สารฟีนอลิกที่มีฤทธิ์ทางชีวภาพจากหาดหนุนและจั่น

นาย บุญชู ศรีตุลารักษ์

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรดุษฎีบัณฑิต
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BIOACTIVE PHENOLICS FROM *ARTOCARPUS GOMEZIANUS* AND *MILLETTIA ERYTHROCALYX*

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บุญชู ศรีตุลารักษ์: สารฟีนอลิกที่มีฤทธิ์ทางชีวภาพจากหาดหนุนและจั่น (BIOACTIVE PHENOLICS FROM *ARTOCARPUS GOMEZIANUS* AND *MILLETTIA ERYTHROCALYX*) อาจารย์ที่ปรึกษา: รศ. ดร. กิตติศักดิ์ ลิขิตวิทยาวุฒิ, 411 หน้า. ISBN 974-17-1374-6

การศึกษาทางพฤกษเคมีของรากหาดหนุน สามารถแยกสารกลุ่ม dimeric stilbene ได้ 2 ชนิด พบว่าเป็นสารชนิดใหม่ 1 ชนิดคือ artogomezianol และอีก 1 ชนิดเป็นสารที่เคยมีรายงานมาแล้วคือ andalasin A การศึกษาทางพฤกษเคมีของเปลือกต้นจั่น สามารถแยกสารใหม่ 3 ชนิด คือ millettocalyxins A-C และสารที่พบครั้งแรกในธรรมชาติ 2 ชนิด คือ pongol methyl ether และ 2'-hydroxy-3,4-methylenedioxy-4'-γ,γ-dimethylallyloxychalcone นอกจากนี้ยังพบสารที่มีรายงานมาแล้วได้อีก 14 ชนิด ได้แก่ derricidin, 7-γ,γ-dimethylallyloxyflavanone, ponganone I, karanjin, milletenone, ovalifolin, milletenin C, 3',4'-methylenedioxy-7-methoxyflavone, pongaglabrone, prunetin, vicenin II, isovitexin, lupeol และ dihydrophaseic acid-4'-*O*-β-D-glucopyranoside ส่วนการศึกษาทางพฤกษเคมีของรากจั่น พบสารใหม่ 2 ชนิด คือ 6-methoxy-[2",3":7,8]-furanoflavanone และ 2,5-dimethoxy-4-hydroxy-[2",3":7,8]-furanoflavan และสารที่พบครั้งแรกในธรรมชาติ คือ 3,4-methylenedioxy-2',4'-dimethoxychalcone รวมทั้งสารที่มีรายงานมาแล้วอีก 10 ชนิด ได้แก่ 1-(4-hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one, derricidin, purpurenone, pongaglabol, ponganone I, pongamol, ovalitenone, milletenone, ponganone V และ lanceolatin B การพิสูจน์โครงสร้างทางเคมีของสารที่แยกได้นี้ อาศัยการวิเคราะห์สเปคตรัมของ UV, IR, MS และ NMR ร่วมกับการเปรียบเทียบข้อมูลของสารที่ทราบโครงสร้างแล้ว ได้ทำการทดลองฤทธิ์การยับยั้งเอนไซม์ tyrosinase, ฤทธิ์จับอนุมูลอิสระและฤทธิ์ต้านไวรัสเริมของสารแต่ละชนิดพบว่า artogomezianol และ andalasin A จากหาดหนุนมีฤทธิ์ปานกลางในการยับยั้งเอนไซม์ tyrosinase และจับอนุมูลอิสระ ยังพบว่า andalasin A มีฤทธิ์แรงในการต้านเชื้อไวรัสเริม HSV-2 แต่มีฤทธิ์ปานกลางต่อเชื้อไวรัสเริม HSV-1 ส่วนสารที่ได้จากจั่นพบว่าทุกชนิดไม่มีฤทธิ์ยับยั้งเอนไซม์ tyrosinase โดยสารเกือบทั้งหมดมีฤทธิ์ต่ำในการจับอนุมูลอิสระและต้านเชื้อไวรัสเริม ยกเว้นสารกลุ่ม flavonoid 4 ชนิดได้แก่ ovalifolin, pongol methyl ether, millettocalyxin A และprunetin ที่พบว่ามีฤทธิ์ปานกลางในการต้านเชื้อไวรัสเริมทั้งสองชนิด

สาขาวิชา เภสัชเคมีและผลิตภัณฑ์ธรรมชาติ ลายมือชื่อนิสิต...
ปีการศึกษา 2545 ลายมือชื่ออาจารย์ที่ปรึกษา..............................

4276955233 MAJOR: PHARMACEUTICAL CHEMISTRY AND NATURAL PRODUCTS

KEY WORD: *ARTOCARPUS GOMEZIANUS*/ *MILLETTIA ERYTHROCALYX*/ TYROSINASE INHIBITORS/ ANTI-HERPES SIMPLEX VIRUS ACTIVITY/ FREE RADICAL SCAVENGING ACTIVITY/ DIMERIC STILBENES/ FLAVONOIDS

BOONCHOO SRITULARAK: BIOACTIVE PHENOLICS FROM *ARTOCARPUS GOMEZIANUS* AND *MILLETTIA ERYTHROCALYX*. THESIS ADVISOR: ASSOCIATE PROFESSOR KITTISAK LIKHITWITAYAWUID, Ph.D, 411 pp. ISBN 974-17-1374-6

Phytochemical study of the roots of *Artocarpus gomezianus* Wall. Ex Tréc. led to the isolation of a new dimeric stilbene, namely, artogomezianol, together with the known stilbene dimer andalasin A. From the stem bark of *Millettia erythrocalyx* Gagnep., 3 new compounds, namely, millettocalyxins A-C, and two new natural products pongol methyl ether and 2'-hydroxy-3,4-methylenedioxy-4'-γ,γ-dimethylallyloxychalcone were isolated, along with 14 other known compounds. The known compounds are derricidin, 7-(γ, γ-dimethylallyloxy)flavanone, ponganone I, karanjin, milletenone, ovalifolin, milletenin C, 3',4'-methylenedioxy-7-methoxyflavone, pongaglabrone, prunetin, vicenin II, isovitexin, lupeol, and dihydrophaseic acid-4'-*O*-β-D-glucopyranoside. The roots of *M. erythrocalyx* Gagnep. yielded 2 new compounds, 6-methoxy-[2'',3'':7,8]-furanoflavanone and 2,5-dimethoxy-4-hydroxy-[2'',3'':7,8]-furanoflavan, and the new natural product 3,4-methylenedioxy-2',4'-dimethoxychalcone, together with 10 other known flavonoids, i.e. 1-(4-hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one, derricidin, purpurenone, pongaglabol, ponganone I, pongamol, ovalitenone, milletenone, ponganone V and lanceolatin B. The structures of all of these isolates were determined by extensive spectroscopic studies, including comparison of their UV, IR, MS and NMR properties with previously reported data. Each of these compounds was evaluated for its tyrosinase inhibitory activity, free radical scavenging activity and anti-herpes simplex virus (HSV-1 and HSV-2) effect. It was found that the stilbene dimers artogomezianol and andalasin A from *A. gomezianus* were moderate tyrosinase inhibitors and moderate free radical scavengers. In addition, andalasin A showed strong activity against HSV-2 but moderate activity against HSV-1. All of the compounds from *M. erythrocalyx* were devoid of tyrosinase inhibitory activity. Most of them showed weak free radical scavenging activity and weak activity against herpes simplex viruses except for 4 flavonoids, including ovalifolin, pongol methyl ether, millettocalyxin A and prunetin, which showed moderate activity against both types of herpes simplex virus.

Field of study Pharmaceutical Chemistry and Natural Products

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Advisor’s signature………………………………….

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LIST OF ABBREVIATIONS AND SYMBOLS

$[\alpha]^{28}_{D}$ = Specific rotation at 28° and sodium D line (589 nm)
α = Alpha
Acetone-d_6 = Deuterated acetone
ax = Axial
β = Beta
br = Broad (for NMR spectra)
C = Concentration
°C = Degree Celsius
calcd = Calculated
CA = Chemical Abstract
CD_{50} = 50% Cytotoxic Dose
CD = Circular Dichroism
$CDCl_3$ = Deuterated chloroform
C_5D_5N = Deuterated pyridine
CD_3OD = Deuterated methanol
$CHCl_3$ = Chloroform
CH_3CN = Acetonitrile
cm = Centimeter
^{13}C NMR = Carbon-13 Nuclear Magnetic Resonance
CO_2 = Carbon dioxide
^{1}H-^{1}H COSY = Homonuclear (Proton-Proton) Correlation Spectroscopy
1-D = One Dimensional
2-D = Two Dimentional
d = Doublet (for NMR spectra)
dd = Doublet of doublets (for NMR spectra)
DEPT = Distortionless Enhancement by Polarization Transfer
DMSO-d_6 = Deuterated dimethylsulfoxide
DPPH = 1,1-Diphenyl-2-picrylhydrazyl
δ = Chemical shift
ED_{50} = 50% Effective Dose

LIST OF ABBREVIATIONS AND SYMBOLS (continued)

EIMS = Electron Impact Mass Spectrometry

eq = Equatorial

ESIMS = Electrospray Ionization Mass Spectrometry

EtOAc = Ethyl acetate

g = Gram

hr = Hour

^{1}H NMR = Proton Nuclear Magnetic Resonance

HMBC = ^{1}H-detected Heteronuclear Multiple Bond Coherence

HMQC = ^{1}H-detected Heteronuclear Multiple Quantum Coherence

H_2O = Water

HPLC = High Performance Liquid Chromatography

HRFABMS = High Resolution Fast Atom Bombardment Mass Spectrum

HSQC = Heteronuclear Single Quantum Correlation

HSV-1 = Herpes Simplex Virus type 1

HSV-2 = Herpes Simplex Virus type 2

Hz = Hertz

IC_{50} = Median Inhibitory Concentration

IR = Infrared Spectrum

J = Coupling constant

KBr = Potassium bromide

Kg = Kilogram

L = Liter

L-DOPA = L-3,4-Dihdroxyphenyl alanine

μg = Microgram

μL = Microliter

μM = Micromolar

λ_{max} = Wavelength at maximal absorption

ε = Molar absorptivity

M^+ = Molecular ion

m = Meta

LIST OF ABBREVIATIONS AND SYMBOLS (continued)

m = Multiplet (for NMR spectra)
MeOH = Methanol
mg = Milligram
$[M+H]^+$ = Protonated molecular ion
MHz = Megahertz
min = Minute
mL = Millimeter
mM = Millimolar
MPLC = Medium Pressure Liquid Chromatography
MW = Molecular weight
m/z = Mass to charge ratio
MS = Mass Spectrometry
mult. = Multiplicity
NaH_2PO_4 = Sodium dihydrogen phosphate
Na_2HPO_4 = Disodium hydrogen phosphate
nm = Nanometer
NMR = Nuclear Magnetic Resonance
NOE = Nuclear Overhauser Effect
NOESY = Nuclear Overhauser Effect Spectroscopy
o = Ortho
p = Para
Pet. ether = Petroleum ether
PE = Petroleum ether
PLC = Preparative Thin Layer Chromatography
ppm = Part per million
pyridine-d_5 = Deuterated pyridine
q = Quartet (for NMR spectra)
RDA = *retro*-Diels-Alder
ROESY = Rotating-Frame Overhauser Enhancement Spectroscopy
spp. = Species

LIST OF ABBREVIATIONS AND SYMBOLS (continued)

TFA = Trifluoro acetic acid

TOCSY = Total Correlation Spectroscopy

ν_{max} = Wave number at maximal absorption

s = Singlet (for NMR spectra)

t = Triplet (for NMR spectra)

TLC = Thin Layer Chromatography

UV = Ultraviolet

UV-VIS = Ultraviolet and Visible Spectrophotometry

VLC = Vacuum Liquid Column Chromatography

CHAPTER I

INTRODUCTION

The genus *Artocarpus* belongs to the family Moraceae of the order Urticarles. This genus consists of about 47 species distributed in Ceylon, India, Pakistan, Burma, Siam, Indo-China, South-China, Malesia and Solomon Islands. Three species (*A. communis*, *A. heterophyllus* and *A. integer*) are cultivated throughout the tropics; 20 spp. in Malaya including the cultivated species (Kochummen, 1978).

Plants in the genus *Artocarpus* are evergreen trees with milky juice. Leaves alternate, coriaceous, often very large, entire, lobe or pinnatifid, penninerved. Flowers monoecious, densely crowded on globose or oblong, 1-sexual solitary usually axillary receptacles, often mixed with scales which are often thickened or peltate at the apex. Male flowers: Perianth 2-4-lobed or -partite; lobes obtus, valvate or slightly imblicate. Stamens 1, erect. Pistillode 0. Female flowers: Perianths tubular, confluent below with the receptacle; mouth minute. Ovary straight; ovule pendulous; style central or lateral; stigma entire (rarely 2-3 fid). Fruit much enlarged fleshy oblong cylindric or subglobose entire or lobed receptacle clothed with the greatly accrescent fleshy perianths and carpels (anthocarps) which have hardened spinescent or truncate or pyramidal or flat apices. Seed pendulous; testa membranous; albumen 0; embryo straight or incurved; cotyledons fleshly equal or unequal; radicle short, superior (Kirtikar and Basu, 1980).

According to Smitinand (2001), the species of genus *Artocarpus* found in Thailand are as follows.

Artocarpus altilis (Parkinson) Fosberg (*A. communis* J.R. & G. Forst., *A. incisa* Linn. f.)	ขนุนสำปะลอ Khanun sampalo (Central); สาเก Sake (Central); Bread fruit tree; Bread nut tree.
A. altissimus J.J. Smith	ไสน Sanai (Surat Thani).
A. chaplasha Roxb.	หาดส้าน Haat san (Chiang Rai).
A. dadah Miq.	ทังคัน Thang khan; ม่วงกวาง Muang kwang, (Yala); หาดรุม Hat rum, หาดลูกใหญ่, Hat luk yai (Trang); หาดขน Hat khon (Narathiwat).

A. elasticus Reinw. ex Blume	กะออก Ka ok, กะเอาะ Ka-o (Peninsular); ตือกะ Tue-ka (Malay-Yala); เอาะ O (Trang,Ranong).
A. gomezianus Wall. ex Trécul	ตะปัง Ta pang, ตำปัง Tam-pang (Malay-Peninsular); หาดหนุน Hat nun (Northern); อี โป้ I po (Trang).
A. heterophyllus Lamk. (*A. integrifolious* Linn. f.)	ขนุน Khanun (General); ขะนู Kha-nu (Chong-Chanthaburi); ขะเนอ Kha-noe (Khmer); ซีคึย Si-khue, ปะหน่อย Pa-noi (Karen-Mae Hong Son); นะยวยซะ Na-yuai-sa (Karen-Kanchanaburi); นากอ Na-ko (Malay-Pattani); เนน Nen (Chaobon-Nakhon Ratchasima); มะหนุน Manun (Northern, Peninsular); ล้าง, ลาง Lang (Shan-Northern); หมักหมี้ Mak mi (Northeastern); หมากลาง Mak lang (Shan-Mae Hong Son); Jack fruit tree.
A. kemando Miq.	ขนุนป่า Khunun pa (Narathiwat); ยาตู Yatu (Malay-Narathiwat).
A. integer (Thunb.) Merr.	จำปาดะ Champada (General); จำปาเดาะ Champado (Peninsular); Champedak.
A. lacucha Roxb. (*A. lakoocha* Roxb.)	กาแย Kaa-yae, ตาแป Ta-pae, ตาแปง Ta-paeng (Malay-Narathiwat); มะหาด Mahat (Peninsular); มะหาดใบใหญ่ Mahat bai yai (Trang); หาด Hat (General).
A. lanceifolius Roxb.	ขนุนป่า Khanun pa (Peninsular); หนังกาปีโต Nang-ka pi-to, หนังกาปีปิ๊ต Nang-ka-pi-pit (Malay-Peninsular); นั่งกาปีแป๊ะ Nang-ka-pi-pae (Malay-Narathiwat).
A. nitidus Tréc subsp. *lingnanensis* Jarrett (*A. parva* Gagnep.)	มะหาดข่อย Mahat khoi (Surat Thani).
A. rigidus Blume subsp. *rigidus*	ขนุนป่า Khanun pa (Peninsular).
A. rigidus bl. subsp. *asperulus* Jarrett. (*A. calophyllus* Kurz)	ขนุนปาน Khanun pan (Surat Thani).

Artocarpus gomezianus Wall. ex Tréc. is an indigenous plant known in Thai as Hat-nun. It is a medium-sized to tall tree reaching 42 m and 210 cm gird. Bark: gray brown, cracking to scaly. Inner bark: pink, soft with creamy sap. Sapwood: pale yellow. Leaves: stalk 1.5-3 cm long; blade leathery, oblong to elliptic, 11-25 x 7-16 cm, apex shortly pointed, base more or less rounded, glabrous on both surfaces, upper surface shining, secondary nerves 10-15 pairs, nervation prominent on both surfaces; midrib and nerves drying black. Flower heads: solitary in leaf axils; male head: obovoid to subglobose, 1-2.5 cm across on 0.7-1.7 cm long stalk. Fruit: subglobose, 8 cm across, yellow pink flesh, drying brown or black, with smooth velutinous surface, stalk 1.5-4.5 cm long. Seeds: ellipsoid, 1.2 x 1 cm (Kochummen, 1978).

The genus *Millettia* (Leguminosae) consists of 134 species distributed in Africa, Indo-Malaya, China and Australia. They are climbing shrubs or trees. Leaves imparipinnate 2 (rarely 1) to many leaflets. Panicles large or reduced to racemes of fascicles. Calyx campanulate; teeth short. Petals white or pink; standard ovate or orbicular; wing oblong. Stamen monodelphous (rarely diadelphous); filaments free at tip. Ovary sessile, rarely stipitate, 3- to many-ovuled. Pod linear or oblong, coriaceous or woody, flattened or thick. Seeds lenticular or reniform (Ridley, 1967).

The species of *Millettia* which have been recorded in Thailand (Smitinand, 2001), are as follows:

Millettia atropurpurea Wall.	=	*Collerya atropurpurea* (Wall.) Schott
M. brandisiana Kurz		กระพี้จั่น Kra phi chan, จั่น Chan, พี้จั่น Phi chan, (General); ปี๊จั่น Pi chan (Northern).
M. caerulea Baker		ปัวเปาะเด๊า Pua-po-do (Karen-Mae Hong Son); ผักเยี่ยววัว Phak yiao wua (Nakhon Sawan, Northern); หางไหลแดง Hang lai daeng (Kanchanaburi).
M. decipiens Prain		ปารี Pa ri (Malay-Narathiwat).
M. extensa Benth. (*M. auriculata* Bak. var. extensa)		ก๋าวเครือ Kao khruea, กวาวเครือ Kwao khruea (Chiang Mai); ตานครบ Tan krop (Lampang).
M. glaucescens Kurz		ยาดา Ya da (Malay-Narathiwat); หยีน้ำ Yi nam (Peninsular).
M. kangensis Craib		กระเจาะ Kra cho, ขะเจาะ Kha cho, ขะเจาะน้ำ Kha cho nam (Chiang Mai).
M. kityana Craib.		เครือข้าวเย็น Khruea khao yen, ลางเย็น Lang yen, ฮางจืด Hang chuet, ฮางเย็น Hang yen (Northern).

M. latifolia Dunn		ขะเจาะ Kha cho (General).
M. leucantha Kurz var. *leucantha*		กะเซาะ Kaso (Central); กระเจาะ Kra cho, ขะเจาะ Khra cho (Northern); กระพี้เขาควาย Kra phi khao khwai (Prachuap Khiri Khan); ขะแมบ Kha maep, คำแมบ Kham maep (Chiang Mai).
M. leucantha Kurz var. *buteoides* (Gagnep.) P.K.Loc (*M. buteoides* Gagnep. var. *siamensis* Craib, *M. pendula* Benth.)		กระเจ๊าะ Kra cho, ขะเจ๊า Kha cho (Lampang); กระท้อน Kra thon (Phetchabun, Phitsanulok); ไม้กระทงน้ำผัก Mai kra thong nam phak (Loei); สะท้อน Sa thon (Saraburi); สาธร Sa thon (Ubon Ratchathani).
M. macrostachya Collett & Hemsl. var. *macrostachya*		ขะเจาะน้ำ Kha cho nam (Chiang Mai).
M. macrostachya Collett & Hemsl. var. *tecta* Craib		ขะเจาะหลวง Kha cho luang, ขะเจาะใหญ่ Kha cho yai (Narathiwat).
M. pachycarpa Benth.		เกถะ Ke-tha (Karen-Chiang Mai); เครือไหล Khruea lai (Chiang Mai).
M. peguensis Ali (*M. ovalifolia* Kurz)		ตอหิ To-hi (Karen-Kanchanaburi).
M. pulchra Benth. Kurz		จันพอ Chan pho (Northern).
M. recemosa (Roxb.) Benth.	=	*Endosamara racemosa* (Roxb.) R. Geesink
M. sericea (Vent.) Benth.		จะไนโค๊ะ Cha-nai-kho, ปาตู Pa-tu (Malay-Narathiwat); นอเราะ No-ro (Malay-Yala, Pattani); ยิมแมเก๊าะ Yim-mae-ko (Malay-Yala); อ้อยสามสวน Oi sam suan (Nong Khai).
M. thorelii Gagnep.	=	*Derris thorelii* Craib
M. utilis Dunn		สะท้อนน้ำผัก Sathon nam phak (Loei).
M. xylocarpa Miq. (*M. hemsleyana* Prain, *M. pubinervis* Kurz)		กะเจ๊าะ Ka che, ขะเจาะ Kha cho (General); คะแมด Kha maet (Chiang Mai); จักจั่น Chakkachan (Loei); พี้พง Phi phong (Phrae); ยะดา Ya-da (Malay-Yala); ไย่ยี Yai-yi (Karen-Mae Hong Son); สาธร Sa thon, หยีน้ำ Yi nam (Pattani, Yala).

Although *Millettia erythrocalyx* Gagnep. has not been recorded in Thai Plant Names (Smitinand, 2001), but the herbarium specimens of this species have been kept at the Royal Forest Department, Ministry of Agriculture and Co-operatives.

Millettia erythrocalyx Gagnep. has a local name as Jun. It is a medium-sized tree reaching 7-8 m. Bark: grayish. Branchlets: rusty strigose, become glabrescent, spotted lenticels prominent. Leaflets: 7-11, ovate- or elliptic-lanceolate, papery, 3-6 x 1.5-2 cm, base narrowly cordate, apex caudate, glabrous and shining adaxially, scarlet strigose on midrib and margins abaxially. Pseudoracemes: axillary on the tip of branches, 6-7 cm, densely dark brown strigose; flower 8-9 mm, calyx 3 mm, deep red, sparsely hairly, teeth truncate, ciliated; corolla lilac, vexillum glabrous, round tapering at base, with 2 minute callus; ovary villose, ovules 4-5. Pod: linear-oblong, 9-10 x 2 cm, flat, slightly curved, tapering to the base, brown tomentose when young, become glabrescent, valves woody, spirally twisted. Seed: 2-3, chestnut brown, 13 x 10 mm, lens-shaped, smooth. It has been found in Thailand, Laos and Cambodia (Gagnepain, 1916; Zhi, 2002).

The isolation of several phenolic compounds from a petroleum ether and a MeOH extracts of the roots of *Artocarpus gomezianus* Wall. ex Tréc. has been earlier described (Sritularak, 1998). These compounds are isocyclomorusin [**25**], cycloartocarpin [**30**], artocarpin [**4**], norartocarpetin [**45**], cudraflavone C [**69**], albanin A [**68**], resveratrol [**177**], resorcinol [**175**]. In the present study, attention has been paid on more polar constituents in the MeOH extract. As for *Millettia erythrocalyx*, no phytochemical work has been reported.

During our preliminary evaluation for biological activities, the extract of *A. gomezianus* showed significant tyrosinase inhibitory activity whereas that of *M. erythrocalyx* was devoid of activity. In addition, both plant extracts exhibited anti-herpes simplex and antioxidant activities (see Results and Discussion section). Therefore, the following objectives are put forwards:

1. Isolation and purification of compounds from the roots of *Artocapus gomezianus*, and from the stem bark and the roots of *Millettia erythrocalyx*.
2. Determination of the chemical structure of each isolated compound.
3. Evaluation of each isolate for its tyrosinase inhibition potential, free radical scavenging activity and anti-herpes simplex effect.

Figure 1 *Artocarpus gomezianus* Wall. ex Tréc.

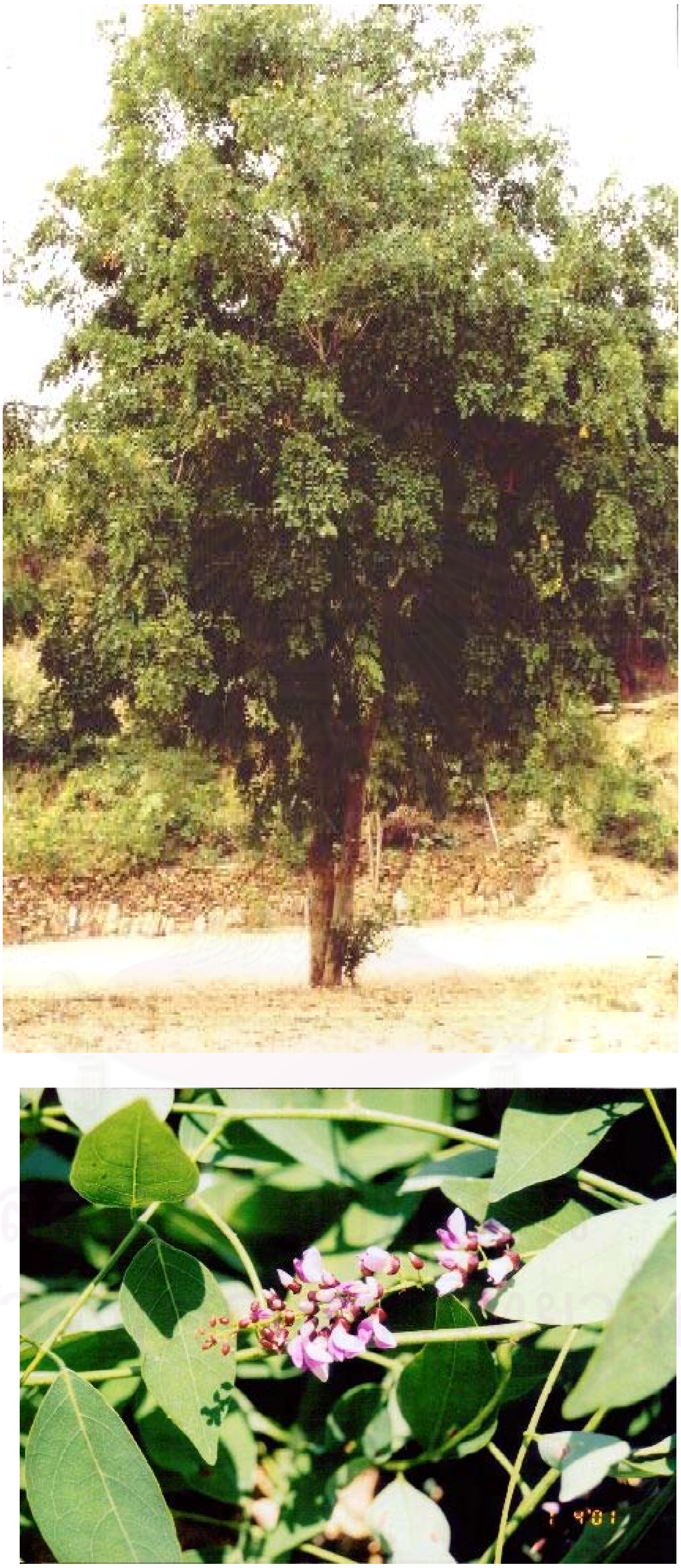


Figure 2 *Millettia erythrocalyx* Gagnep.

CHAPTER II

HISTORICAL

1. Chemical Constituents of *Artocarpus* spp.

A number of compounds have been isolated from the genus *Artocarpus*. They can be classified as flavonoids, triterpenoids, steroids, stilbenes and miscellaneous substances (Tables 1-3).

Table 1 Distribution of flavonoids in *Artocarpus*.

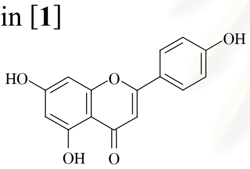
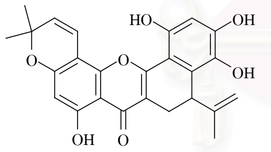
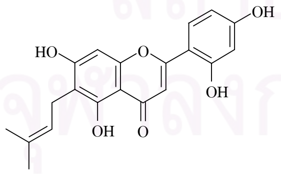
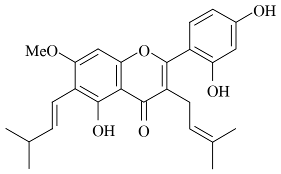
Plant and chemical compound	Plant part	Reference
Artocarpus altilis		
Apigenin [**1**]  	Heartwood	Shimizu *et al*., 1998
Artobiloxanthone [**2**]  	Stem bark	Aida *et al*., 1997
Artocarpesin [**3**]  	Heartwood	Shimizu *et al*., 1998
Artocarpin[**4**]  	Heartwood	Venkataraman, 1972

Table 1 (continued)

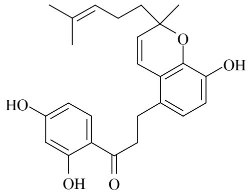
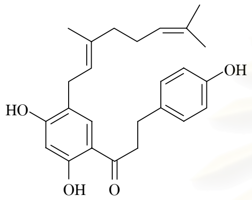
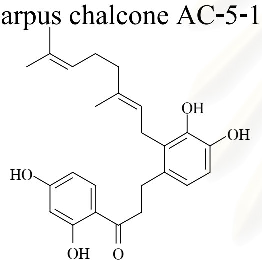
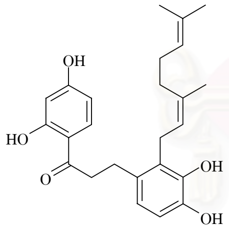
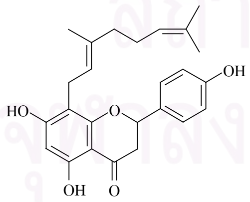
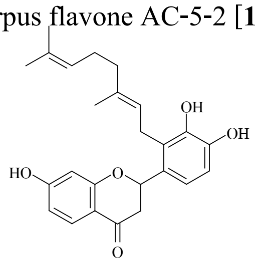
Plant and chemical compound	Plant part	Reference
Artocarpus chalcone AC-3-1 [**5**] 	Flower	Fujimoto *et al.*, 1987
Artocarpus chalcone AC-3-2 [**6**] 	Flower	Fujimoto *et al.*, 1987
Artocarpus chalcone AC-5-1 [**7**] 	Flower	Fujimoto *et al.*, 1987
Artocarpus chalcone I [**8**] 	Flower	Fujimoto, Agusutein, and Made, 1987
Artocarpus flavone AC-3-3 [**9**] 	Flower	Fujimoto *et al.*, 1987
Artocarpus flavone AC-5-2 [**10**] 	Flower	Fujimoto *et al.*, 1987

Table 1 (continued)

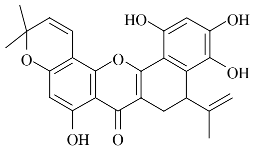
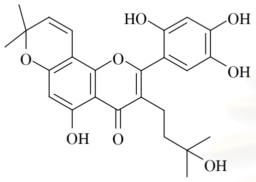
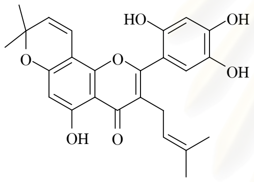
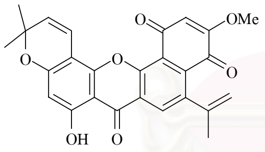
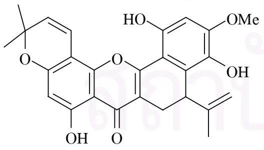
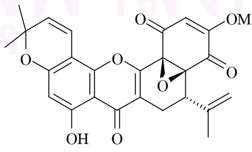
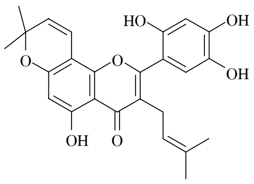
Plant and chemical compound	Plant part	Reference
Artocarpus flavone KB-1 [**11**] 	Stem bark	Fujimoto *et al*., 1990
Artocarpus flavone KB-2 [**12**] 	Stem bark	Fujimoto *et al*., 1990
Artocarpus flavone KB-3 [**13**] (Artonin E) 	Stem bark	Fujimoto *et al*., 1990
Artomunoxanthentrione [**14**] 	Root bark	Shieh and Lin, 1992
Artomunoxanthone [**15**] 	Root bark	Shieh and Lin, 1992
Artomunoxanthotrione epoxide [**16**] 	Root bark	Lin, Shieh, and Jong, 1992
Artonin E [**13**] 	Stem bark	Hano *et al*., 1990

Table 1 (continued)

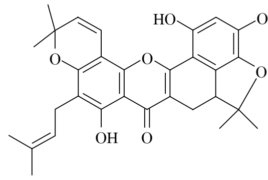
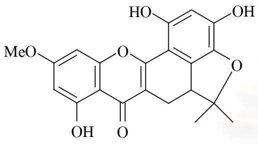
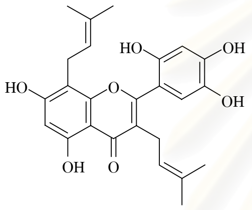
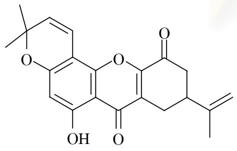
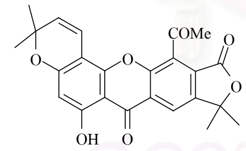
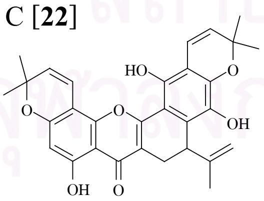
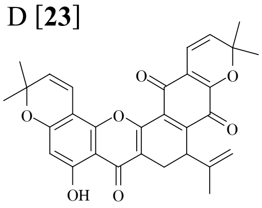
Plant and chemical compound	Plant part	Reference
Artonin F [**17**] 	Stem bark	Hano *et al*., 1990
Artonin K [**18**] 	Stem bark	Aida *et al*., 1997
Artonin V [**19**] 	Root bark	Hano, Inami, and Nomura, 1994
Artonol A [**20**] 	Stem bark	Aida *et al*., 1997
Artonol B [**21**] 	Stem bark	Aida *et al*., 1997
Artonol C [**22**] 	Stem bark	Aida *et al*., 1997
Artonol D [**23**] 	Stem bark	Aida *et al*., 1997

Table 1 (continued)

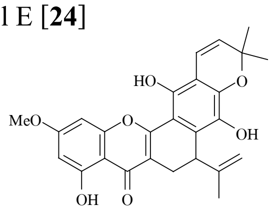
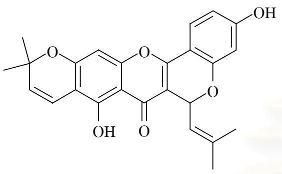
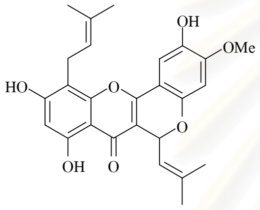
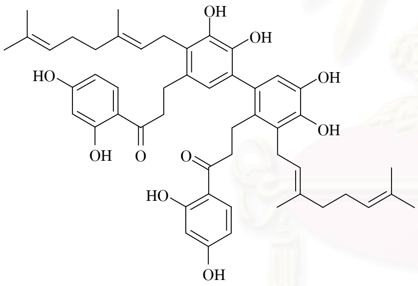
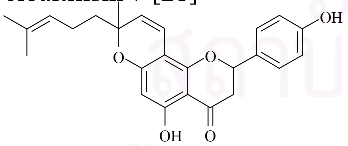
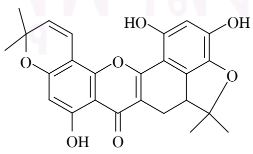
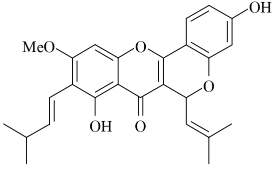
Plant and chemical compound	Plant part	Reference
Artonol E [**24**] 	Stem bark	Aida *et al*., 1997
Cudraflavone A [**25**] 	Root bark	Shieh and Lin, 1992
Cycloaltilisin [**26**] 	Stem	Chen *et al*., 1993
Cycloaltilisin 6 [**27**] 	Bud cover	Patil *et al*., 2002
Cycloaltilisin 7 [**28**] 	Bud cover	Patil *et al*., 2002
Cycloartobiloxanthone [**29**] 	Stem bark	Hano *et al*., 1990
Cycloartocarpin [**30**] 	Heartwood	Venkataraman, 1972

Table 1 (continued)

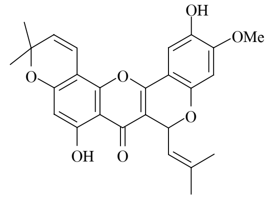
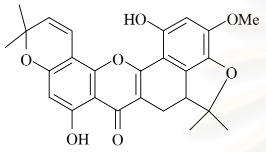
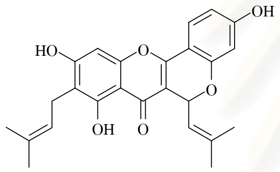
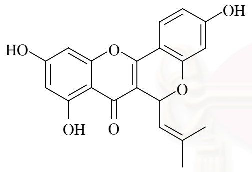
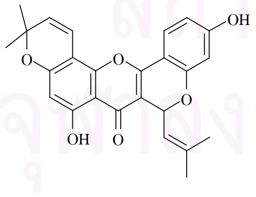
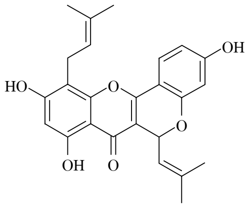
Plant and chemical compound	Plant part	Reference
Cycloartomunin [**31**] 	Root bark	Lin and Shieh, 1991
Cycloartomunoxanthone [**32**] 	Root bark	Lin and Shieh, 1991
Cyclocommunin [**33**] 	Root bark	Lin and Shieh, 1991
Cyclocommunol [**34**] 	Root bark	Lin and Shieh, 1991
Cyclomorusin [**35**] 	Root bark Stem	Lin and Shieh, 1991; Chen *et al*., 1993
Cyclomulberrin [**36**] 	Root bark Stem	Lin and Shieh, 1992; Chen *et al*., 1993

Table 1 (continued)

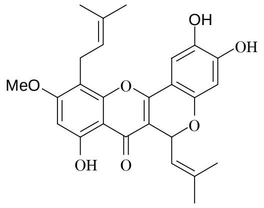
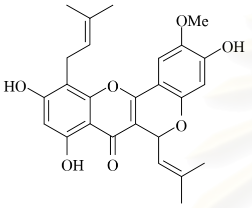
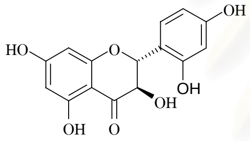
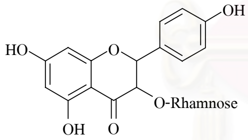
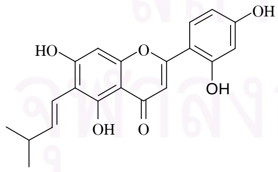
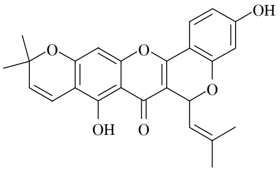
Plant and chemical compound	Plant part	Reference
Dihydrocycloartomunin [**37**] 	Root bark	Lin and Shieh, 1991
Dihydroisocycloartomunin [**38**] 	Root bark	Lin and Shieh, 1992
Dihydromorin [**39**] 	Heartwood	Shimizu *et al*., 1998
Engeletin [**40**] 	Stem	Chen *et al*., 1993
Isoartocarpesin [**41**] 	Heartwood	Shimizu *et al*., 1998
Isocyclomorusin (Cudraflavone A) [**25**] 	Stem	Chen *et al*., 1993

Table 1 (continued)

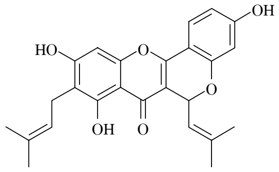
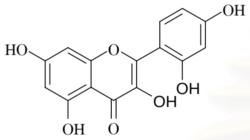
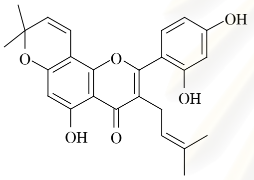
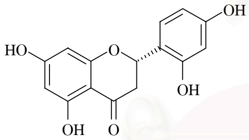
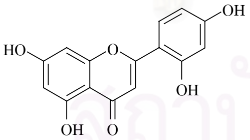
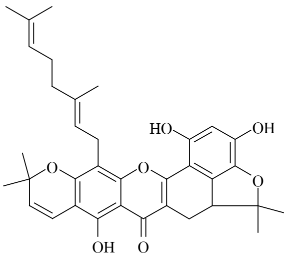
Plant and chemical compound	Plant part	Reference
Isocyclomulberrin (Cyclocommunin) [**33**]  	Stem	Chen *et al*., 1993
Morin [**42**]  	Heartwood	Venkataraman, 1972
Morusin [**43**]  	Stem bark	Fujimoto *et al*., 1990
(+)-Norartocarpone [**44**]  	Heartwood	Shimizu *et al*., 1998
Norartocarpetin [**45**]  	Heartwood	Venkataraman, 1972
A. champeden		
Artoindonesianin A [**46**]  	Root	Hakim *et al*., 1999

Table 1 (continued)

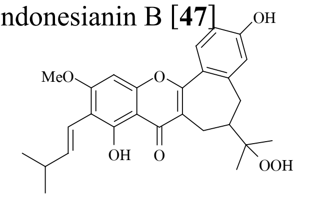
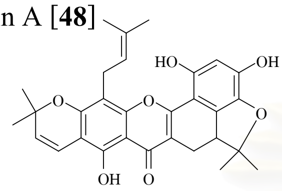
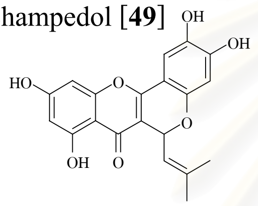
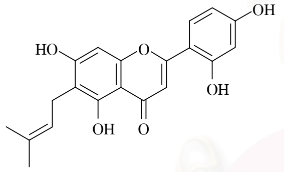
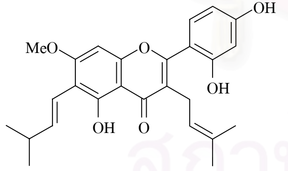
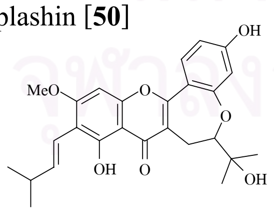
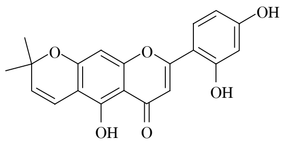
Plant and chemical compound	Plant part	Reference
Artoindonesianin B [**47**] 	Root	Hakim *et al*., 1999
Artonin A [**48**] 	Root	Hakim *et al*., 1999
Cyclochampedol [**49**] 	Stem bark	Achmad *et al*., 1996; Paolo *et al*, 1998
A. chaplacha		
Artocarpesin [**3**] 	Heartwood	Rao, Rathi, and Venkataraman, 1972
Artocarpin [**4**] 	Heartwood	Rao *et al*., 1972
Chaplashin [**50**] 	Heartwood	Rao *et al*., 1972
Cycloartocarpesin [**51**] 	Heartwood	Rao *et al*., 1972

Table 1 (continued)

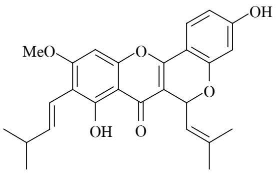
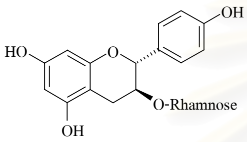
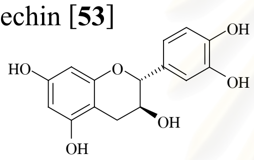
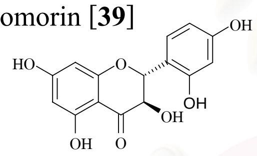
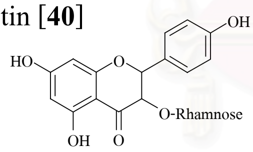
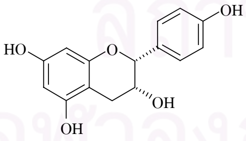
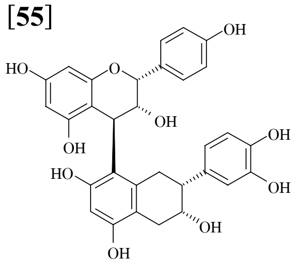
Plant and chemical compound	Plant part	Reference
Cycloartocarpin [**30**]  	Heartwood	Rao *et al.*, 1972
A. dadah		
Afzelechin-3-*O*-α-L-rhamnopyranoside [**52**]  	Stem bark Twig	Su *et al.*, 2002
(+)-Catechin [**53**]  	Stem bark Twig	Su *et al.*, 2002
Dihydromorin [**39**]  	Stem bark	Su *et al.*, 2002
Engeletin [**40**]  	Twig	Su *et al.*, 2002
(-)-Epiafzelechin [**54**]  	Stem bark	Su *et al.*, 2002
(-)-Epiafzelechin-(4β⟶8)-epicatechin [**55**]  	Stem bark	Su *et al.*, 2002

Table 1 (continued)

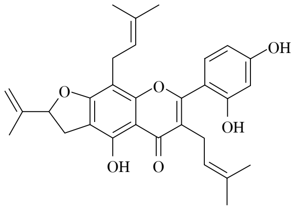
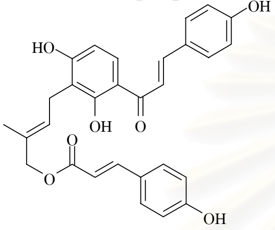
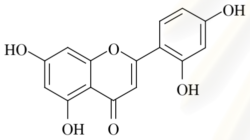
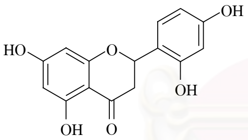
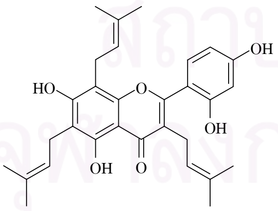
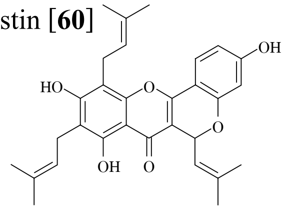
Plant and chemical compound	Plant part	Reference
Gemichalcone B [**56**] 	Twig	Su *et al*., 2002
Isogemichalcone B [**57**] 	Twig	Su *et al*., 2002
Norartocarpetin [**45**] 	Twig	Su *et al*., 2002
Steppogenin [**58**] 	Twig	Su *et al*., 2002
A. elasticus		
Artelasticin [**59**] 	Heartwood	Kijjoa *et al*., 1996
Artelastin [**60**] 	Heartwood	Kijjoa *et al*., 1996

Table 1 (continued)

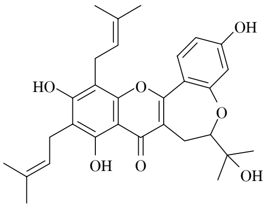
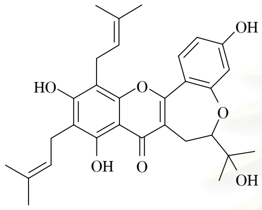
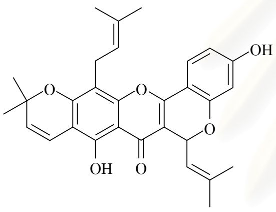
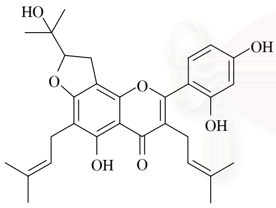
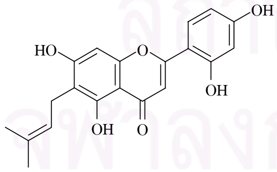
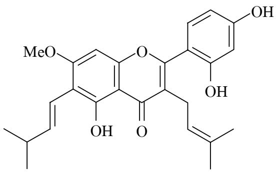
Plant and chemical compound	Plant part	Reference
Artelastinin [**61**] 	Heartwood	Kijjoa *et al.*, 1998
Artelastocarpin [**62**] 	Heartwood	Cidade *et al.*, 2001
Artelastochromene [**63**] 	Heartwood	Kijjoa *et al.*, 1996
Artelastofuran [**64**] 	Heartwood	Kijjoa *et al.*, 1998
Artocarpesin [**3**] 	Heartwood	Kijjoa *et al.*, 1996
Artocarpin [**4**] 	Heartwood	Kijjoa *et al.*, 1976

Table 1 (continued)

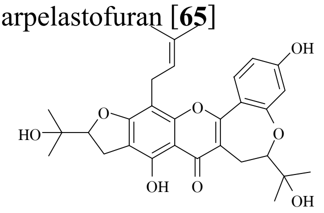
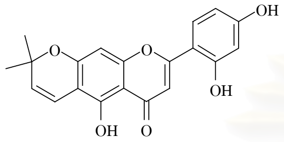
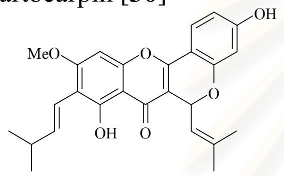
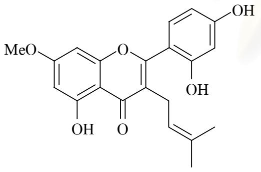
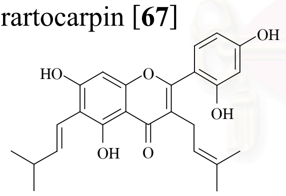
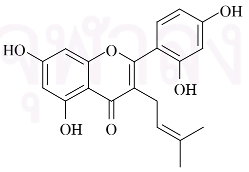
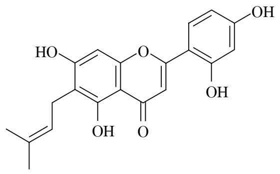
Plant and chemical compound	Plant part	Reference
Carpelastofuran [**65**] 	Heartwood	Cidade *et al*., 2001
Cycloartocarpesin [**51**] 	Heartwood	Pendse *et al*., 1976
Cycloartocarpin [**30**] 	Heartwood	Pendse *et al*., 1976
Integrin [**66**] 	Heartwood	Pendse *et al*., 1976
Norartocarpin [**67**] 	Heartwood	Pendse *et al*., 1976
A. gomezianus		
Albanin A [**68**] 	Root	Sritularak, 1998; Likhitwitayawuid, Sritularak, and De-Ek-Namkul, 2000
Artocarpesin [**3**] 	Heartwood	Venkataraman, 1972

Table 1 (continued)

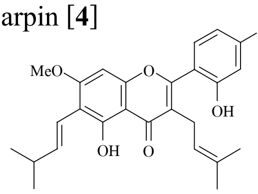
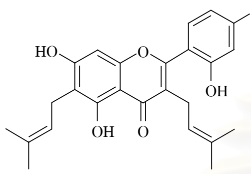
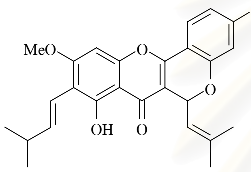
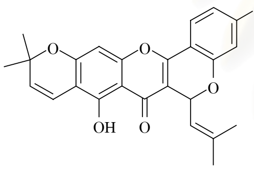
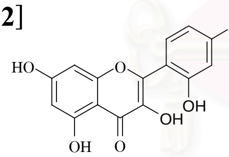
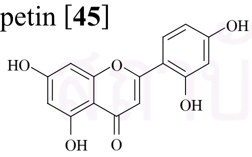
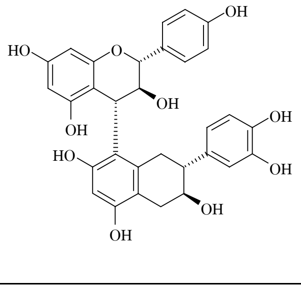
Plant and chemical compound	Plant part	Reference
Artocarpin [**4**]  	Heartwood	Venkataraman, 1972
Cudraflavone C [**69**]  	Root	Sritularak, 1998; Likhitwitayawuid, Sritularak, and De-Ek-Namkul, 2000
Cycloartocarpin [**30**]  	Heartwood	Venkataraman, 1972
Isocyclomorusin [**25**]  	Root	Sritularak, 1998; Likhitwitayawuid, Sritularak, and De-Ek-Namkul, 2000
Morin [**42**]  	Heartwood	Venkataraman, 1972
Norartocarpetin [**45**]  	Heartwood	Venkataraman, 1972
A. heterophyllus		
Afzelechin-(4α→8)-catechin [**70**]  	Leaf	An *et al*., 1992

Table 1 (continued)

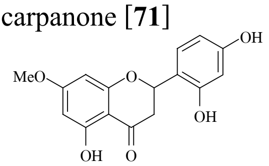
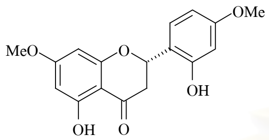
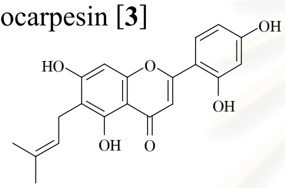
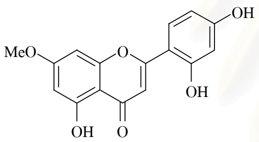
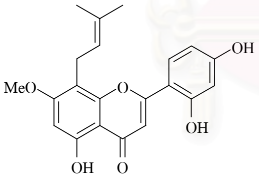
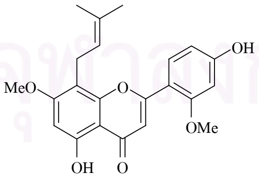
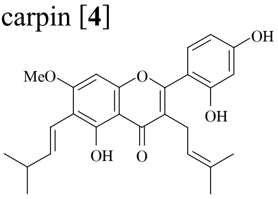
Plant and chemical compound	Plant part	Reference
Artocarpanone [**71**] 	Heartwood	Radhakrishnan, Rao, and Venkataraman, 1965
Artocarpanone A [**72**] 	Root bark	Lin *et al*., 1995
Artocarpesin [**3**] 	Heartwood	Radhakrishnan *et al*., 1965
Artocarpetin [**73**] 	Heartwood	Venkataraman, 1972
Artocarpetin A [**74**] 	Root bark	Lin *et al*., 1995
Artocarpetin B [**75**] 	Root	Chung *et al*., 1995
Artocarpin [**4**] 	Heartwood	Radhakrishnan *et al*., 1965

Table 1 (continued)

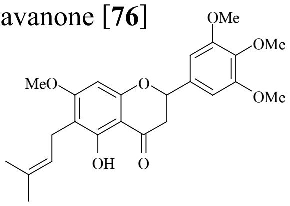
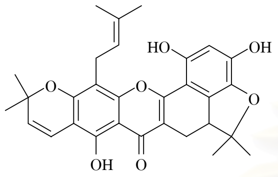
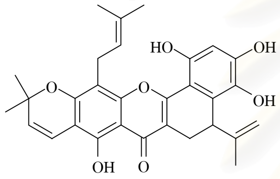
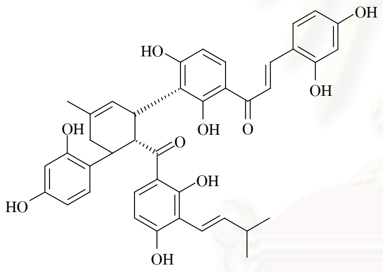
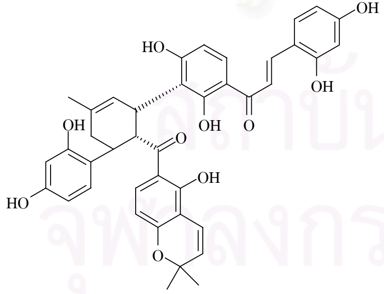
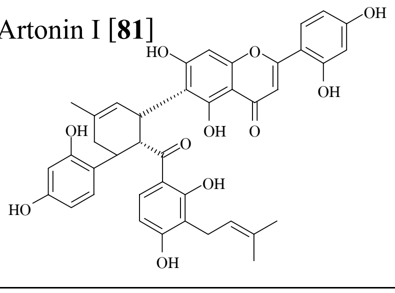
Plant and chemical compound	Plant part	Reference
Artoflavanone [**76**] 	Root	Dayal and Seshadri, 1974
Artonin A [**77**] 	Root bark	Hano *et al*., 1989
Artonin B [**78**] 	Root bark	Hano *et al*., 1989
Artonin C [**79**] 	Root bark	Hano, Aida, and Nomura, 1990
Artonin D [**80**] 	Root bark	Hano, Aida, and Nomura, 1990
Artonin I [**81**] 	Root bark	Hano *et al*., 1989

Table 1 (continued)

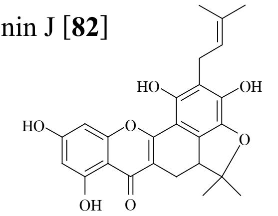
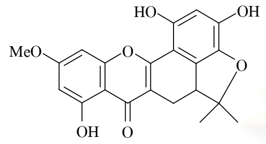
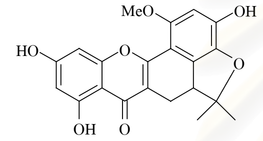
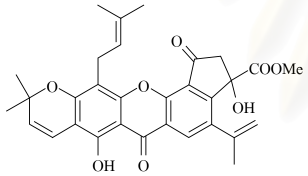
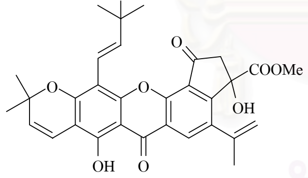
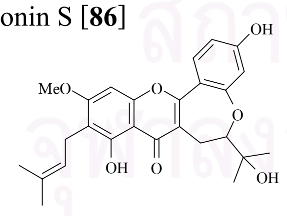
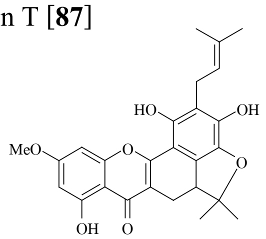
Plant and chemical compound	Plant part	Reference
Artonin J [**82**] 	Root bark	Aida *et al*., 1993
Artonin K [**18**] 	Root bark	Aida *et al*., 1993
Artonin L [**83**] 	Root bark	Aida *et al*., 1993
Artonin Q [**84**] 	Stem bark	Aida *et al*., 1994
Artonin R [**85**] 	Stem bark	Aida *et al*., 1994
Artonin S [**86**] 	Stem bark	Aida *et al*., 1994
Artonin T [**87**] 	Stem bark	Aida *et al*., 1994

Table 1 (continued)

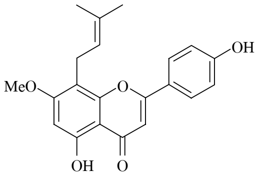
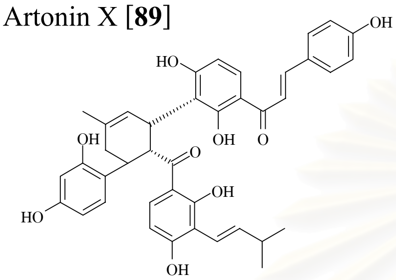
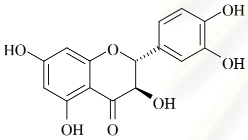
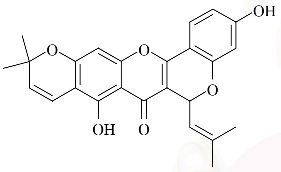
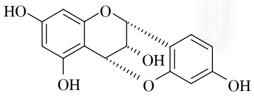
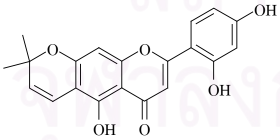
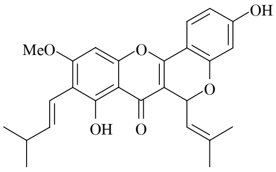
Plant and chemical compound	Plant part	Reference
Artonin U [**88**] 	Stem bark	Aida *et al*., 1994
Artonin X [**89**] 	Stem bark	Shinomiya *et al*., 1995
Catechin [**90**] 	Leaf	Yamazaki *et al*., 1987
Cudraflavone A [**25**] 	Root bark	Lin *et al*., 1995
Cyanomaclurin [**91**] 	Heartwood	Radhakrishnan *et a*l., 1965
Cycloartocarpesin [**51**] 	Heartwood	Parthasarathy *et al*., 1969
Cycloartocarpin [**30**] 	Heartwood	Venkataraman, 1972

Table 1 (continued)

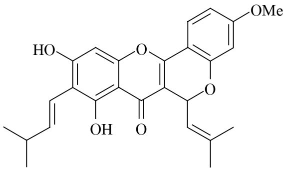
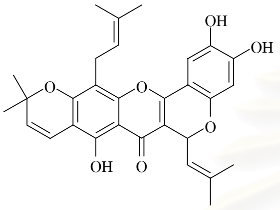
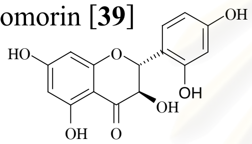
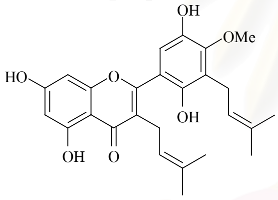
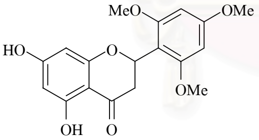
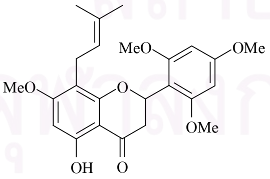
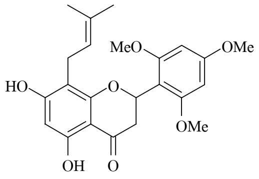
Plant and chemical compound	Plant part	Reference
Cycloartocarpin A [**92**] 	Root bark	Lu and Lin, 1994
Cycloheterophyllin [**93**] 	Stem bark Root bark	Rao, Varadan, and Venkataraman, 1971; Hano *et al*., 1989
Dihydromorin [**39**] 	Heartwood	Venkataraman, 1972
Heteroartonin A [**94**] 	Root	Chung *et al*., 1995
Heteroflavanone A [**95**] 	Root bark	Lu and Lin, 1993
Heteroflavanone B [**96**] 	Root bark	Lu and Lin, 1993
Heteroflavanone C [**97**] 	Root bark	Lu and Lin, 1994

Table 1 (continued)

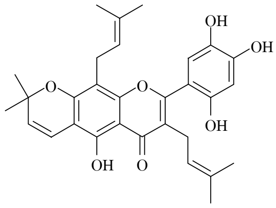
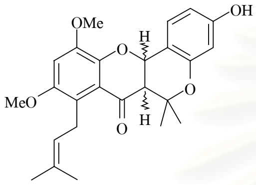
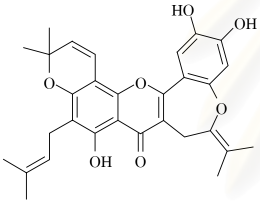
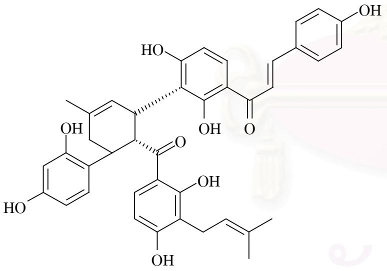
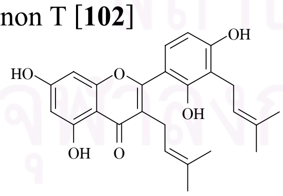
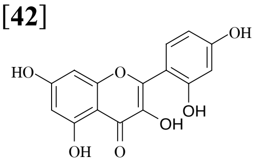
Plant and chemical compound	Plant part	Reference
Heterophyllin [**98**] 	Root bark	Hano *et al*., 1989
Heterophylol [**99**] 	Root bark	Lin and Lu, 1993
Isocycloheterophyllin [**100**] 	Stem bark	Rao, Varadan, and Venkataraman, 1973
Kuwanon R [**101**] 	Root bark	Shinomiya *et al*., 1995
Kuwanon T [**102**] 	Root bark	Shinomiya *et al*., 1995
Morin [**42**] 	Heartwood	Radhakrishnan *et al*., 1965; Parthasarathy *et al*., 1969

Table 1 (continued)

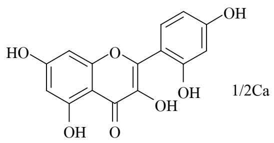
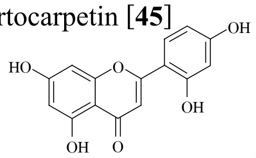
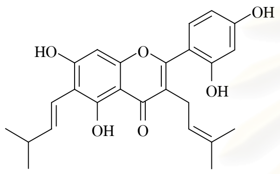
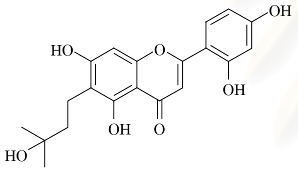
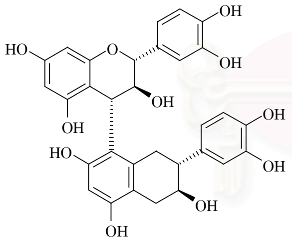
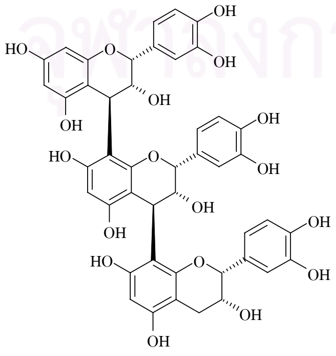
Plant and chemical compound	Plant part	Reference
Morin-calcium-chelate [**103**] 	Heartwood	Mu and Li, 1982
Norartocarpetin [**45**] 	Heartwood	Radhakrishnan *et al*., 1965
Norartocarpin [**67**] 	Heartwood	Venkataraman, 1972
Oxydihydroartocarpesin [**104**] 	Heartwood	Pathasarathy *et al*., 1969
Procyanidin B-3 [**105**] 	Leaf	An *et al*., 1992
Procyanidin C-1 [**106**] 	Leaf	An *et al*., 1992

Table 1 (continued)

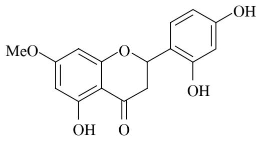
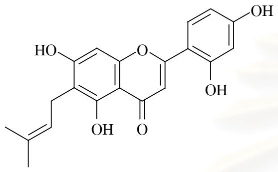
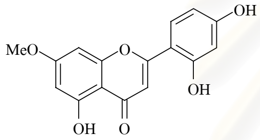
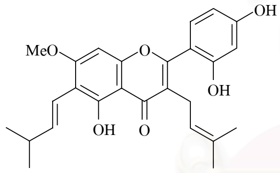
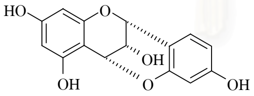
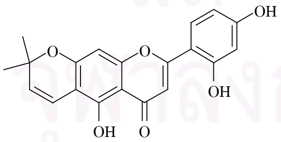
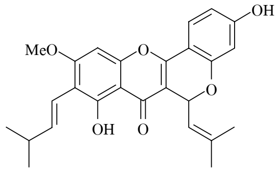
Plant and chemical compound	Plant part	Reference
A. hirsuta		
Artocarpanone [**71**] 	Heartwood	Venkataraman, 1972
Artocarpesin [**3**] 	Heartwood	Venkataraman, 1972
Artocarpetin [**73**] 	Heartwood	Venkataraman, 1972
Artocarpin [**4**] 	Heartwood	Venkataraman, 1972
Cyanomaclurin [**91**] 	Heartwood	Venkataraman, 1972
Cycloartocarpesin [**51**] 	Heartwood	Venkataraman, 1972
Cycloartocarpin [**30**] 	Heartwood	Venkataraman, 1972

Table 1 (continued)

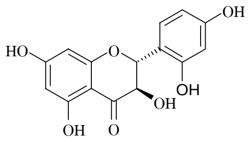
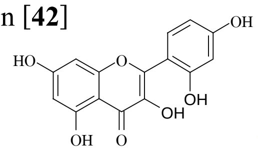
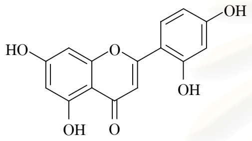
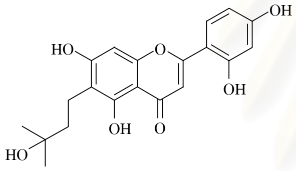
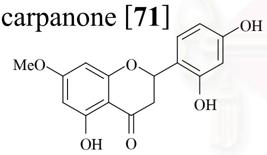
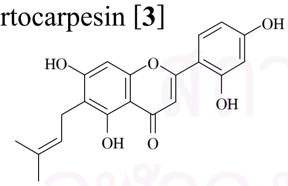
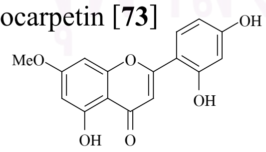
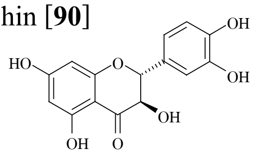
Plant and chemical compound	Plant part	Reference
Dihydromorin [**39**] 	Heartwood	Venkataraman, 1972
Morin [**42**] 	Heartwood	Venkataraman, 1972
Norartocarpetin [**45**] 	Heartwood	Venkataraman, 1972
Oxydihydroartocarpesin [**104**] 	Heartwood	Venkataraman, 1972
A. integer		
Artocarpanone [**71**] 	Heartwood	Pendse *et al*., 1976
Artocarpesin [**3**] 	Heartwood	Pendse *et al*., 1976
Artocarpetin [**73**] 	Heartwood	Pendse *et al*., 1976
Catechin [**90**] 	Leaf	Yamazaki *et al*., 1987

Table 1 (continued)

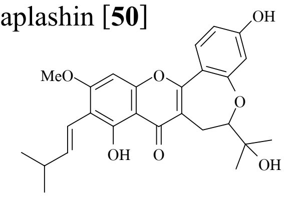
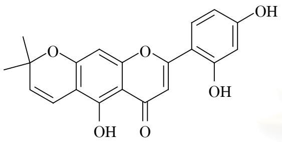
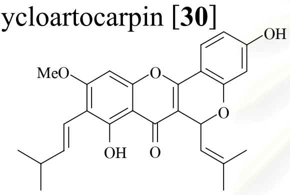
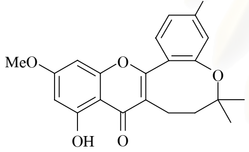
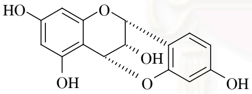
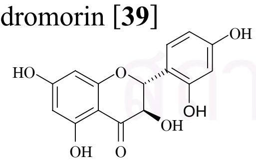
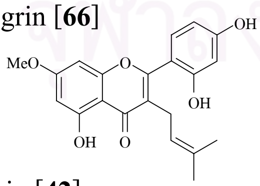
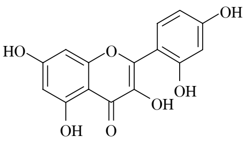
Plant and chemical compound	Plant part	Reference
Chaplashin [**50**] 	Heartwood	Pendse *et al*., 1976
Cycloartocarpesin [**51**] 	Heartwood	Pendse *et al*., 1976
Cycloartocarpin [**30**] 	Heartwood	Pendse *et al*., 1976
Cyclointegrin [**107**] 	Heartwood	Pendse *et al*., 1976
Cyanomaclurin [**91**] 	Heartwood	Pendse *et al*., 1976
Dihydromorin [**39**] 	Heartwood	Pendse *et al*., 1976
Integrin [**66**] 	Heartwood	Pendse *et al*., 1976
Morin [**42**] 	Heartwood	Pendse *et al*., 1976

Table 1 (continued)

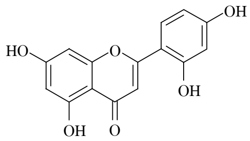
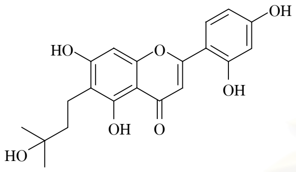
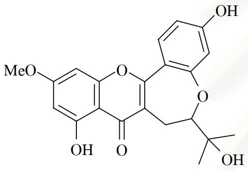
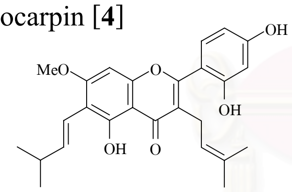
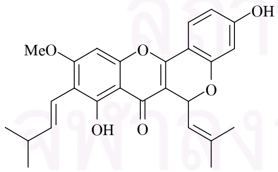
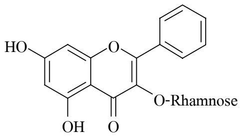
Plant and chemical compound	Plant part	Reference
Norartocarpetin [**45**] 	Heartwood	Pendse *et al*., 1976
Oxydihydroartocarpesin [**104**] 	Heartwood	Pendse *et al*., 1976
Oxyisocyclointegrin [**108**] 	Heartwood	Pendse *et al*., 1976
A. lakoocha		
Artocarpin [**4**] 	Heartwood	Venkataraman, 1972
Cycloartocarpin [**30**] 	Heartwood	Venkataraman, 1972
5,7-Dihydroxyflavone-3-*O*-α-L-rhamnoside [**109**] 	Root bark	Chauhan and Kumari, 1979

Table 1 (continued)

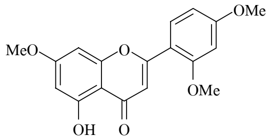
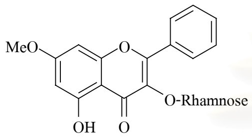
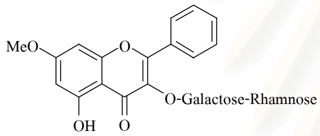
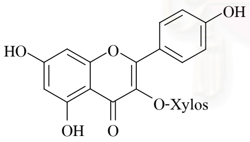
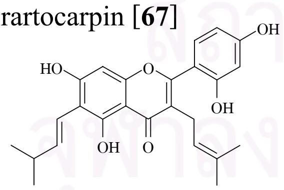
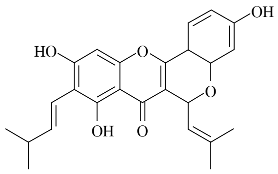
Plant and chemical compound	Plant part	Reference
5-Hydroxy-7,2',4'-trimethoxyflavone [**110**] 	Stem wood	Pavaro and Reutrakul, 1976
Galangin-3-*O*-α-L-(-)-rhamno-Pyranoside [**111**] 	Root bark	Chauhan and Kumari, 1979
Galangin-3-*O*-β-D-galactopyranosyl-(1⟶4)-α-L- Rhamnopyranoside [**112**] 	Root bark	Chauhan, Kumari and Saraswat, 1979
Kaempferol-3-*O*-β-D-xylanopyranoside [**113**] 	Root bark	Chauhan *et al.*, 1982
Norartocarpin [**67**] 	Heartwood	Venkataraman, 1972
Norcycloartocarpin [**114**] 	Heartwood	Venkataraman, 1972

Table 1 (continued)

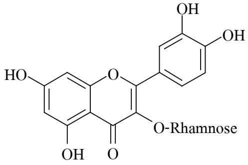
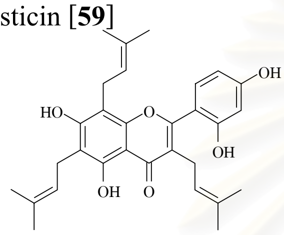
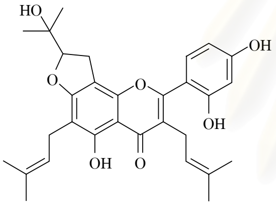
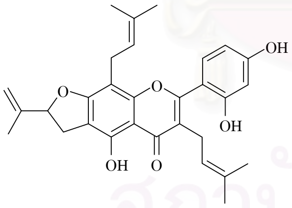
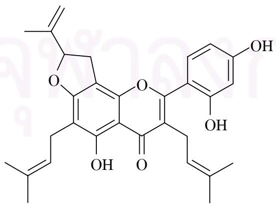
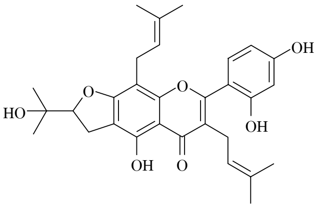
Plant and chemical compound	Plant part	Reference
Quercetin-3-*O*-α-L-rhamno-pyranoside [**115**] 	Root bark	Chauhan *et al*., 1982
A. lanceifolius		
Artelasticin [**59**] 	Heartwood	Syah *et al*., 2001
Artelastofuran [**64**] 	Heartwood	Syah *et al*., 2001
Artoindonesianin G [**116**] 	Heartwood	Syah *et al*., 2001
Artoindonesianin H [**117**] 	Heartwood	Syah *et al*., 2001
Artoindonesianin I [**118**] 	Heartwood	Syah *et al*., 2001

Table 1 (continued)

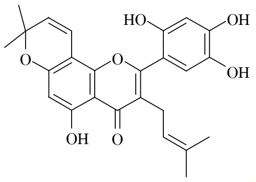
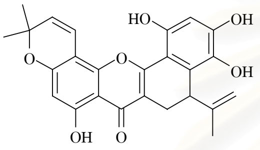
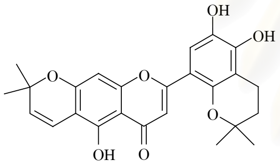
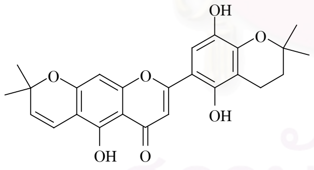
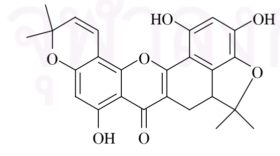
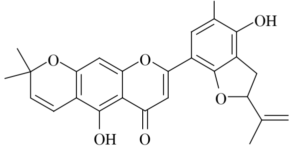
Plant and chemical compound	Plant part	Reference
A. nobilis		
Artobilochromen [**13**] (Artonin E) 	Stem bark	Pavanasasivam, Sultanbawa and Mageswaran, 1974; Kumar *et al.*, 1977; Sultanbawa and Surendrakumar, 1989
Artobiloxanthone [**119**] 	Stem bark	Sultanbawa and Surendrakumar, 1989
Chromanoartobilochromen A [**120**] 	Stem bark	Kumar *et al.*, 1977
Chromanoartobilochromen B [**121**] 	Stem bark	Pavanasasivum *et al.*, 1974; Kumar *et al.*, 1977
Cycloartobiloxanthone [**122**] 	Stem bark	Sultanbawa and Surendrakumar, 1989
Furanoartobilochromene A [**123**] 	Stem bark	Pavanasasivum *et al.*, 1974; Kumar *et al.*, 1977

Table 1 (continued)

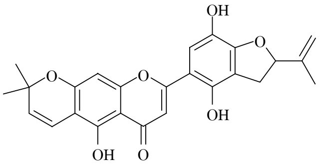
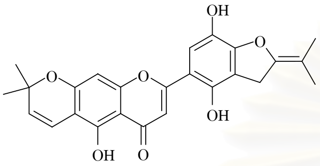
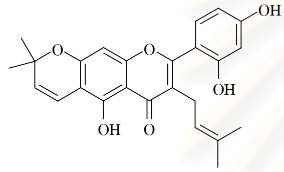
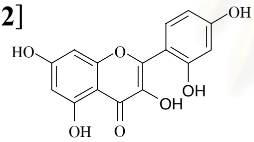
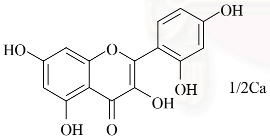
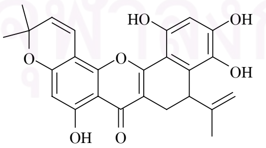
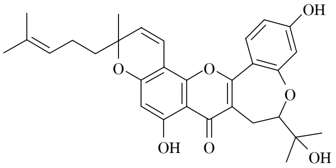
Plant and chemical compound	Plant part	Reference
Furanoartobilochromene B-1 [**124**] 	Stem bark	Pavanasasivum *et al*., 1974; Kumar *et al*., 1977
Furanoartobilochromene B-2 [**125**] 	Stem bark	Pavanasasivum *et al*., 1974; Kumar *et al*., 1977
Oxydihydromorusin [**126**] 	Stem bark	Kumar *et al*., 1977; Fukai and Nomura, 1993
A. pithecogalla		
Morin [**42**] 	Heartwood	Mu and Li, 1982
Morin-calcium-chelate [**103**] 	Heartwood	Mu and Li, 1982
A. rigida		
Artobiloxanthone [**119**] 	Stem bark	Hano, Inami, and Nomura, 1990
Artocarpol B [**127**] 	Root bark	Ko, Lin, and Yang, 2000

Table 1 (continued)

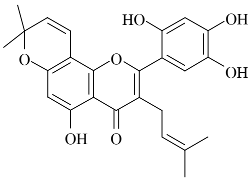
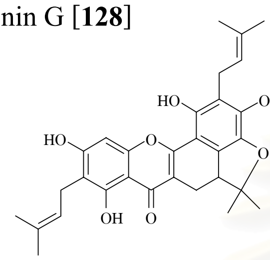
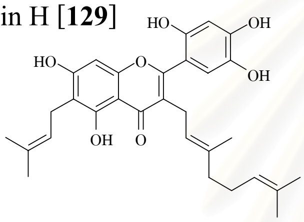
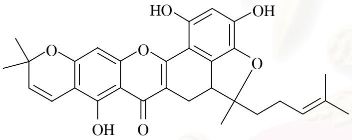
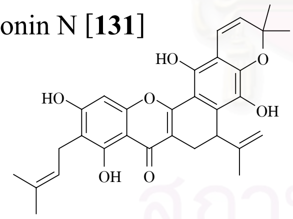
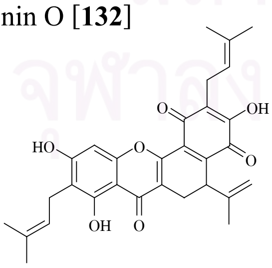
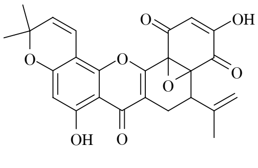
Plant and chemical compound	Plant part	Reference
Artonin E [**13**] 	Stem bark	Hano, Inami, and Nomura, 1990
Artonin G [**128**] 	Stem bark	Hano, Inami, and Nomura, 1990
Artonin H [**129**] 	Stem bark	Hano, Inami, and Nomura, 1990
Artonin M [**130**] 	Stem bark	Hano, Inami, and Nomura, 1993
Artonin N [**131**] 	Stem bark	Hano, Inami, and Nomura, 1993
Artonin O [**132**] 	Stem bark	Hano, Inami, and Nomura, 1993
Artonin P [**133**] 	Stem bark	Hano, Inami, and Nomura, 1993

Table 1 (continued)

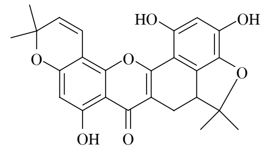
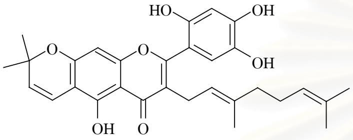
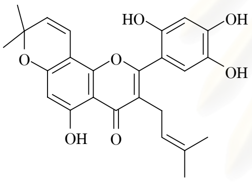
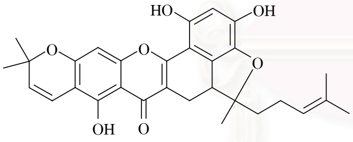
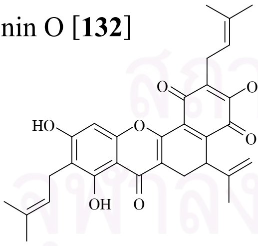
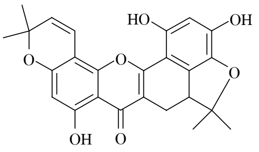
Plant and chemical compound	Plant part	Reference
Cycloartobiloxanthone [**122**] 	Stem bark	Hano, Inami, and Nomura, 1990
A. rotunda		
Artoindonesianin L [**134**] 	Root bark	Suhartati *et al*., 2001
Artonin E [**13**] 	Root bark	Suhartati *et al*., 2001
Artonin M [**130**] 	Root bark	Suhartati *et al*., 2001
Artonin O [**132**] 	Root bark	Suhartati *et al*., 2001
Cycloartobiloxanthone [**122**] 	Root bark	Suhartati *et al*., 2001

Table 1 (continued)

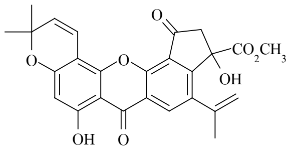
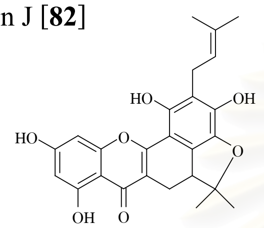
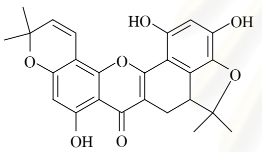
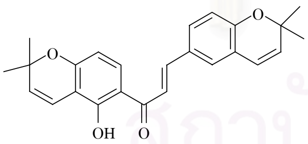
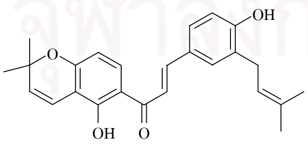
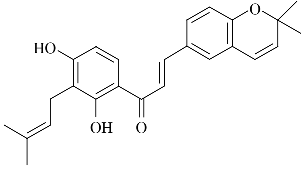
Plant and chemical compound	Plant part	Reference
A. teysmanii		
Artoindonesianin C [**135**] 	Root bark	Makmur *et al*., 2000
Artonin J [**82**] 	Root bark	Makmur *et al*., 2000
Cycloartobiloxanthone [**122**] 	Root bark	Makmur *et al*., 2000
A. tonkiensis		
Artotonkin [**136**]	Stem bark	Lein *et al*., 1998
A. venenosa		
Paratocarpin A [**137**] 	Stem bark	Hano *et al*., 1995a; Nomura, Hano, and Aida, 1998
Paratocarpin B [**138**] 	Stem bark	Hano *et al*., 1995a; Nomura, Hano, and Aida, 1998
Paratocarpin C [**139**] 	Stem bark	Hano *et al*., 1995a; Nomura, Hano, and Aida, 1998

Table 1 (continued)

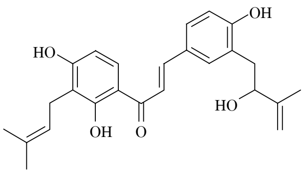
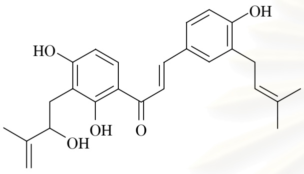
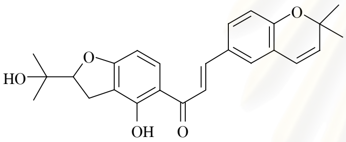
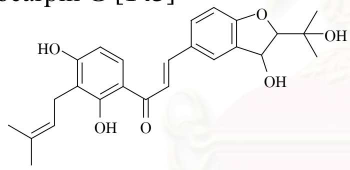
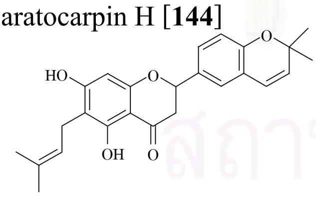
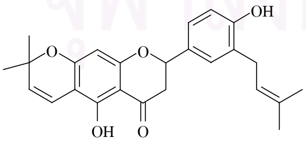
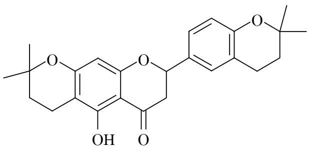
Plant and chemical compound	Plant part	Reference
Paratocarpin D [**140**] 	Stem bark	Hano *et al*., 1995a; Nomura, Hano, and Aida, 1998
Paratocarpin E [**141**] 	Stem bark	Hano *et al*., 1995a; Nomura, Hano, and Aida, 1998
Paratocarpin F [**142**] 	Stem bark	Hano *et al*., 1995b; Nomura, Hano, and Aida, 1998
Paratocarpin G [**143**] 	Stem bark	Hano *et al*., 1995b; Nomura, Hano, and Aida, 1998
Paratocarpin H [**144**] 	Stem bark	Hano *et al*., 1995b; Nomura, Hano, and Aida, 1998
Paratocarpin I [**145**] 	Stem bark	Hano *et al*., 1995b; Nomura, Hano, and Aida, 1998
Paratocarpin J [**146**] 	Stem bark	Hano *et al*., 1995b; Nomura, Hano, and Aida, 1998

Table 1 (continued)

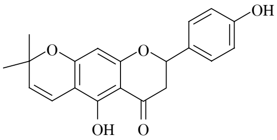
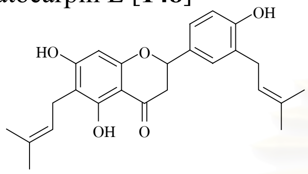
Plant and chemical compound	Plant part	Reference
Paratocarpin K [**147**] 	Stem bark	Hano *et al*., 1995b; Nomura, Hano, and Aida, 1998
Paratocarpin L [**148**] 	Stem bark	Hano *et al*., 1995b; Nomura, Hano, and Aida, 1998

Table 2 Distribution of triterpenoids in *Artocarpus*.

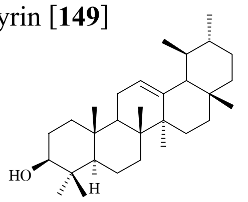
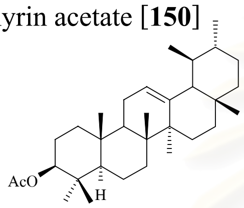
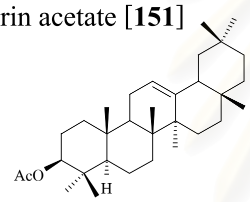
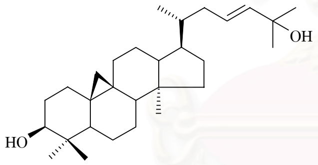
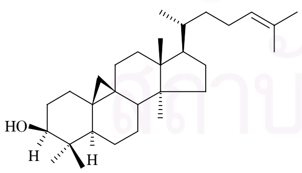
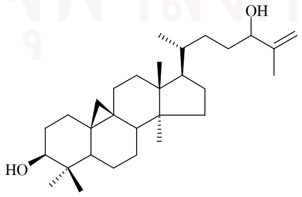
Plant and chemical compound	Plant part	Reference
A. altilis		
α-Amyrin [**149**] 	Latex	Ultee, 1949
α-Amyrin acetate [**150**] 	Fruit	Altman and Zito, 1976
β-Amyrin acetate [**151**] 	Latex	Ultee, 1949
Cycloart-23-ene-3β-25-diol [**152**] 	Fruit	Altman and Zito, 1976
Cycloart-24-ene-3β-ol [**153**] 	Fruit	Altman and Zito, 1976
Cycloart-25-ene-3β-24-diol [**154**] 	Fruit	Altman and Zito, 1976

Table 2 (continued)

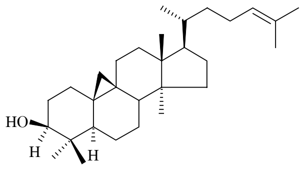
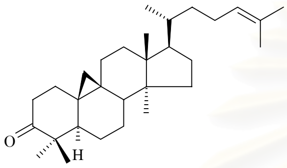
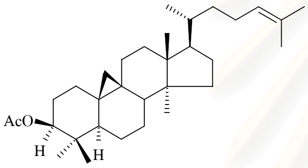
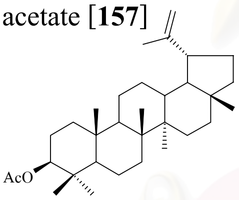
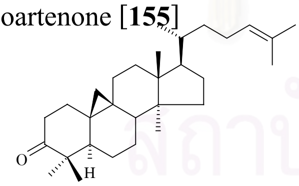
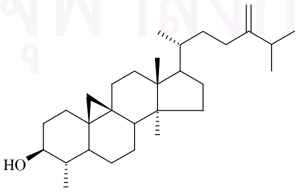
Plant and chemical compound	Plant part	Reference
Cycloartenol [**153**] (Cycloart-24-ene-3β-ol) 	Stem bark	Pavanasasivam and Sultanbawa, 1973
Cycloartenone [**155**] 	Stem bark	Pavanasasivam and Sultanbawa, 1973
Cycloartenyl acetate [**156**] 	Stem bark	Pavanasasivam and Sultanbawa, 1973
Lupeol acetate [**157**] 	Root bark	Shieh and Lin, 1992
A. champeden		
Cycloartenone [**155**] 	Stem bark	Achmad *et al*., 1996
Cycloeucalenol [**158**] 	Stem bark	Achmad *et al*., 1996

Table 2 (continued)

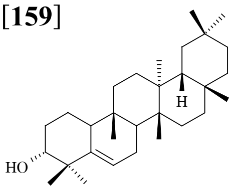
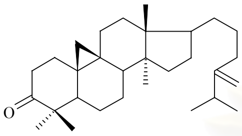
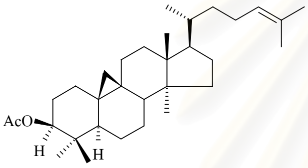
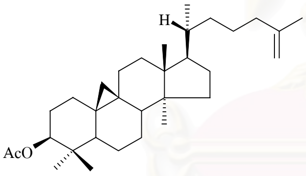
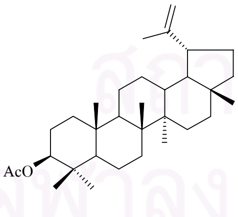
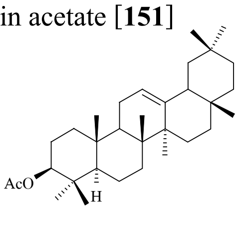
Plant and chemical compound	Plant part	Reference
Glutinol [**159**] 	Stem bark	Achmad *et al.*, 1996
24-Methylenecycloartanone [**160**] 	Stem bark	Achmad *et al.*, 1996
A. chaplasha		
Cycloartenyl acetate [**156**] 	Stem bark	Mahato, Banerjee, and Chakravarti, 1971
Isocycloartenol acetate [**161**] 	Stem bark	Mahato *et al.*, 1971
Lupeol acetate [**157**] 	Stem bark	Mahato *et al.*, 1971
A. elasticus		
β-Amyrin acetate [**151**] 	Latex	Ultee, 1949

Table 2 (continued)

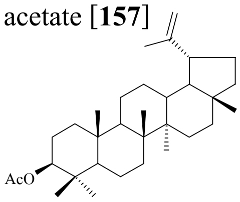
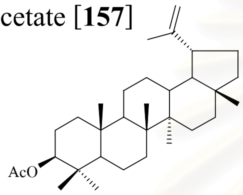
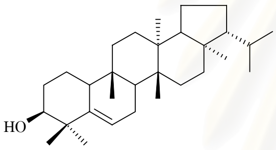
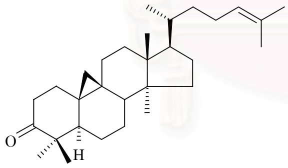
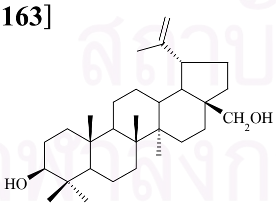
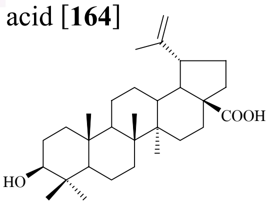
Plant and chemical compound	Plant part	Reference
Lupeol acetate [**157**]  	Latex	Ultee, 1949
A. gomezianus		
Lupeol acetate [**157**]  	Leaf	Kingroungpet, 1994
Simiarenol [**162**]  	Leaf	Kingroungpet, 1994
A. heterophyllus		
Artostenone (Cycloartenone) [**155**]  	Fruit	Nath and Mukherjee, 1939
Betulin [**163**]  	Root bark	Lu and Lin, 1994
Betulinic acid [**164**]  	Root Root bark	Dayal and Seshadri, 1974; Lu and Lin, 1994

Table 2 (continued)

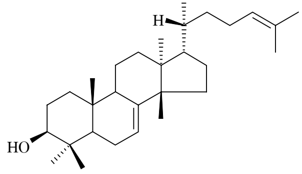
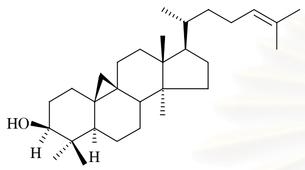
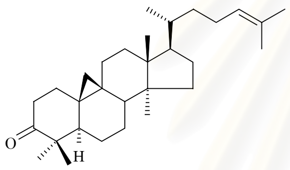
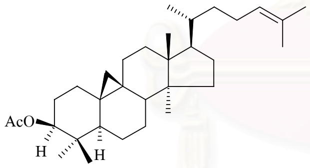
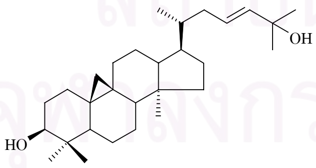
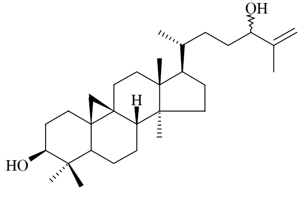
Plant and chemical compound	Plant part	Reference
Butyrospermol [**165**] 	Fruit	Barton, 1951
Cycloartenol [**153**] 	Fruit Wood Stem bark Latex	Barton, 1951; Nogueira and Correia, 1958; Pavanasasivam and Sultanbawa, 1973; Barik *et al*., 1994
Cycloartenone [**155**] 	Fruit Stem bark Root Latex	Barton, 1951; Pavanasasivam and Sultanbawa, 1973; Dayal and Seshadri, 1974; Pant and Chaturvedi, 1989; Barik *et al*., 1994
Cycloartenyl acetate [**156**] 	Stem bark	Pavanasasivam and Sultanbawa, 1973
9,19-Cyclolanost-23-ene-3β,25-diol (Cycloart-23-ene-3,25-diol) [**152**] 	Fruit	Kielland and Malterud, 1994
9,19-Cyclolanost-25-ene-3β,24-diol [**166**] 	Fruit	Kielland and Malterud, 1994; Barik *et al*., 1997

Table 2 (continued)

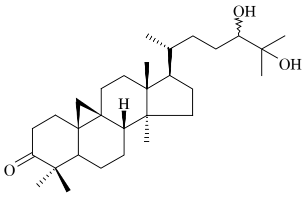
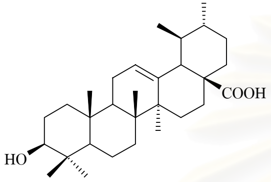
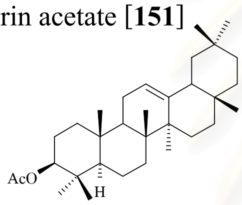
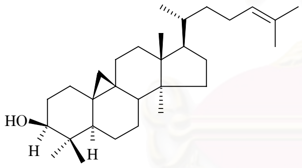
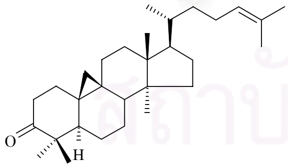
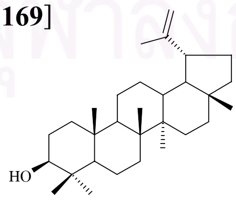
Plant and chemical compound	Plant part	Reference
9,19-Cyclolanost-3-one-24,25-diol [**167**] 	Latex	Barik *et al.*, 1994
Ursolic acid [**168**] 	Root Root bark	Dayal and Seshadri, 1974; Lu and Lin, 1994
A. lakoocha		
β-Amyrin acetate [**151**] 	Stem bark	Kapil and Joshi, 1960
Cycloartenol [**153**] 	Stem bark	Pavanasasivam and Sultanbawa, 1973
Cycloartenone [**155**] 	Stem bark	Pavanasasivam and Sultanbawa, 1973
Lupeol [**169**] 	Root bark	Chauhan and Kumari, 1979;

Table 2 (continued)

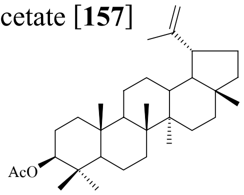
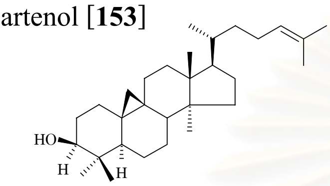
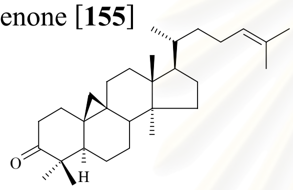
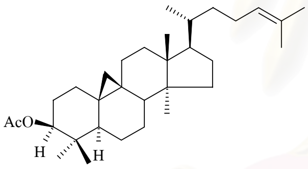
Plant and chemical compound	Plant part	Reference
Lupeol acetate [**157**] 	Stem bark	Kapil and Joshi, 1960
A. nobilis		
Cycloartenol [**153**] 	Stem bark Heartwood	Pavanasasivam and Sultanbawa, 1973
Cycloartenone [**155**] 	Stem bark Heartwood	Pavanasasivam and Sultanbawa, 1973
Cycloartenyl acetate [**156**] 	Stem bark Heartwood	Pavanasasivam and Sultanbawa, 1973

Table 3 Distribution of miscellaneous compounds in *Artocarpus*.

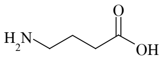
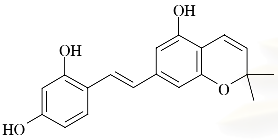
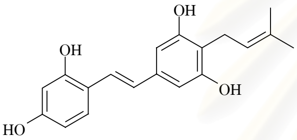
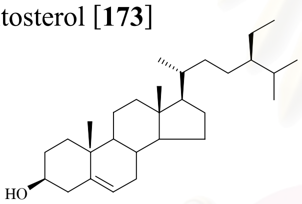
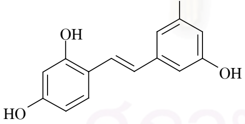
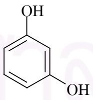
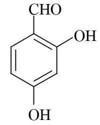
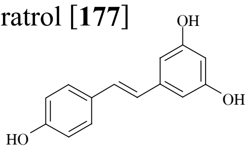
Plant and chemical compound	Category	Plant part	Reference
A. altilis			
γ-Aminobutyric acid [**170**] 	Amino acid	Leaf	Durand *et al.*, 1962
Artocarbene [**171**] 	Stilbene	Heartwood	Shimizu *et al.*, 1997
4-Prenyloxyresveratrol [**172**] 	Stilbene	Heartwood	Shimizu *et al.*, 1997
β-Sitosterol [**173**] 	Steroid	Root bark	Shieh and Lin, 1992
A. chaplasha			
Oxyresveratrol [**174**] 	Stilbene	Heartwood	Rao *et al.*, 1972
Resorcinol [**175**] 	Benzenoid	Heartwood	Rao *et al.*, 1972
β-Resorcyaldehyde [**176**] 	Benzenoid	Heartwood	Rao *et al.*, 1972
Resveratrol [**177**] 	Stilbene	Heartwood	Rao *et al.*, 1972

Table 3 (continued)

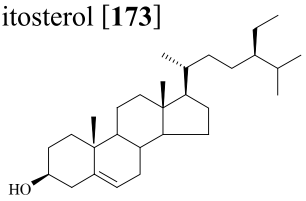
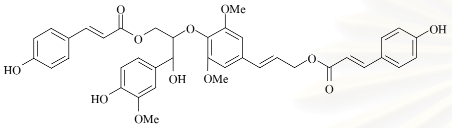
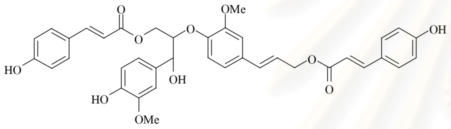
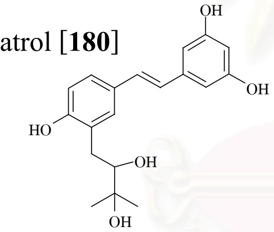
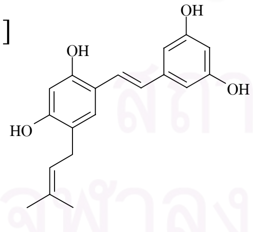
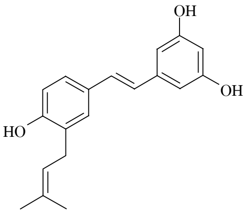
Plant and chemical compound	Category	Plant part	Reference
β-Sitosterol [**173**] 	Steroid	Stem bark	Mahato *et al.*, 1971
A. dadah			
Dadahol A [**178**] 	Neolignan	Twig	Su *et al.*, 2002
Dadahol B [**179**] 	Neolignan	Twig	Su *et al.*, 2002
3-(2,3-dihydroxy-3-methylbutyl)-resveratrol [**180**] 	Stilbene	Stem bark	Su *et al.*, 2002
3-(γ,γ-dimethylallyl)oxyresveratrol [**181**] 	Stilbene	Stem bark	Su *et al.*, 2002
3-(γ,γ-dimethylallyl)resveratrol [**182**] 	Stilbene	Stem bark	Su *et al.*, 2002

Table 3 (continued)

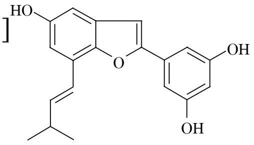
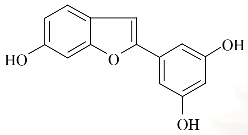
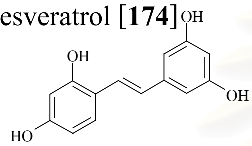
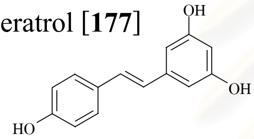
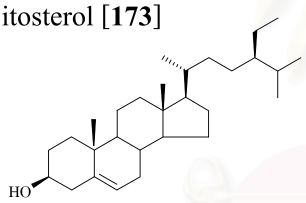
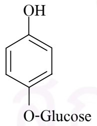
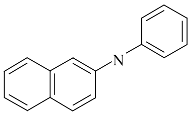
Plant and chemical compound	Category	Plant part	Reference
3-(γ,γ-dimethylpropenyl)moracin M [**183**] 	Stilbene	Stem bark	Su *et al.*, 2002
Moracin M [**184**] 	Stilbene	Twig	Su *et al.*, 2002
Oxyresveratrol [**174**] 	Stilbene	Stem bark Twig	Su *et al.*, 2002
Resveratrol [**177**] 	Stilbene	Twig	Su *et al.*, 2002
A. elasticus			
β-Sitosterol [**173**] 	Steroid	Heartwood	Pendse *et al.*, 1976
A. gomezianus			
Arbutin [**185**] 	Phenolic glycoside	Leaf	Kingroungpet, 1994
1-Dotriacontanol [**186**] $HOCH_2CH_2(CH_2)_{29}CH_3$	Alcohol	Leaf	Kingroungpet, 1994
Mesoerythritol [**187**]	Phenolic compound	Heartwood	Venkataraman, 1972
Phenyl-β-naphthylamine [**188**] 	Naphthalene	Root	Sritularak, 1998; Likhitwitayawuid, Sritularak and De-Ek-Namkul, 2000

Table 3 (continued)

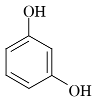
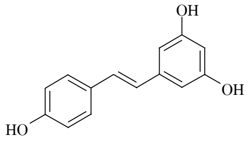
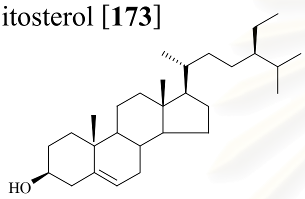
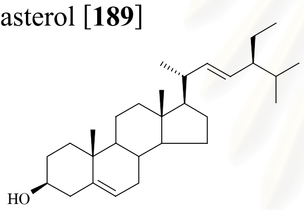
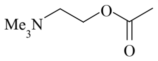
Plant and chemical compound	Category	Plant part	Reference
Resorcinol [**175**] 	Benzenoid	Root	Sritularak, 1998
Resveratrol [**177**] 	Stilbene	Root	Sritularak, 1998; Likhitwitayawuid, Sritularak and De-Ek-Namkul, 2000
β-Sitosterol [**173**] 	Steroid	Leaf	Kingroungpet, 1994
Stigmasterol [**189**] 	Steroid	Root	Sritularak, 1998
A. heterophyllus			
Acetylcholine [**190**] 	Amine	Seed	Pereira, Medina and Bustos, 1962
Artocarpus integra α-D-Galactose specific lectin [**191**]	Lectin	Seed	Suresh, Appukuttan, and Basu, 1982
Artocarpus integrifolia lectin [**192**]	Lectin	Seed	Chatterjee, Sarkar, and Rao, 1982; Namjuntra and Culavatnatol, 1984
Artocarpus lectin CE-A-I [**193**]	Lectin	Seed	Ferreira *et al*., 1992

Table 3 (continued)

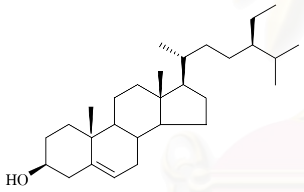
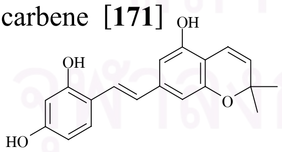
Plant and chemical compound	Category	Plant part	Reference
Aurantiamide acetate [**194**]	Protein	Seed	Chakraborty and Mandal, 1981
9-Hydroxytridecyl docosanoate [**195**] $CH_3(CH_2)_{20}COO(CH_2)_8CH(OH)(CH_2)_3CH_3$	Lipid	Root bark	Lu and Lin, 1994
4-Hydroxyundecyl docosanoate [**196**] $CH_3(CH_2)_{20}COO(CH_2)_3CH(OH)(CH_2)_6CH_3$	Lipid	Latex	Pant and Chaturvedi, 1989
Jacalin [**197**]	Lectin	Seed	Hagiwara *et al*., 1988; Ferreira *et al*., 1992
Lymphoagglutinin [**198**]	Lectin	Seed	Arora *et al*., 1987
Recinoleic acid [**199**] $CH_3(CH_2)_5CH(OH)CH_2CH=CH(CH_2)_7COOH$	Lipid	Seed oil	Daulatabad and Mirajkar, 1989
β-Sitosterol [**173**]	Steroid	Heartwood	Pathasarathy *et al.,* 1969;
		Root	Dayal and Seshadri, 1974;
		Root bark	Lu and Lin, 1994
A. hirsuta			
Lymphoagglutinin [**198**]	Lectin	Seed	Arora *et al*., 1987
A. integer			
Artocarbene [**171**] 	Stilbene	Aerial part	Boonlaksiri *et al*., 2000
Artocarpus lectin C [**200**]	Lectin	Seed	Hashim, Gendeh and Jaafar, 1992

Table 3 (continued)

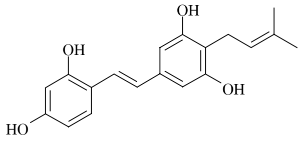
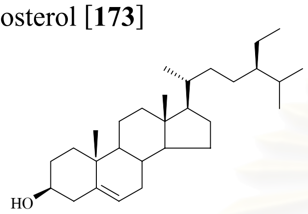
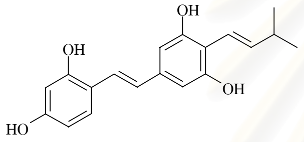
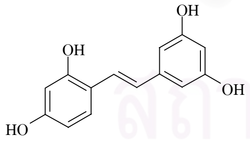
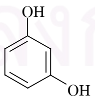
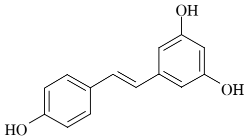
Plant and chemical compound	Category	Plant part	Reference
4-Prenyloxyresveratrol [**172**] 	Stilbene	Aerial part	Boonlaksiri *et al.*, 2000
β-Sitosterol [**173**] 	Steroid	Heartwood	Pendse *et al.*, 1976
Tran-4-(3-methyl-E-but-1-enyl)-3,5,2',4'-tetrahydroxystilbene [**201**] 	Stilbene	Aerial part	Boonlaksiri *et al.*, 2000
A. lakoocha			
ALA-I [**202**]	Isolectin	Seed	Wongkham *et al.*, 1995
ALA-II [**203**]	Isolectin	Seed	Wongkham *et al.*, 1995
Artocarpus lakoocha lectin [**204**]	Lectin	Seed	Chatterjee *et al.*, 1982
Lymphoagglutinin [**198**]	Lectin	Seed	Arora *et al.*, 1987
Oxyresveratrol [**174**] 	Stilbene	Heartwood	Venkataraman, 1972; Mongolsuk,Robertson and Towers, 1957
Resorcinol [**175**] 	Benzenoid	Heartwood	Venkataraman, 1972
Resveratrol [**177**] 	Stilbene	Heartwood	Venkataraman, 1972

Table 3 (continued)

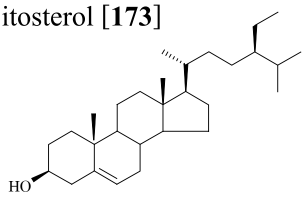
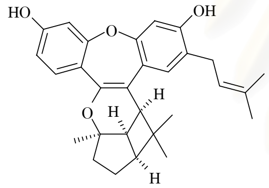
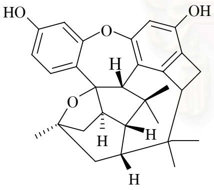
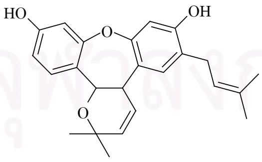
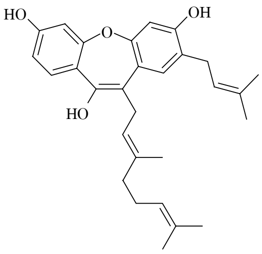
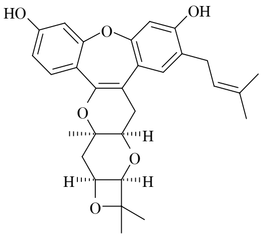
Plant and chemical compound	Category	Plant part	Reference
β-Sitosterol [**173**] 	Steroid	Root bark	Chauhan and Kumari, 1979
A. lignanensis Artocarpus lectin [**205**]		Seed	Zhang *et al.*, 1999
A. masticatus Artocarpus lectin AM [**206**]		Seed	Blasco *et al.*, 1996
A. melinoxylus Artocarpus lectin AME [**207**]		Seed	Blasco *et al.*, 1996
A. rigida Artocarpol A [**208**] 	Phenolics	Root bark	Chung *et al.*, 2000
Artocarpol C [**209**] 	Phenolics	Root bark	Ko, Lin, and Yang, 2000
Artocarpol D [**210**] 	Phenolics	Root bark	Ko, Lin, and Yang, 2000
Artocarpol E [**211**] 	Phenolics	Root bark	Ko, Lin, and Yang, 2000

Table 3 (continued)

Plant and chemical compound	Category	Plant part	Reference
Artocarpol F [**212**] 	Phenolics	Root bark	Ko, Yang, and Lin, 2001

2. Chemical Constituents of *Millettia* spp.

Chemical investigations of a number of *Millettia* species have shown them to be a good source of flavonoids. In addition, other classes of natural compounds such as coumarins, nitrogenous compound, terpenoids and miscellaneous substances have been found (Tables 4-5).

Table 4 Distribution of flavonoids in *Millettia*.

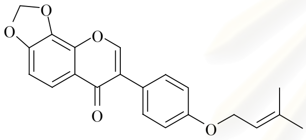
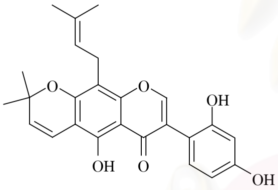
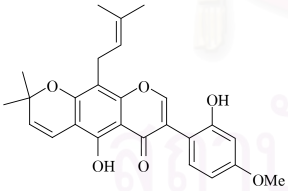
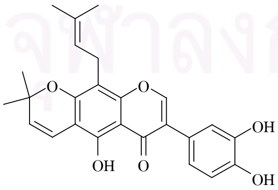
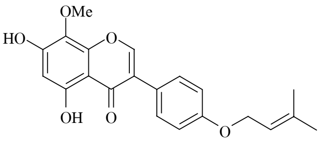
Plant and chemical compound	Plant part	Reference
Millettia auriculata		
Auricularin [**213**] 	Root	Rao, Prasad, and Ganapaty, 1992
Auriculatin [**214**] 	Root	Raju *et al*., 1981; Rao, Prasad, and Ganapaty, 1992
Auriculin [**215**] 	Root	Rao, Prasad, and Ganapaty, 1992
Auriculasin [**216**] 	Seed Leaf	Raju and Srimannarayana, 1978
Aurmillone [**217**] 	Seed	Raju and Srimannarayana, 1978

Table 4 (continued)

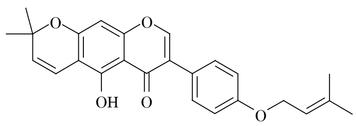
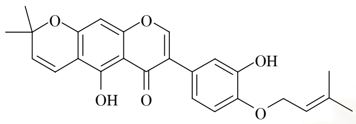
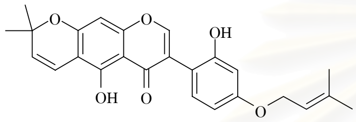
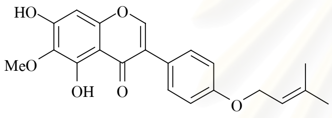
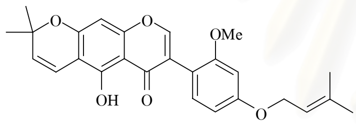
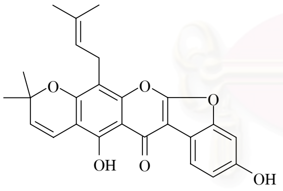
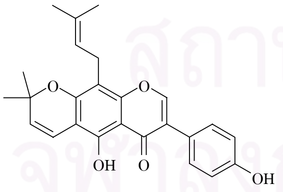
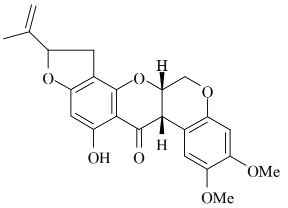
Plant and chemical compound	Plant part	Reference
2'-Deoxyisoauriculatin [**218**] 	Root	Rao, Prasad, and Ganapaty, 1992
Isoauriculasin [**219**] 	Leaf	Minhaj *et al*., 1976
Isoauriculatin [**220**] 	Root	Minhaj *et al*., 1976
Isoaurmillone [**221**] 	Pod	Gupta *et al*., 1983
2'-*O*-Methylisoauriculatin [**222**] 	Root	Rao, Prasad, and Ganapaty, 1992
Millettin [**223**] 	Root	Rao, Prasad, and Ganapaty, 1992
Scandenone [**224**] 	Root	Rao, Prasad, and Ganapaty, 1992
Sumatrol [**225**] 	Root Seed	Rao, Prasad, and Ganapaty, 1992

Table 4 (continued)

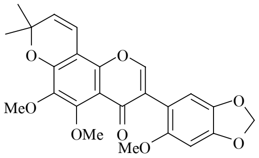
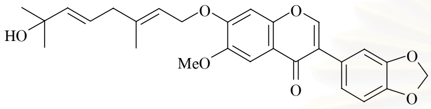
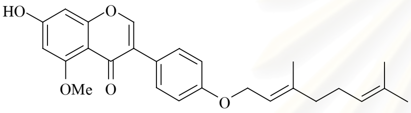
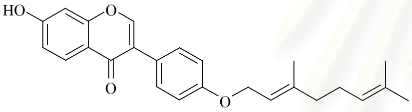
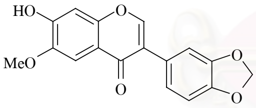
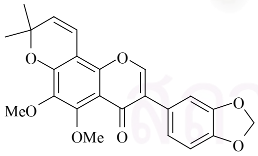
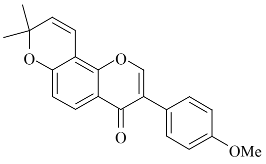
Plant and chemical compound	Plant part	Reference
M. conraui		
Conrauinone A [**226**] 	Stem bark	Fuendjiep *et al*., 1998a
Conrauinone B [**227**] 	Stem bark	Fuendjiep *et al*., 1998a
Conrauinone C [**228**] 	Stem bark	Fuendjiep *et al*., 1998b
Conrauinone D [**229**] 	Stem bark	Fuendjiep *et al*., 1998b
7-Hydroxy-6-methoxy-3',4'-methylenedioxyisoflavone [**230**] 	Stem bark	Fuendjiep *et al*., 1998b
5-Methoxydurmillone [**231**] 	Stem bark	Fuendjiep *et al*., 1998a
M. dura		
Calopogonium isoflavone A [**232**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1996

Table 4 (continued)

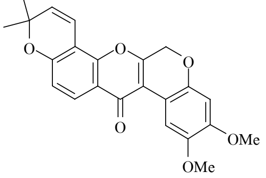
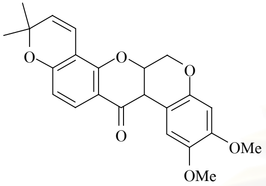
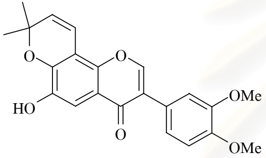
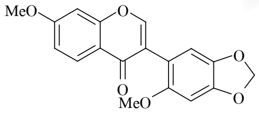
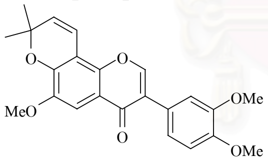
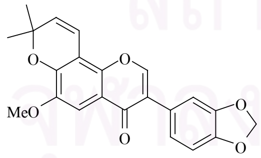
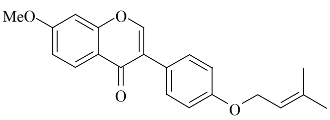
Plant and chemical compound	Plant part	Reference
6a,12a-Dehydrodeguelin [**233**] 	Seed	Ollis, Rhodes, and Sutherland, 1967
Deguelin [**234**] 	Seed	Dagne, Mammo, and Bekele, 1991
6-Demethyldurallone [**235**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1996
7,2'-Dimethoxy-4',5'-methylenedioxy-isoflavone [**236**] 	Stem bark Root bark	Dagne, Mammo, and Bekele, 1991
Durallone [**237**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1996
Durmillone [**238**] 	Stem bark Seed	Ollis, Rhodes, and Sutherland, 1967
Durlettone [**239**] 	Seed	Ollis, Rhodes, and Sutherland, 1967

Table 4 (continued)

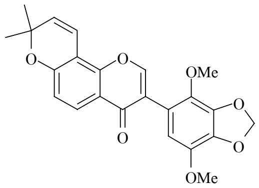
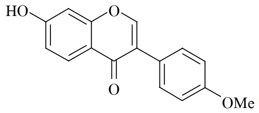
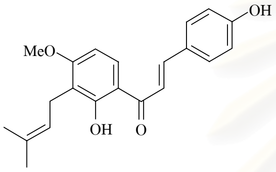
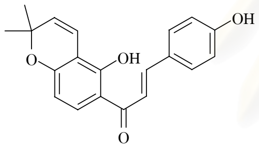
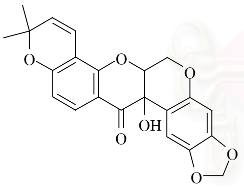
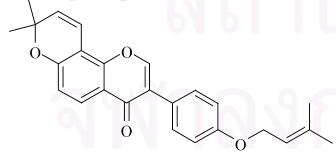
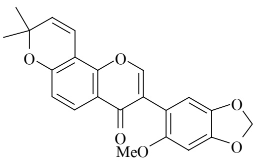
Plant and chemical compound	Plant part	Reference
Ferrugone [**240**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1997
Formononetin [**241**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1997
4-Hydroxyderricin [**242**] 	Stem bark Root bark	Dagne, Mammo, and Bekele, 1991
4-Hydroxylonchocarpin [**243**] 	Stem bark Root bark	Dagne, Mammo, and Bekele, 1991
12-Hydroxymilletone [**244**] 	Seed	Dagne, Mammo, and Bekele, 1991
Isoerythrin-A 4'-(3-methylbut-2-enyl) Ether [**245**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1996
Jamaicin [**246**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1997

Table 4 (continued)

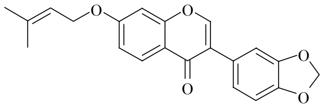
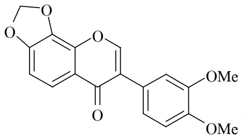
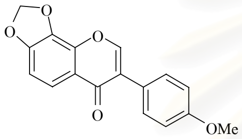
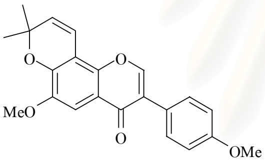
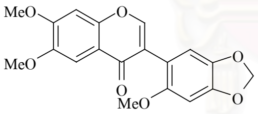
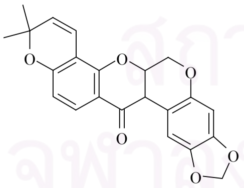
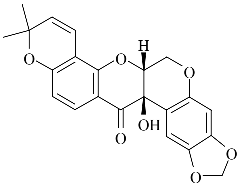
Plant and chemical compound	Plant part	Reference
Maximaisoflavone B [**247**] 	Stem bark Root bark	Dagne, Mammo, and Bekele, 1991
Maximaisoflavone D [**248**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1996
Maximaisoflavone H [**249**] 	Stem bark Root bark	Dagne, Mammo, and Bekele, 1991
6-Methoxycalopogonium isoflavoneA [**250**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1997
Milldurone [**251**] 	Seed	Ollis, Rhodes, and Sutherland, 1967
Millettone [**252**] 	Seed pod Seed	Ollis, Rhodes, and Sutherland, 1967
(-)-Millettosin [**253**] 	Seed	Ollis, Rhodes, and Sutherland, 1967

Table 4 (continued)

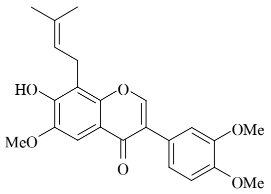
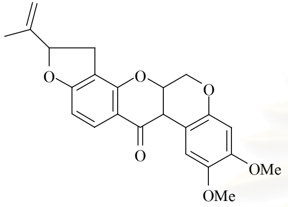
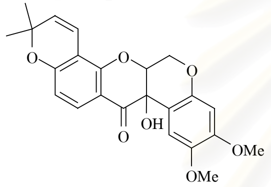
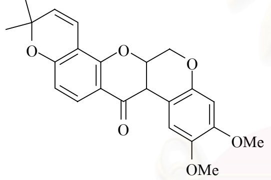
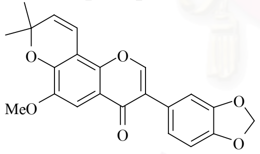
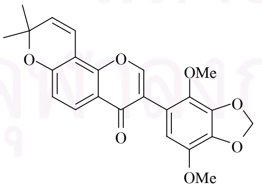
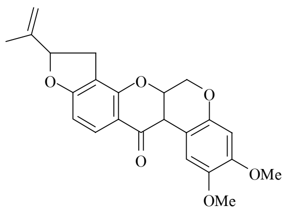
Plant and chemical compound	Plant part	Reference
Predurallone [**254**] 	Seed pod	Yenesew, Midiwo, and Waterman, 1996
Rotenone [**255**] 	Seed	Ollis, Rhodes, and Sutherland, 1967
Tephrosin [**256**] 	Seed pod Seed	Ollis, Rhodes, and Sutherland, 1967
M. ferruginea		
Deguelin [**234**] 	Seed	Highet and Highet, 1967
Durmillone [**238**] 	Seed	Highet and Highet, 1967
Ferrugone [**240**] 	Seed	Highet and Highet, 1967
Rotenone [**255**] 	Seed	Highet and Highet, 1967

Table 4 (continued)

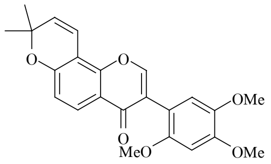
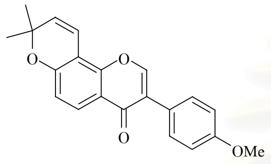
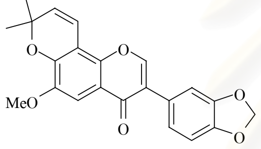
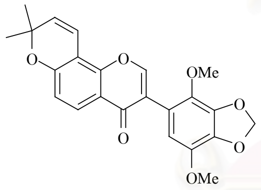
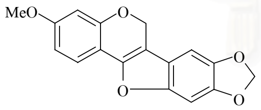
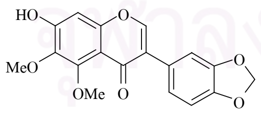
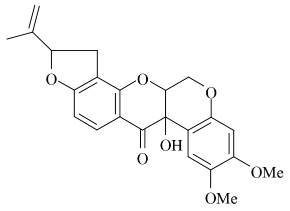
Plant and chemical compound	Plant part	Reference
M. ferruginea* subsp. *darassana		
Barbigerone [**257**] 	Seed	Dagne and Bekele, 1990
Calopogonium isoflavone-A [**232**] 	Seed	Dagne and Bekele, 1990
Durmillone [**238**] 	Seed Seed pod	Dagne and Bekele, 1990; Dagne and Bekele, 1989
Ferrugone [**240**] 	Seed Stem bark	Dagne and Bekele, 1990; Dagne and Bekele, 1989
Flemichapparin [**258**] 	Stem bark	Dagne and Bekele, 1989
7-Hydroxy-5,6-dimethoxy-3',4'-methylenedioxyisoflavone [**259**] 	Stem bark	Dagne and Bekele, 1989
12a-Hydroxyrotenone [**260**] 	Seed	Dagne and Bekele, 1990

Table 4 (continued)

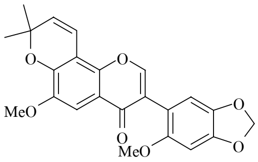
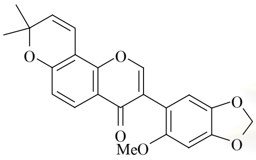
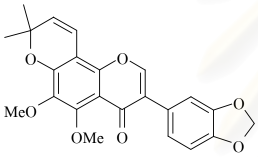
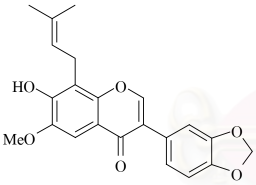
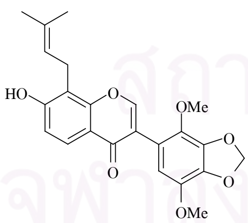
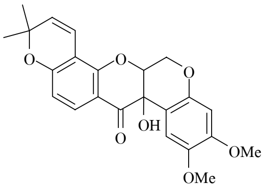
Plant and chemical compound	Plant part	Reference
Ichthynone [**261**] 	Bark	Dagne and Bekele, 1989
Jamaicin [**246**] 	Bark	Dagne and Bekele, 1989
5-Methoxydurmillone [**231**] 	Bark	Dagne and Bekele, 1989
Predurmillone [**262**] 	Seed	Dagne and Bekele, 1990
Preferrugone [**263**] 	Seed	Dagne and Bekele, 1990
Tephrosin [**256**] 	Seed	Dagne and Bekele, 1990

Table 4 (continued)

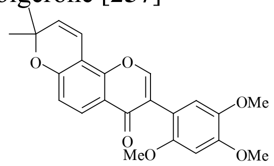
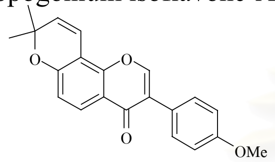
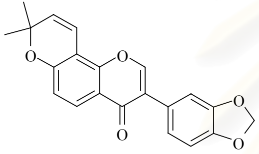
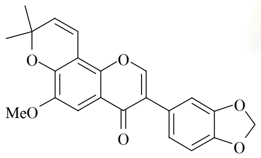
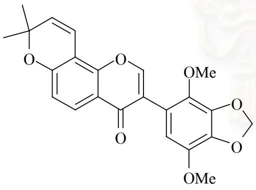
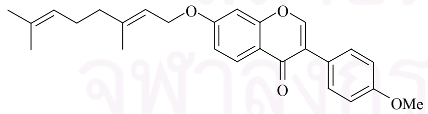
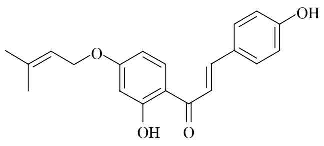
Plant and chemical compound	Plant part	Reference
M. ferruginea* subsp. *ferruginea		
Barbigerone [**257**] 	Seed	Dagne and Bekele, 1990
Calopogonium isoflavone-A [**232**] 	Seed	Dagne and Bekele, 1990
Calopogonium isoflavone-B [**264**] 	Stem bark	Dagne and Bekele, 1989
Durmillone [**238**] 	Seed	Dagne and Bekele, 1990
Ferrugone [**240**] 	Seed Root bark	Dagne and Bekele, 1990; Dagne *et al.*, 1990
7-*O*-geranylformononetin [**265**] 	Root bark	Dagne *et al.*, 1990
4'-*O*-geranylisoliquiritigenin [**266**] 	Root bark	Dagne *et al.*, 1990

Table 4 (continued)

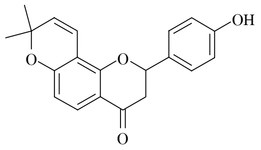
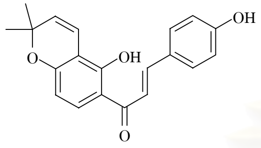
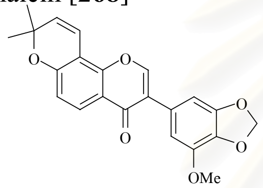
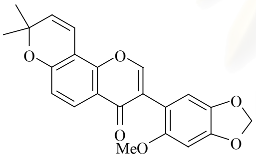
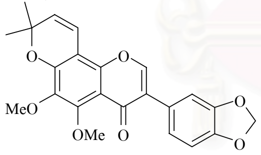
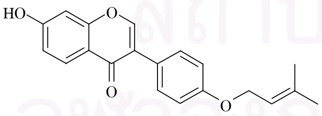
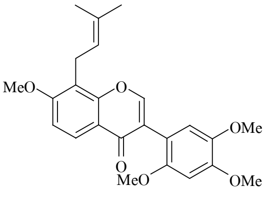
Plant and chemical compound	Plant part	Reference
4'-Hydroxyisolonchocarpin [**267**] 	Stem bark	Dagne and Bekele, 1989
4-Hydroxylonchocarpin [**243**] 	Stem bark	Dagne and Bekele, 1989
Isojamaicin [**268**] 	Stem bark	Dagne and Bekele, 1989
Jamaicin [**246**] 	Stem bark Root bark	Dagne and Bekele, 1989; Dagne *et al.*, 1990
5-Methoxydurmillone [**231**] 	Stem bark Root bark	Dagne and Bekele, 1989; Dagne *et al.*, 1990
Nordurlettone [**269**] 	Seed	Dagne *et al.*, 1990
Prebarbigerone [**270**] 	Seed	Dagne and Bekele, 1990

Table 4 (continued)

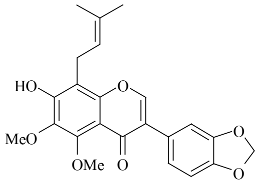
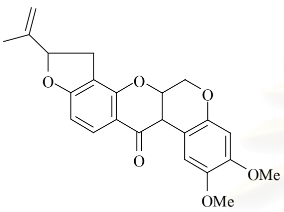
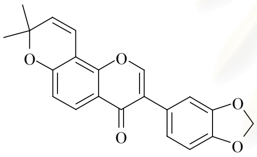
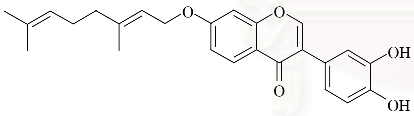
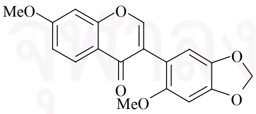
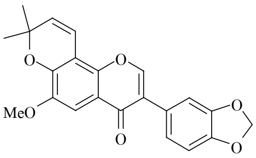
Plant and chemical compound	Plant part	Reference
Pre-5-methoxydurmillone [**271**] 	Root bark	Dagne and Bekele, 1990
Rotenone [**255**] 	Seed	Dagne and Bekele, 1990
M. griffoniana		
Calopogonium isoflavone B [**264**] 	Root bark	Yankep, Fomum, and Dagne, 1997
3',4'-Dihydroxy-7-O-[(E)-3,7-dimethyl-2,6-octadienyl]-isoflavone [**272**] 	Root bark	Yankep *et al*., 1998
7,2'-Dimethoxy-4',5'-methylenedioxy isoflavone [**236**] 	Root bark	Yankep, Fomum, and Dagne, 1997
Durmillone [**238**] 	Root bark	Yankep, Fomum, and Dagne, 1997

Table 4 (continued)

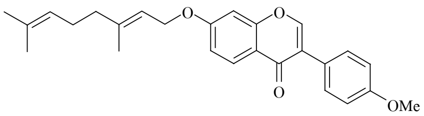
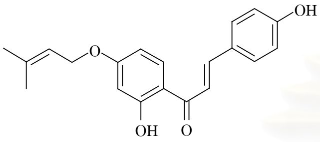
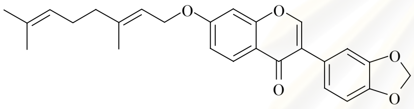
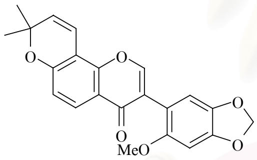
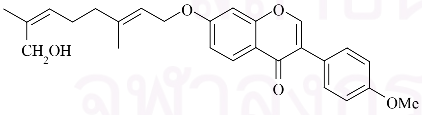
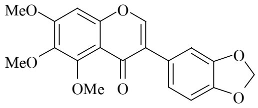
Plant and chemical compound	Plant part	Reference
7-*O*-Geranylformononetin [**265**] 	Root bark	Yankep, Fomum, and Dagne, 1997
4'-*O*-Geranylisoliquiritigenin [**266**] 	Root bark	Yankep, Fomum, and Dagne, 1997
7-*O*-Geranylpseudobaptigenin [**273**] 	Root bark	Yankep, Fomum, and Dagne, 1997
Jamaicin [**246**] 	Root bark	Yankep, Fomum, and Dagne, 1997
4'-Methoxy-7-O[(E)-3-methyl-7-hydroxymethyl-2,6-octadienyl]isoflavone [**274**] 	Root bark	Yankep *et al.*, 1998
Odorantin [**275**] 	Root bark	Yankep, Fomum, and Dagne, 1997

Table 4 (continued)

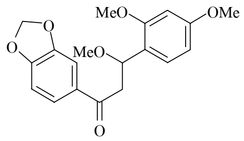
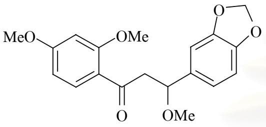
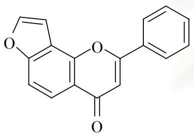
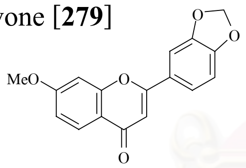
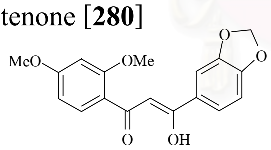
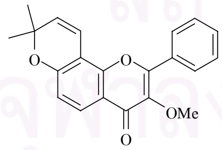
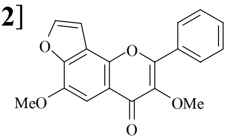
Plant and chemical compound	Plant part	Reference
M. hemsleyana		
Dihydroisomilletenone methyl ether [**276**]  	Stem bark	Mahmoud and Waterman, 1985
Dihydromilletenone methyl ether [**277**]  	Stem bark	Mahmoud and Waterman, 1985
Lanceolatin B [**278**]  	Stem bark	Mahmoud and Waterman, 1985
3',4'-Methylenedioxy-7-methoxy-flavone [**279**]  	Stem bark	Mahmoud and Waterman, 1985
Milletenone [**280**]  	Stem bark	Mahmoud and Waterman, 1985
Pongaflavone [**281**]  	Stem bark	Mahmoud and Waterman, 1985
M. ichthyochtona		
3,6-Dimethoxyfurano[4",5":8,7] flavone [**282**]  	Leaf and branch	Kamperdick *et al*., 1998

Table 4 (continued)

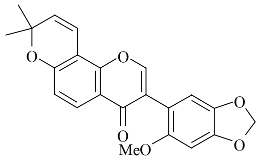
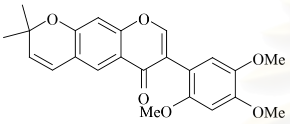
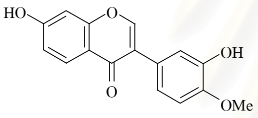
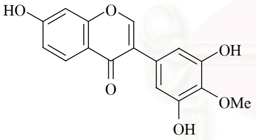
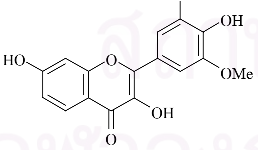
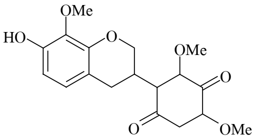
Plant and chemical compound	Plant part	Reference
Jamaicin [**246**] 	Leaf	Kamperdick *et al*., 1998
2',4',5'-Trimethoxy-2",2"-dimethylpyrano[5",6":6,7]isoflavone [**283**] 	Leaf	Kamperdick *et al*., 1998
M. laurentii		
Calycosin [**284**] 	Wood	Kamnaing *et al*., 1999
Gliricidin [**285**] 	Wood	Kamnaing *et al*., 1999
Laurentinol [**286**] 	Wood	Kamnaing *et al*., 1999
Laurentiquinone [**287**] 	Wood	Kamnaing *et al*., 1999

Table 4 (continued)

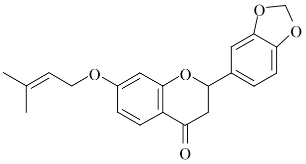
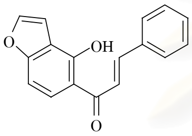
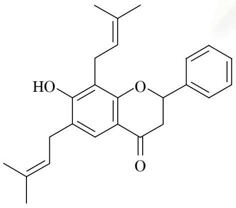
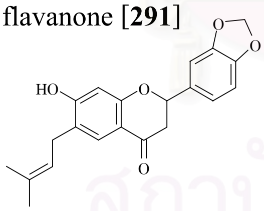
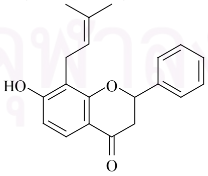
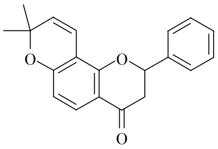
Plant and chemical compound	Plant part	Reference
M. ovalifolia		
7-*O*-(γ,γ-Dimethyl-allyloxy)-3',4'-methylenedioxy flavanone [**288**] 	Seed	Islam, Gupta, and Krishnamurti, 1980
1-(4-Hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one [**289**] 	Root	Asomaning *et al.*, 1999
7-Hydroxy-6,8-diprenylflavanone [**290**] 	Seed	Gupta, and Krishnamurti, 1976b
7-Hydroxy-3',4'-methylenedioxy-6-C-prenyl flavanone [**291**] 	Seed	Islam, Gupta, and Krishnamurti, 1980
7-Hydroxy-8-prenylflavanone [**292**] 	Seed	Gupta, and Krishnamurti, 1976b
Isolonchocarpin [**293**] 	Seed	Krishnamurti and Islam, 1987

Table 4 (continued)

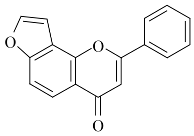
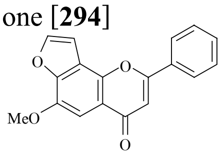
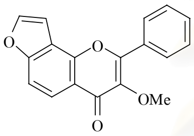
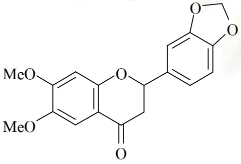
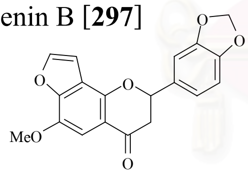
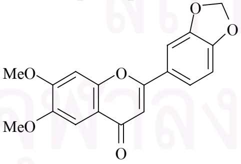
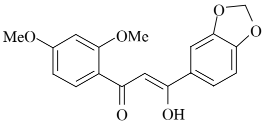
Plant and chemical compound	Plant part	Reference
Lanceolatin B [**278**] 	Leaf Seed	Khan and Zaman, 1974; Gupta, and Krishnamurti, 1976b
Kanjone [**294**] 	Seed	Gupta, and Krishnamurti, 1976b
Karanjin [**295**] 	Seed	Gupta, and Krishnamurti, 1976b
Milletenin A [**296**] 	Leaf	Khan and Zaman, 1974
Milletenin B [**297**] 	Leaf	Khan and Zaman, 1974
Milletenin C [**298**] 	Leaf	Khan and Zaman, 1974
Milletenone [**280**] 	Leaf Root	Khan and Zaman, 1974; Asomaning *et al*., 1999

Table 4 (continued)

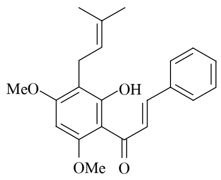
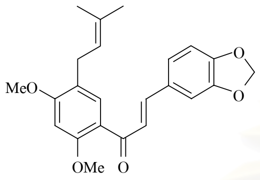
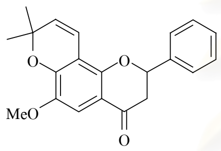
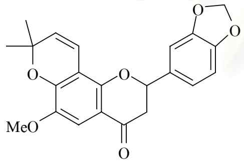
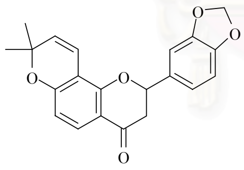
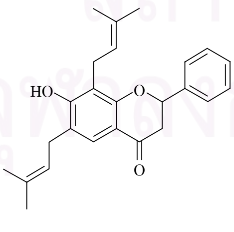
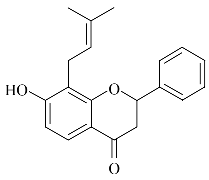
Plant and chemical compound	Plant part	Reference
Ovalichalkone [**299**]  	Seed	Gupta and Krishnamurti, 1977a
Ovalichalkone A [**300**]  	Seed	Gupta and Krishnamurti, 1979a
Ovalichromene [**301**]  	Seed	Gupta and Krishnamurti, 1976c
Ovalichromene A [**302**]  	Seed	Gupta and Krishnamurti, 1976a
Ovalichromene B [**303**]  	Seed	Gupta and Krishnamurti, 1976a
Ovaliflavanone A [**304**]  	Seed	Gupta and Krishnamurti, 1976b
Ovaliflavanone B [**305**]  	Seed	Gupta and Krishnamurti, 1976b

Table 4 (continued)

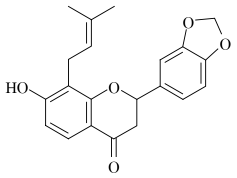
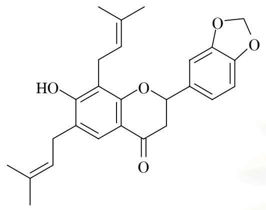
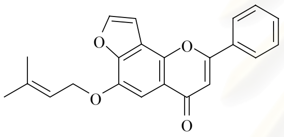
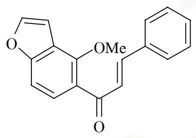
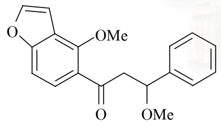
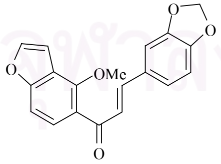
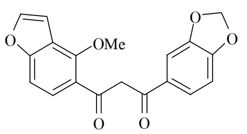
Plant and chemical compound	Plant part	Reference
Ovaliflavanone C [**306**] 	Seed	Islam, Gupta, and Krishnamurti, 1980
Ovaliflavanone D [**307**] 	Seed	Islam, Gupta, and Krishnamurti, 1980
Ovalifolin [**308**] 	Leaf	Khan and Zaman, 1974
Ovalitenin A [**309**] 	Seed	Gupta and Krishnamurti, 1977b
Ovalitenin B [**310**] 	Seed	Gupta and Krishnamurti, 1977b
Ovalitenin C [**311**] 	Seed	Islam, Gupta, and Krishnamurti, 1980
Ovalitenone [**312**] 	Seed Root	Gupta and Krishnamurti, 1977b; Asomaning *et al*., 1999

Table 4 (continued)

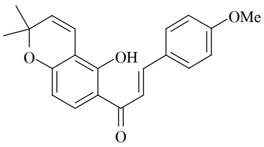
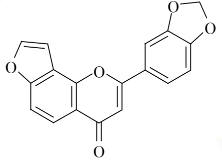
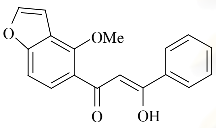
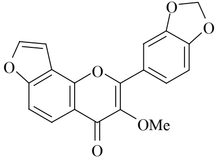
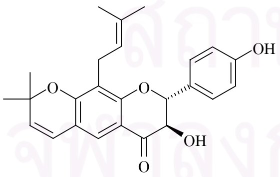
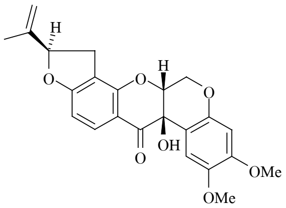
Plant and chemical compound	Plant part	Reference
Pongachalkone-I [**313**] 	Seed	Gupta and Krishnamurti, 1976a
Pongaglabrone [**314**] 	Seed	Gupta, and Krishnamurti, 1976b
Pongamol [**315**] 	Seed Root	Gupta and Krishnamurti, 1976a; Asomaning *et al*., 1999
Pongapin [**316**] 	Seed	Gupta and Krishnamurti, 1976a
M. pachycarpa		
(2*R*,3*R*)-5,4'-Dihydroxy-8-prenyl-6",6"-dimethylpyrano[2",3":7,6]-dihydroflavonol [**317**] 	Aerial part	Singhal *et al*., 1980
cis-12a-Hydroxyrotenone [**260**] 	Root	Singhal *et al*., 1982

Table 4 (continued)

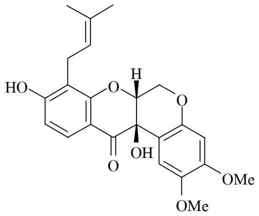
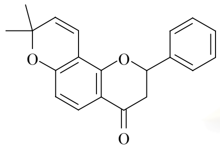
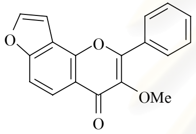
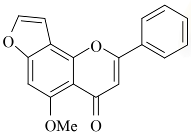
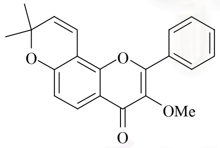
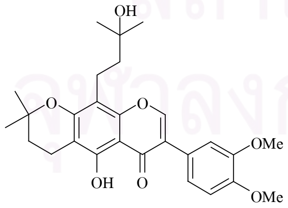
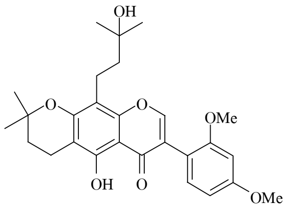
Plant and chemical compound	Plant part	Reference
cis-12a-Hydroxyrot-2-enonic acid [**318**] 	Root	Singhal *et al.*, 1982
(-)-Isolonchocarpin [**293**] 	Root	Shao *et al.*, 2001a
Karanjin [**295**] 	Root	Chen *et al.*, 1999
5-Methoxykaranjin [**319**] 	Root	Lu *et al.*, 1999
3-Methoxypyrano(5",6":7,8)-flavone (Pongaflavone) [**281**] 	Root	Shao *et al.*, 2001a
Millettia isoflavone 7A [**320**] 	Leaf	Singhal *et al.*, 1981
Millettia isoflavone 7B [**321**] 	Leaf	Singhal *et al.*, 1981

Table 4 (continued)

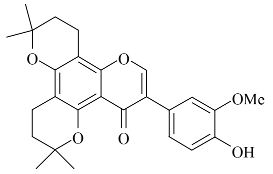
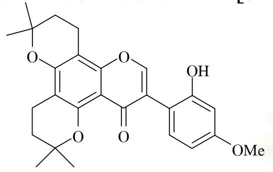
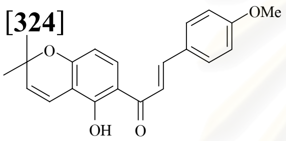
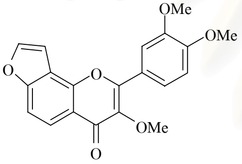
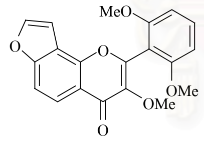
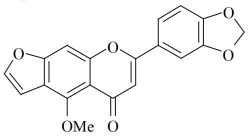
Plant and chemical compound	Plant part	Reference
Millettia isoflavone 10B [**322**]  	Leaf	Singhal *et al*., 1981
Millettia isoflavone 11A [**323**]  	Leaf	Singhal *et al*., 1981
Millettia pachycarpa pyranochalcone 3A [**324**]  	Seed	Singhal *et al*., 1983
Pachycarin A [**325**]  	Root	Chen *et al*., 1999
Pachycarin B [**326**]  	Root	Lu *et al*., 1999
Pachycarin C [**327**]	Root	Shao *et al*., 2001b
Pachycarin D [**328**]	Root	Shao *et al*., 2001b
Pachycarin E [**329**]	Root	Shao *et al*., 2001b
Pinnatin [**330**]  	Root	Shao *et al*., 2001a

Table 4 (continued)

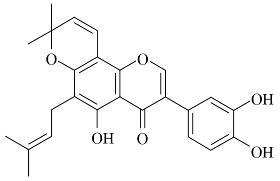
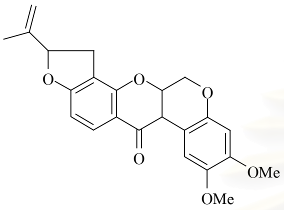
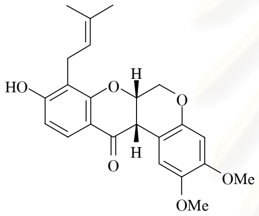
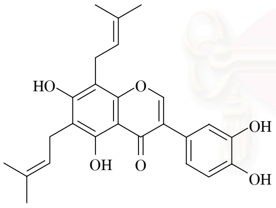
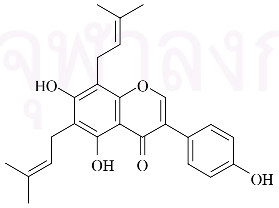
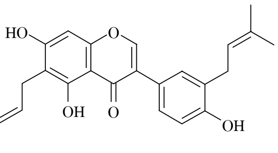
Plant and chemical compound	Plant part	Reference
Pomiferin [**331**] 	Seed	Singhal *et al*., 1981
Rotenone [**255**] 	Root	Singhal *et al*., 1982
Rot-2'-enonic acid [**332**] 	Root	Singhal *et al*., 1982
5,7,3',4'-Tetrahydroxy-6,8-diprenyl-isoflavone [**333**] 	Aerial part Seed	Singhal *et al*., 1980; Singhal *et al*., 1981
5,7,4'-Trihydroxy-6,8-diprenyliso-flavone [**334**] 	Aerial part Seed	Singhal *et al*., 1980; Singhal *et al*., 1981
5,7,4'-Trihydroxy-6,3'-diprenyliso-flavone [**335**] 	Aerial part	Singhal *et al*., 1980

Table 4 (continued)

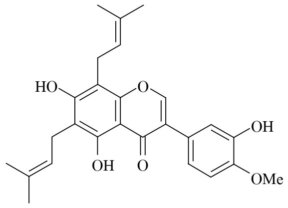
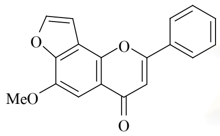
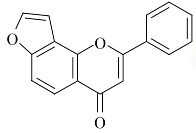
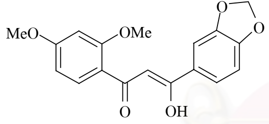
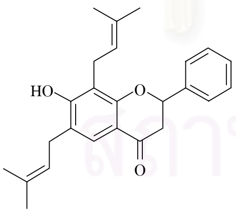
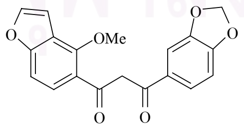
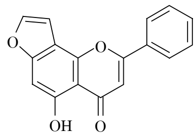
Plant and chemical compound	Plant part	Reference
3',5,7-Trihydroxy-4'-methoxy-6,8-diprenylisoflavone [**336**]  	Seed	Singhal *et al*., 1981
M. peguensis		
Kanjone [**294**]  	Stem bark	Ganapaty *et al*., 1998
Lanceolatin B [**278**]  	Stem bark	Ganapaty *et al*., 1998
Milletenone [**280**]  	Stem bark	Ganapaty *et al*., 1998
Ovaliflavanone A [**304**]  	Stem bark	Ganapaty *et al*., 1998
Ovalitenone [**312**]  	Stem bark	Ganapaty *et al*., 1998
Pongaglabol [**337**]  	Leaf	Ganapaty *et al*., 1998

Table 4 (continued)

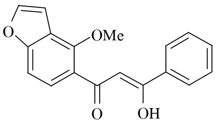
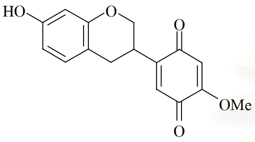
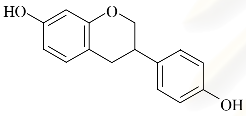
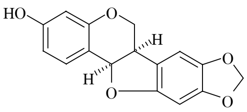
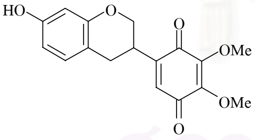
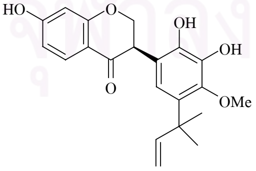
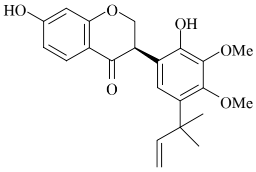
Plant and chemical compound	Plant part	Reference
Pongamol [**315**] 	Leaf	Ganapaty *et al*., 1998
M. pendura		
Claussequinone [**338**] 	Heart wood	Hayashi *et al*., 1978
Equol [**339**] 	Heart wood	Hayashi *et al*., 1978
(-)-Maackianin [**340**] 	Heart wood	Hayashi *et al*., 1978
Pendulone [**341**] 	Heart wood	Hayashi *et al*., 1978
M. pervilleana		
3'-*O*-Demethylpervilleanone [**342**] 	Root bark	Galeffi *et al*., 1997
Pervilleanone [**343**] 	Root bark	Galeffi *et al*., 1997

Table 4 (continued)

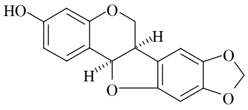
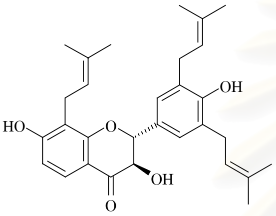
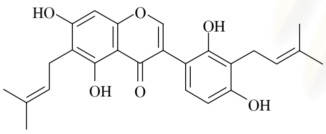
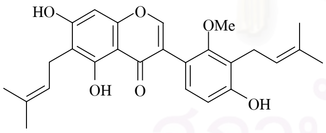
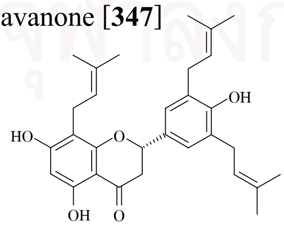
Plant and chemical compound	Plant part	Reference
M. pulchra		
(-)-Maackianin [**340**] 	Aerial part	Baruah *et al.*, 1984
(2*R*,3*R*)-7,4'-Dihydroxy-8,3',5'-triprenyldihydroflavanol [**344**] 	Aerial part	Baruah *et al.*, 1984
5,7,2',4'-Tetrahydroxy-6,3'-diprenylisoflavone [**345**] 	Aerial part	Baruah *et al.*, 1984
5,7,4'-Trihydroxy-2'-methoxy-6,3'-diprenylisoflavone [**346**] 	Aerial part	Baruah *et al.*, 1984
(2*S*)-5,7,4'-Trihydroxy-8,3',5'-triprenylflavanone [**347**] 	Aerial part	Baruah *et al.*, 1984

Table 4 (continued)

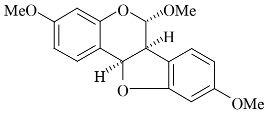
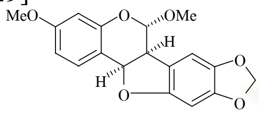
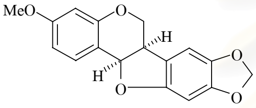
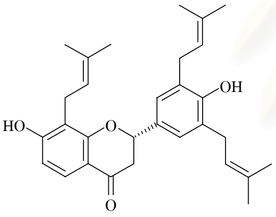
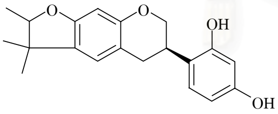
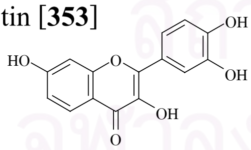
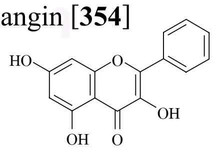
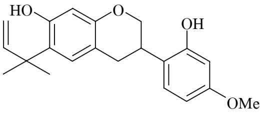
Plant and chemical compound	Plant part	Reference
(6*S*,6a*S*,11a*R*)-6α-methoxyhomopterocarpin [**348**] 	Aerial part	Baruah *et al*., 1984
(6*S*,6a*S*,11a*R*)-6α-methoxypterocarpin [**349**] 	Aerial part	Baruah *et al*., 1984
(-)-Pterocarpin [**350**] 	Aerial part	Baruah *et al*., 1984
(-)-Sophoranone [**351**] 	Aerial part	Baruah *et al*., 1984
M. racemosa		
3*R*(+)-Cyclomillinol [**352**] 	Stem	Kumar, Krupadanam, and Srimannarayana, 1989
Fisetin [**353**] 	Leaf	Ganapaty, Pushpalatha, and Naidu, 1999
Galangin [**354**] 	Leaf	Ganapaty, Pushpalatha, and Naidu, 1999
3*R*(-)-Isomillinol-B [**355**] 	Stem	Rao and Krupadanam, 1994

Table 4 (continued)

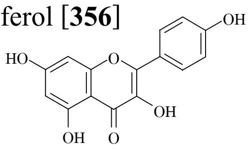
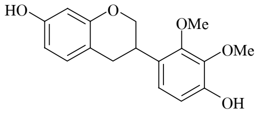
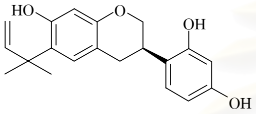
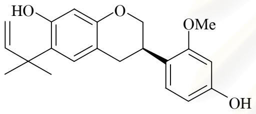
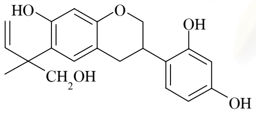
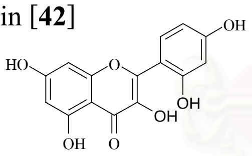
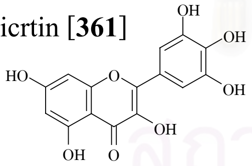
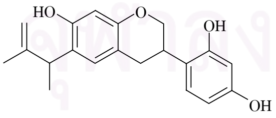
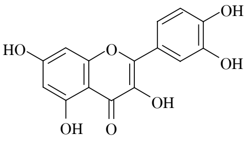
Plant and chemical compound	Plant part	Reference
Kaemferol [**356**] 	Leaf	Ganapaty, Pushpalatha, and Naidu, 1999
3*R*(-)-Laxifloran [**357**] 	Stem	Rao and Krupadanam, 1994
3*R*(+)-Millinol [**358**] 	Stem	Kumar, Krupadanam, and Srimannrayana, 1989
3*R*(+)-Millinol-B [**359**] 	Stem	Kumar, Krupadanam, and Srimannrayana, 1989
Millinolol [**360**] 	Stem	Rao, Prashant, and Krupadanam, 1996
Morin [**42**] 	Leaf	Ganapaty, Pushpalatha, and Naidu, 1999
Myricrtin [**361**] 	Leaf	Ganapaty, Pushpalatha, and Naidu, 1999
Neomillinol [**362**] 	Stem	Rao, Prashant, and Krupadanam, 1996
Quercetin [**363**] 	Leaf	Ganapaty, Pushpalatha, and Naidu, 1999

Table 4 (continued)

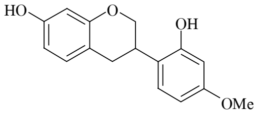
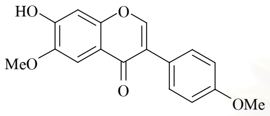
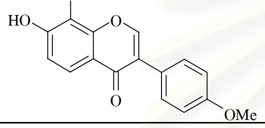
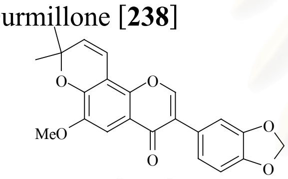
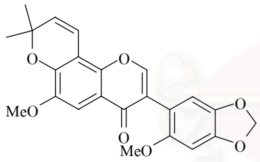
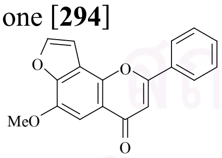
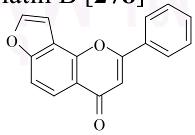
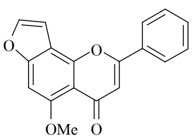
Plant and chemical compound	Plant part	Reference
3*R*(-)-Vestitol [**364**] 	Stem	Rao and Krupadanam, 1994
M. reticulata		
Afrormosin [**365**] 	Stem	Chen *et al*., 1983
7-Hydroxy-4',8-dimethoxyisoflavone [**366**] 	Stem	Chen *et al*., 1983
M. rubiginosa		
Durmillone [**238**] 	Root	Desai *et al*., 1977
Ichthynone [**261**] 	Root	Desai *et al*., 1977
M. sanagana		
Kanjone [**294**] 	Root bark	Mbafor *et al*., 1995
Lanceolatin B [**278**] 	Root bark	Mbafor *et al*., 1995
5-Methoxy furano [7,8:4",5"] flavone [**319**] 	Root bark	Mbafor *et al*., 1995

Table 4 (continued)

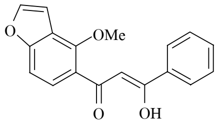
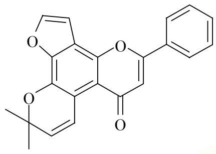
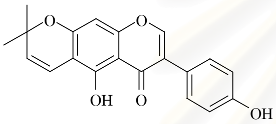
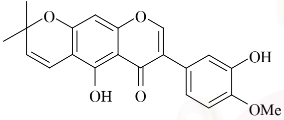
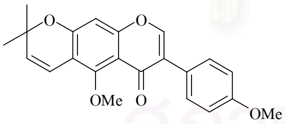
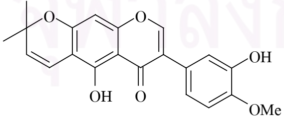
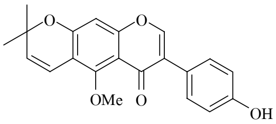
Plant and chemical compound	Plant part	Reference
Pongamol [**315**] 	Root bark	Mbafor *et al*., 1995
Sanaganone [**367**] 	Root bark	Mbafor *et al*., 1995
M. thonningii		
Alpinum isoflavone [**368**] 	Seed Root Entire plant	Olivares *et al*., 1982; Asomaning *et al*., 1999 Khalid *et al*., 1986
3',5-Dihydroxy-4'-methoxy-2",2"-dimethylpyrano-(5",6":6,7)-isoflavone [**369**] 	Seed	Olivares *et al*., 1982
O,*O*-Dimethylalpinumisoflavone [**370**] 	Root bark Seed Root Pod	Asomaning *et al*., 1995; Olivares *et al*., 1982; Asomaning *et al*., 1999
3'-Hydroxy-4'-*O*-methylalpinumisoflavone [**371**] 	Pod	Asomaning *et al*., 1999
5-*O*-Methylalpinumisoflavone [**372**] 	Root	Asomaning *et al*., 1999

Table 4 (continued)

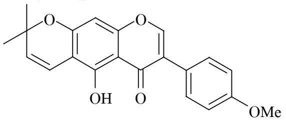
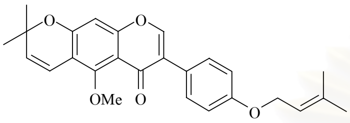
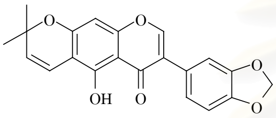
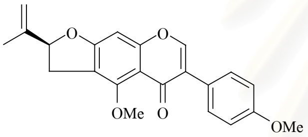
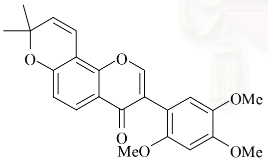
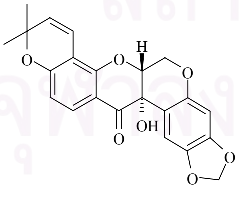
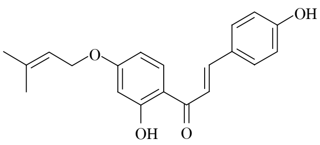
Plant and chemical compound	Plant part	Reference
4'-Methylalpinumisoflavone [**373**] 	Seed	Olivares *et al*., 1982
5-*O*-Methyl-4'-*O*-(3-methyl-2-butenyl)alpinumisoflavone [**374**] 	Root bark Root	Asomaning *et al*., 1995; Asomaning *et al*., 1999
Robustone [**375**] 	Seed Root	Khalid and Waterman, 1983; Asomaning *et al*., 1999
Thonninginisoflavone [**376**] 	Root bark	Asomaning *et al*., 1995
M. usaramensis		
subsp. *usaramensis*		
Barbigerone [**257**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
(+)-12a-Epimillettosin [**377**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
4'-*O*-Geranylisoliquiritigenin [**266**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998

Table 4 (continued)

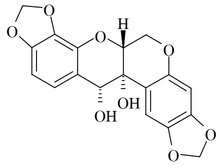
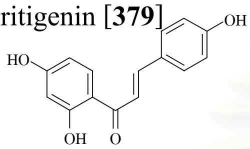
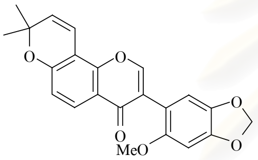
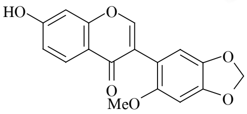
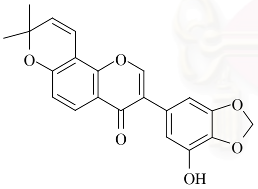
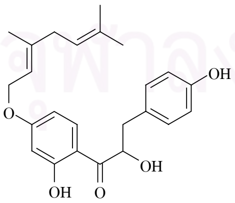
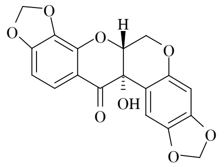
Plant and chemical compound	Plant part	Reference
(+)-12α-Hydroxy-12-dihydrousara-rotenoid A [**378**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
Isoliquiritigenin [**379**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
Jamaicin [**246**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
Maximaisoflavone G [**380**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
Norisojamaicin [**381**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
α,4,2'-Trihydroxy-4'-*O*-geranyldihy-drochalcone [**382**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
(+)-Usararotenoid-A [**383**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998

Table 4 (continued)

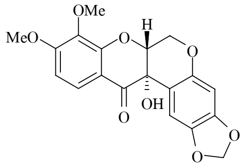
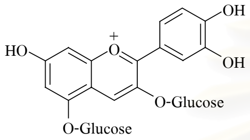
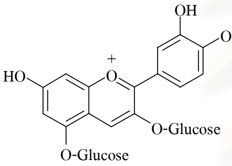
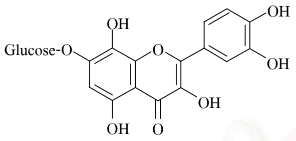
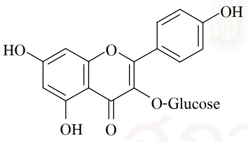
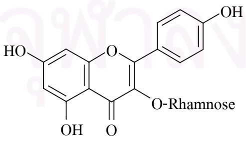
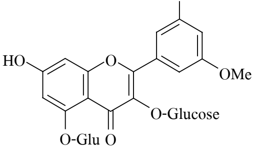
Plant and chemical compound	Plant part	Reference
(+)-Usararotenoid-B [**384**] 	Stem bark	Yenesew, Midiwo, and Waterman, 1998
M. zechiana		
Cyanin [**385**] 	Flower	Ogbeide and Parvez, 1992
Cyanidin-3,5-di-*O*-glucoside [**386**] 	Flower	Parvez and Ogbeide, 1990
8-Hydroxyquercetin-7-*O*-glucoside [**387**] 	Flower	Parvez and Ogbeide, 1990
Kaempferol-3-*O*-glucoside [**388**] 	Flower	Parvez and Ogbeide, 1990
Kaemferol-3-*O*-rhamnoside [**389**] 	Flower	Ogbeide and Parvez, 1992
Malvidin-3,5-di-*O*-glucoside [**390**] 	Flower	Parvez and Ogbeide, 1990; Ogbeide and Parvez, 1992

Table 4 (continued)

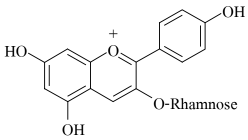
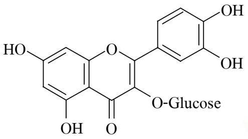
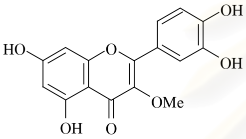
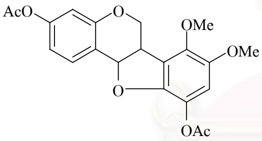
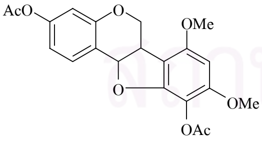
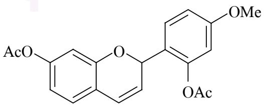
Plant and chemical compound	Plant part	Reference
Pelargonidin-3-*O*-rhamnoside [**391**] 	Flower	Parvez and Ogbeide, 1990
Quercetin-3-*O*-glucoside [**392**] 	Flower	Parvez and Ogbeide, 1990
Quercetin-3-methylether [**393**] 	Flower	Ogbeide and Parvez, 1992
***Millettia* sp.**		
3,10-Diacetoxy-7,8-dimethoxypterocarpan [**394**] 	Heartwood	Mitsunaga, Kondo, and Imamura, 1987a
3,10-Diacetoxy-7,9-dimethoxypterocarpan [**395**] 	Heartwood	Mitsunaga, Kondo, and Imamura, 1987a
2',7-Diacetoxy-4'-methoxyflav-3-ene [**396**] 	Heartwood	Mitsunaga, Kondo, and Imamura, 1987a

Table 4 (continued)

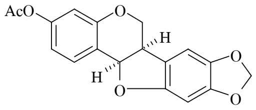
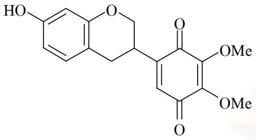
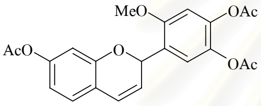
Plant and chemical compound	Plant part	Reference
Maackiain acetate [**397**] 	Heartwood	Mitsunaga, Kondo, and Imamura, 1987a
Pendulone [**341**] 	Heartwood	Mitsunaga, Kondo, and Imamura, 1987a
3',4',7-Triacetoxy-6'-methoxyflav-3-ene [**398**] 	Heartwood	Mitsunaga, Kondo, and Imamura, 1987a; Mitsunaga, Kondo, and Imamura, 1987b

Table 5 Distribution of miscellaneous compounds in *Millettia*.

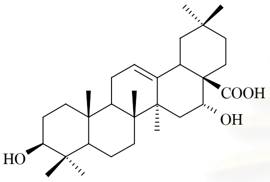
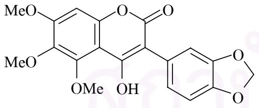
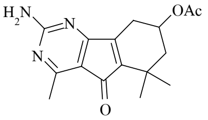
Plant and chemical compound	Category	Plant part	Reference
M. duchesnei			
Arabinose [**399**]	Carbohydrate	Root bark	Kapundu, Nzundu, and Delaude, 1984
Echinocystic acid [**400**]  	Triterpenoid	Root bark	Kapundu, Nzundu, and Delaude, 1984
Fucose [**401**]	Carbohydrate	Root bark	Kapundu, Nzundu, and Delaude, 1984
Glucose [**402**]	Carbohydrate	Root bark	Kapundu, Nzundu, and Delaude, 1984
Rhamnose [**403**]	Carbohydrate	Root bark	Kapundu, Nzundu, and Delaude, 1984
M. griffoniana			
4-Hydroxy-5,6,7-trimethoxy-3-(3',4'-methylenedioxy)phenylcoumarin [**404**]  	Coumarin	Root bark	Yankep *et al*., 1998
M. laurentii			
O-Acetylmillaurine [**405**]  	Alkaloid	Seed	Ngamga *et al*., 1993

Table 5 (continued)

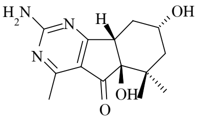
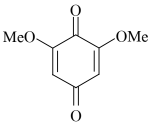
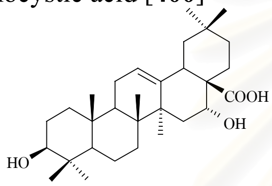
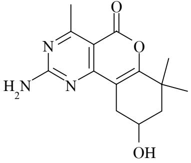
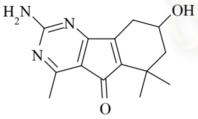
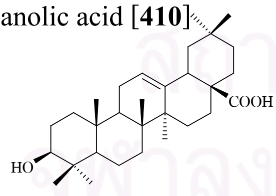
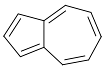
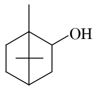
Plant and chemical compound	Category	Plant part	Reference
5a,9a-Dihydro-5a-hydroxymillaurine [**406**] 	Alkaloid	Seed	Ngamga, Free, and Fomum, 1994
2,6-Dimethoxy-*p*-benzoquinone [**407**] 	Quinone	Wood	Schmalle and Jarchow, 1977; Hausen, 1978
Echinocystic acid [**400**] 	Triterpenoid	Entire plant	Kapundu *et al*., 1978
Millettonine [**408**] 	Alkaloid	Stem bark	Kamnaing *et al*., 1994
Millaurine [**409**] 	Alkaloid	Seed	Ngamga *et al*., 1993
Oleanolic acid [**410**] 	Triterpenoid	Entire plant	Kapundu *et al*., 1978
M. ovalifolia			
Azulene [**411**] 	MonoterpeNoid	Leaf	Nigam *et al*., 1982
α-Boneol [**412**] 	MonoterpeNoid	Leaf	Nigam *et al*., 1982

Table 5 (continued)

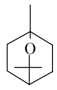
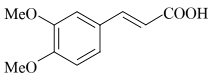
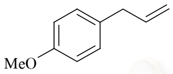
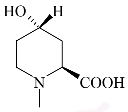
Plant and chemical compound	Category	Plant part	Reference
1,8-Cineol [**413**] 	Monoterpenoid	Leaf	Nigam *et al.*, 1982
3,4-Dimethoxycinnamic acid [**414**]  	Phenylpropanoid	Seed	Krishnamurti and Islam, 1987
Heptacosanol [**415**] $H_3C(CH_2)_{25}CH_2OH$	Alkane	Seed	Krishnamurti and Islam, 1987
Linelyl acetate [**416**]	MonoterpeNoid	Leaf	Nigam *et al.*, 1982
Methyl chavicol [**417**]  	Phenylpropanoid	Leaf	Nigam *et al.*, 1982
Ovalin [**418**]  	Alkaloid	Seed	Gupta and Krishnamurti, 1979b
19-Oxo-5α-carda-14,20(22)-dienolide-3-*O*-β-D-glucopyranoside [**419**]	Cardenolide	Root	Bose and Chakraborty, 2000

Table 5 (continued)

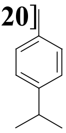
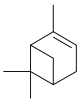
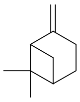
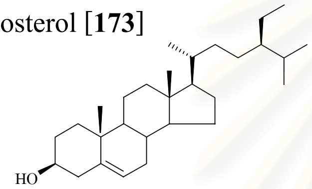
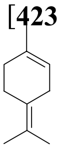
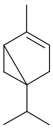
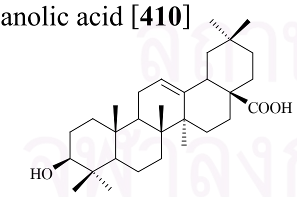
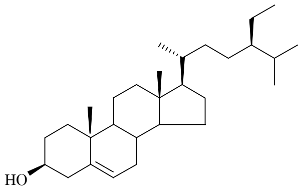
Plant and chemical compound	Category	Plant part	Reference
Pi-cymene [**420**] 	Monoterpe-Noid	Leaf	Nigam *et al*., 1982
α-Pinene [**421**] 	Monoterpe-Noid	Leaf	Nigam *et al*., 1982
β-Pinene [**422**] 	Monoterpe-Noid	Leaf	Nigam *et al*., 1982
β-Sitosterol [**173**] 	Steroid	Seed	Gupta and Krishnamurti, 1976b
α-Terpinolene [**423**] 	Monoterpe-Noid	Leaf	Nigam *et al*., 1982
α-Thujene [**424**] 	Monoterpe-Noid	Leaf	Nigam *et al*., 1982
M. pachycarpa			
Oleanolic acid [**410**] 	Triterpenoid	Root	Chen *et al*., 1999
β-Sitosterol [**173**] 	Steroid	Root	Chen *et al*., 1999

Table 5 (continued)

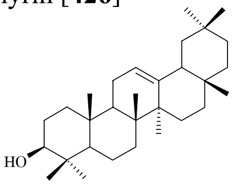
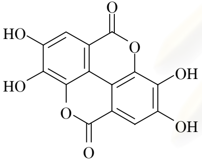
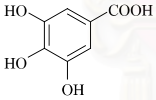
Plant and chemical compound	Category	Plant part	Reference
M. peguensis			
Canavanine [**425**]	Proteid	Seed	Rao, 1983
M. pendura			
β-Amyrin [**426**] 	Triterpenoid	Stem bark	Rathore, Nagar, and Gupta, 1983
Daucosterol [**427**]	Steroid	Seed	Rathore, Nagar, and Gupta, 1983
Ellagic acid [**428**] 	Coumarin	Stem bark	Rathore, Nagar, and Gupta, 1983
Galactose [**429**]	Carbohydrate	Stem bark	Rathore, Nagar, and Gupta, 1983
Gallic acid [**430**] 	Benzenoid	Stem bark	Rathore, Nagar, and Gupta, 1983
β-Methylgalactoside [**431**]	Carbohydrate	Stem bark	Rathore, Nagar, and Gupta, 1983
Octacosan-1-ol [**432**] $H_3C(CH_2)_{26}CH_2OH$	Alkane	Stem bark	Rathore, Nagar, and Gupta, 1983
Rhamnose [**403**]	Carbohydrate	Stem bark	Rathore, Nagar, and Gupta, 1983

Table 5 (continued)

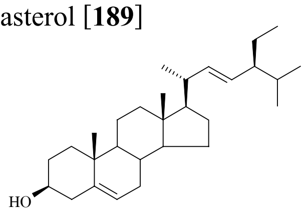
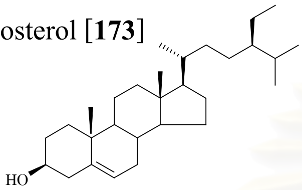
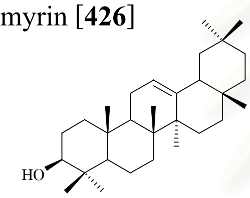
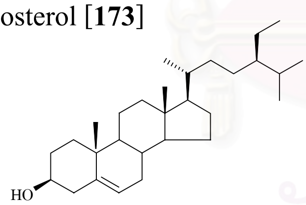
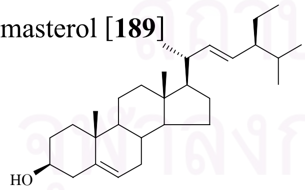
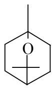
Plant and chemical compound	Category	Plant part	Reference
Stigmasterol [**189**] 	Steroid	Stem bark	Rathore, Nagar, and Gupta, 1983
β-Sitosterol [**173**] 	Steroid	Stem bark	Rathore, Nagar, and Gupta, 1983
M. racemosa			
β-Amyrin [**426**] 	Triterpenoid	Stem	Rao and Krupadanam, 1994
Behenic acid [**433**] $H_3C(CH_2)_{20}COOH$	Lipid	Stem	Rao and Krupadanam, 1994
β-Sitosterol [**173**] 	Steroid	Stem Root	Rao and Krupadanam, 1994
Stigmasterol [**189**] 	Steroid	Root	Desai *et al*., 1977
M. reticulata			
1,8-Cineol [**413**] 	Monoterpenoid	Flower	Gong and Wu, 1998

Table 5 (continued)

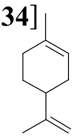
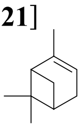
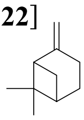
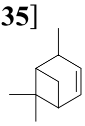
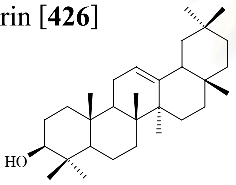
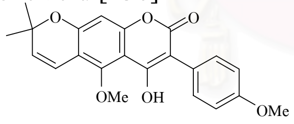
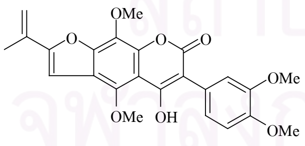
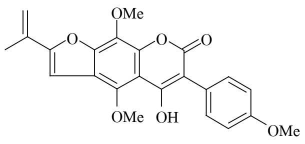
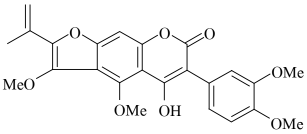
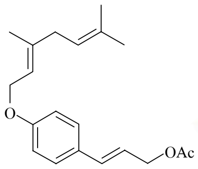
Plant and chemical compound	Category	Plant part	Reference
Limonene [**434**] 	Monoterpe-noid	Flower	Gong and Wu, 1998
α-Pinene [**421**] 	Monoterpe-noid	Flower	Gong and Wu, 1998
β-Pinene [**422**] 	Monoterpe-noid	Flower	Gong and Wu, 1998
δ-Pinene [**435**] 	Monoterpe-noid	Flower	Gong and Wu, 1998
M. thonningii			
β-Amyrin [**426**] 	Steroid	Root bark	Asomaning *et al*., 1995
Robustic acid [**436**] 	Coumarin	Seed Entire plant	Olivares *et al*., 1982 Khalid *et al*., 1986
Thonningine-A [**437**] 	Coumarin	Seed	Khalid and Waterman, 1983
Thonningine-B [**438**] 	Coumarin	Seed	Khalid and Waterman, 1983

Table 5 (continued)

Plant and chemical compound	Category	Plant part	Reference
Thonningine-C [**439**] 	Coumarin	Seed	Saxena *et al.*, 1987
M. usaramensis **subsp. *usaramensis*** 4-*O*-Geranylcinnamyl acetate [**440**] 	Phenylpro -panoid	Stem bark	Yenesew, Miwido, and Waterman, 1998

3. Traditional Uses and Biological Activities of *Artocarpus* Constituents.

Artocarpus plants have been used in traditional medicine in many countries with several purposes. In West Indies, many parts of *A. altilis* or bread-fruit tree were used: ground fruit rind tea for oliguria, yellow leaf tea for hypertension and diabetes, latex cataplasm for pain, crushed leaves for pain, crushed leaves bound to head for headache and boil leaves for tea for high blood pressure (Ayensu, 1981). In Indonesia, *A. altilis* the bark of the seedless form is one of the constituents of medicine administered postpartum. The ashes of the leaves, with coconut oil and *Curcuma*, are applied to a skin disease with creeps like herpes. A poultice of the roasted and crushed leaves with water is applied to enlarged spleen. The heated flowers, after cooling, are rubbed on the gums to ease toothache. The fruit meat is used to treat cough, the root bark to treat diarrhea and dysentery and the seeds as an aphrodisiac. In the Philippines, a decoction of the bark is employed as a vulnerary and also to treat stomachache. In New Guinea, the latex is a medicine taken to cure dysentery (Perry, 1980). The stems and roots have been used for the treatment of liver cirrhosis and hypotension in Taiwan (Chen *et al*., 1993).

Concerning *A. hetterophyllus* or Jackfruit tree, the fruit is edible. The sap is used to treat ulcers and abscess in Burma, China and the Philippines, and bark as poultices for the same in Malay and Peninsular. The roots are used to treat diarrhea, and in a compound extract to treat fever in Burma. In Indo-China, the wood is used as a sedative to treat convulsions. The boiled leaves are given to both animals and women to activate the secretion of milk and the sap is antisyphilitic and vermifuge (Perry, 1980).

Traditional uses of the other *Artocarpus* species have been recorded. The sap from the wounded bark of *A. dadah* is employed to clean foul leg-wounds in Indonesia. In the same country, a strip of *A. elasticus* pounded is applied as a bandage to treat lumbago. Its leaves mixed with rice are ingested for the treatment of tuberculosis, and the latex is used to treat dysentery. In Indo-China, the latex of *A. rigidus* is applied to wounds of domestic animals. The *A. lakoocha* roots are tonic and deobstruent and its leaves are used for treating dropsy. The boiled bark of *A. ovatus* is used for treating stomachache. The fresh leaves of *A. rubrovenious* is administered for fevers. In Burma, the juice and seeds of *A. lakoocha* are purgative and the bark is astringent (Perry, 1980).

A famous Thai traditional medicine from *A. lakoocha* known as “Puag-Haad” has been used as an anthelmintic and antipyretic. Puag-Haad is an aqueous extract of the heartwood of *A. lakoocha* and its activities come from 2,4,3',5'-tetrahydroxystilbene. (Farnworth and Bunyapraphatsara, 1992; Poopyruchpong *et al*., 1978).

The anti-inflammatory activities of *Artocarpus* flavonoids have been studied by testing these compounds for their inhibitory actions on arachidonate 5-lipooxygenase. Several compounds such as artonin E [**13**] from the bark (Reddy *et al*., 1991) and Artocarpus chalcone (AC-5-1) [**7**] from the dried flowers of *A. communis* (Koshihara *et al*., 1988; Nomura *et al*., 1998) showed potent inhibition on arachidonate 5-lipoxygenase.

According to a review by Nomura and co-workers in 1998, the *Artocarpus* flavonoids have been studied for many biological activities. Morusin [**43**] has been found to be an anti-tumor in a two-stage carcinogenesis experiment with teleocidin, and several *Artocarpus* flavonoids act as anti-tumor promotors against the okadaic acid type promotion. About cytotoxic activities, *Artocarpus* flavonoids such as artonin E [**13**], heterophyllin [**98**] and cycloheterophyllin [**93**] showed cytotoxic activities against the cancer cells mouse-L1210 and colon 38. Artomunoxanthotrione epoxide [**16**], cyclocommunol [**34**], cyclomulberrin [**36**] and cyclocommunin [**33**] showed in *vitro* cytotoxic effects against human hepatoma PLC/PRF/5 and KB cells. The prenyl flavonoids isolated from Formosan *A. communis* and *A. heterophyllus* showed inhibition of arachidonic acid-induced platelet aggregation. Furthermore, the extract of *A. heterophyllus* showed intensive antibacterial activities against cariogenic bacteria and also inhibited the growth of *Streptococci* on plaque forming. In 1998, Shimizu *et al*. reported that the compounds of heartwood of *A. incisus* showed potent inhibitory activity for tyrosinase enzyme. Norartocarpetin [**45**] and resveratrol [**177**] from *A. gomezianus* exhibited potent tyrosinase inhibitory activity (Likhitwitayawuid, Sritularak and De-Eknamkul, 2000).

4. Traditional Uses and Biological Activities of *Millettia* Constituents.

Plants of the genus *Millettia* have been used medicinally in several countries. In Cameroon, various parts of *Millettia* plants are used as a cure for intestinal parasites and cholic in children (Mbafor *et al*., 1995). In East Africa, the roots of *M. usaramensis* are supposedly used as a remedy for snake bite (Yenesew, Midiwo and Waterman, 1998). The bark pulp of *M. zechiana*, with sea water and Guinea grains diluted with warm water, is used as a gargle for rhinopharyngal and bronchial troubles and the purple leaves are rubbed on painful parts in Guinea (Parvez and Ogbeide, 1990). In East Africa, the roots of *M. eriocalyx* is used as application to skin eruption (Watt and Breyer-Brandwijk, 1962). *M. thonningii* is used in Ghana as an anthelmintic and as a purgative agent (Perrett *et al*., 1995) and this plant is also used as a laxative, a blood purifier, a dewormer, an analgesic and for the treatment of diarrhea (Asomaning *et al*., 1995).

The seed and the other parts of *Millettia* species have been shown to have insecticidal, piscidal and molluscicidal activities. In Ethiopia, the seeds of *M. ferruginea* are used as fish poisoning (Dagne and Bekele, 1990). In Cameroon, various parts of *Millettia* plants are used as insecticides and piscicide, as agent for the destruction of worms and snails (Ngamga *et al*., 1993).

A number of biological investigations of *Millettia* species have been reported. Extracts and aqueous suspension of finely ground seeds of *M. pachycarpa* are reported to possess considerable insecticidal activity when used in sprays against a variety of insects, e.g. houseflies, bean aphids, pentatomids, leaf beetles and cabbage worms. They act both as stomach and contact poisons and are also ovicidal (Singhal *et al*., 1983). The aqueous extract from *M. pachycarpa* showed very strong inhibitory effects on murine retroviral reverse transcriptase and human DNA polymerase (Ono *et al*., 1989). The juice from the leaves of *M. thonningii* can kill the *Bilinus* snail, the vector for schistosomiasis (Asomaning *et al*., 1999) and a chloroform extract of the seeds from this plant also have been found to be effective in preventing schistosomal infections (Perrett *et al*., 1995). Furthermore, a dichloromethane extract of *M. thonningii* seeds is extremely efficient at disrupting embryonic development of Biomphalaria glabrata eggs *in situ* within masses and killing such embryos (Tang, Whitfield and Perrett, 1995).

Some flavonoids from *M. racemosa*, such as neomillinol [**362**] and millinolol [**360**] showed moderate bactericidal activity against *Staphylococcus aureus* (Rao, Prashant and Krupadanam, 1996). 3*R*(-)-Isomillinol-B [**355**] and 3*R*(-)-vestitol [**364**] showed highly toxic to *Staphylococcus aureus* and *Escherichia coli* (Rao and Krupadanam, 1994) and insecticidal activity against *Spodoptera litura* (Kumar, Krupadanam and Srimannarayana, 1989). Pongamol [**315**] from *M. ovalifolia* showed potent inhibition in the growth of fungus, *Helminthosporium oryzae* (Saxena *et al*., 1987). The flavonoids from *M. thonningii*, such as alpinum isoflavone [**368**] and robustic acid [**436**] have been shown to be effective in killing the egg masses of *B. glabrata* and stopping ciliary beating on the gill of the mussel *Mytilus* (Perrett and Whitfield, 1995) and these compounds also displayed moderate antimalarial activity against *Plasmodium falciparum* (Khalid *et al*., 1986). Moreover, *O*,*O*-dimethylalpinumisoflavone [**370**] from *M. thonningii* were active in the brine shrimp lethality bioassay (Asomaning *et al*., 1995).

CHAPTER III

EXPERIMENTAL

1. Sources of Plant Materials

The roots of *Artocarpus gomezianus* Wall ex Tréc. were collected from Trang province, Thailand in May 1998. The plant was identified by Dr. Thawatchai Santisuk of the Botanical Section, Royal Forest Department, Ministry of Agriculture and Co-operatives, Bangkok, Thailand. A voucher specimen, KL-052541, is on deposit at the Department of Pharmacognosy, Faculty of Pharmaceutical Sciences, Chulalongkorn University.

The stem bark and the roots of *Millettia erythrocalyx* Gagnep. were collected from Tayang district, Petchaburi Province, Thailand, in April 1999, and April 2000, respectively. Authentication was performed by comparison with herbarium specimens at the Royal Forest Department, Ministry of Agriculture and Co-operatives, and voucher specimens (KL-032542 and KL-042543) are on deposit at the Department of Pharmacognosy, Faculty of Pharmaceutical Sciences, Chulalongkorn University.

2. General Techniques

2.1 Analytical Thin-Layer Chromatography (TLC)

Technique : One dimension, ascending

Adsorbent : Silica gel 60 F_{254} (E. Merck) precoated plate

Layer thickness : 0.2 mm

Distance : 6 cm

Temperature : Laboratory temperature (30-35 $^{\circ}$C)

Detection : 1.Ultraviolet light (254 and 365 nm)

2. Anisaldehyde and heating at 105 $^{\circ}$C for 10 min.

2.2 Preparative Thin-Layer Chromatography (PLC)

Technique : One dimension, ascending

Adsorbent : Silica gel 60 F_{254} (E. Merck) precoated plate

Layer thickness : 1 mm

Distance : 15 cm

Temperature : Laboratory temperature (30-35 $^{\circ}$C)

Detection : Ultraviolet light (254 and 365 nm)

2.3 Column Chromatography

2.3.1 Vacuum Liquid Column Chromatography

Adsorbent : Silica gel 60 (No. 7734) particle size 0.063-0.200 nm (70-230 mesh ASTM) (E. Merck)

Packing method : Dry packing

Sample loading : The sample was dissolved in a small amount of organic solvent, mixed with a small quantity of adsorbent, triturated, dried and then placed gently on top of the column.

Detection : Fractions were examined by TLC observing under UV light (254 and 365 nm).

2.3.2 Flash Column Chromatography

Adsorbent : Silica gel 60 (No. 9385) particle size 0.400-0.063 nm (70-230 mesh ASTM) (E. Merck)

Packing method : Wet packing

Sample loading : The sample was dissolved in a small amount of eluent and then applied gently on top of the column.

Detection : Fractions were examined in the same manner as described in section 2.3.1.

2.3.3 Medium Pressure Liquid Chromatography

Adsorbent : 1. Silica gel 60 (No. 9385) particle size 0.400-0.063 nm (70-230 mesh ASTM) (E. Merck)

2. Polyamide SC60 (0.05-0.16 mm; Machery-Nagel)

Packing method : Dry packing

Sample loading : The sample was dissolved in a small amount of eluent and then applied gently on top of the column.

Detection : Fractions were examined in the same manner as described in section 2.3.1.

2.3.4 Gel Filtration Chromatography

Gel filter : Sephadex LH20 (Pharmacia)

Packing method : Gel filter was suspended in the eluent and left standing to swell for 24 hours prior to use. It was then poured into the column and allowed to set tightly.

Sample loading : The sample was dissolved in a small volume of eluent and applied on top of the column.

2.3.5 High Pressure Liquid Chromatography (HPLC)

Column (Semi-prep.) : 1. Shim-pack PREP-SIL No. 2025810
2. Bisschoff HPLC, LiChrospher 100 RP-18, 10 μm
3. Machery-Nagel SP 250/21 Nucleosil 100-7 C-18 No. 8065013
4. Merck LichroCART 250-10, LiChrospher 100 Diol, 10 μm
5. Bisschoff HPLC, Spherisorb CN, 5 μm

(Analytical) : 1. Merck LichroCART, LiChrospher 100 RP18, 5 μm
2. Merch LichroCART, LiChrospher 100 Diol, 5 μm
3. Merch LichroCART, LiChrospher 100 CN, 6 μm

Flow rate : 1. 4 mL/min for semi-preparative column
2. 1 mL/min for analytical column

Mobile phase : 1. Ethylacetate-hexane for normal phase
2. Gradient Acetonitrile-H_2O + 0.005% Trifluoro acetic acid (TFA) for RP-18 and CN columns
3. Isocratic 95% acetonitrile (MeCN) in H_2O + 0.005% TFA for Diol column

Sample preparation : The sample was dissolved in a small amount of eluent and filtered through millipore filter paper before injection.

Injection volume : 1 mL

Pump : 1. LC-8A (Shimadzu)
2. K-1001 (Knauer)

Detector : 1. SPD-10A UV Detector (Shimadzu)
2. K-2001 (Knauer)

Recorder : 1. C-R6A Chromatopac (Shimadzu)
2. ECW 2000 Integration Package

Temperature : Room temperature

2.4 Spectroscopy

2.4.1 Ultraviolet (UV) Absorption Spectra

UV (in methanol) spectra were obtained on a Milton Roy Spectronic 3000 Array spectrophotometer (Pharmaceutical Research Instrument Center, Faculty of Pharmaceutical Sciences, Chulalongkorn University).

2.4.2 Infrared (IR) Absorption Spectra

IR spectra (KBr disc and film) were recorded on a Perkin Elmer FT-IR 1760X spectrometer (Scientific and Technological Research Equipment Center, Chulalongkorn University).

2.4.3 Mass Spectra

Electron impact and high-resolution electron impact mass spectra (EIMS and HREIMS) were obtained with a Varian MAT 311A, Electrospray Ionization mass spectrometry (ESIMS) on a Finnigan MAT TSQ 700 and high-resolution fast atom bombardment mass spectrometry (HRFABMS) on a Finnigan MAT 95 (University of Hohenheim, Stuttgart, Germany).

2.4.4 Proton and Carbon-13 Nuclear Magnetic Resonance (^{1}H and ^{13}C-NMR) Spectra

^{1}H NMR (300 MHz) and ^{13}C NMR (75 MHz) spectra were obtained with a Bruker Avance DPX-300 FT-NMR spectrometer (Faculty of Pharmaceutical Sciences, Chulalongkorn University) or a Varian Unity Inova 300 MHz instrument (University of Hohenheim, Stuttgart, Germany).

^{1}H NMR (500 MHz) and ^{13}C NMR (125 MHz) spectra were obtained with a JEOL JMN-A 500 NMR spectrometer (Scientific and Technological Research Equipment Center, Chulalongkorn University) or a Varian Unity Inova 500 MHz instrument (University of Hohenheim, Stuttgart, Germany).

Solvents for NMR spectra were deuterated dimethylsulfoxide (DMSO-d_6), deuterated chloroform (chloroform-*d*), deuterated acetone (acetone-d_6), deuterated methanol (methanol-d_3) and deuterated pyridine (pyridine-d_5). Chemical shifts were reported in ppm scale using the chemical shift of the solvent as the reference signal.

2.5 Physical Properties

2.5.1 Melting Points

Melting points were obtained on a Fisher-Johns Melting Point Apparatus (Department of Pharmaceutical Botany, Faculty of Pharmaceutical Sciences, Chulalongkorn University).

2.5.2 Optical Rotations

Optical rotations were measured on a Perkin Elmer Polarimeter 341 (Pharmaceutical Research Instrument Center, Faculty of Pharmaceutical Sciences, Chulalongkorn University).

2.5.3 Circular Dichroism (CD) Spectra

CD Spectra were recorded on a JASCO J-715 spectropolarimeter (Pharmaceutical Research Instrument Center, Faculty of Pharmaceutical Sciences, Chulalongkorn University).

2.6 Solvents

Throughout this work, all organic solvents were of commercial grade and were redistilled prior to use.

3. Extraction and Isolation

3.1 Extraction and Isolation of Compounds from *Artocapus gomezianus*

3.1.1 Extraction

The dried roots of *Artocarpus gomezianus* (8.5 kg) were chopped, ground and then extracted with petroleum ether (2 x 30 L), ethyl acetate (3 x 30 L) and methanol (2 x 30 L) to give, after removal of the organic solvent, a pet ether extract (24.95 g, 0.29% based on dried weight of roots), an ethyl acetate extract (486.1 g, 5.72% based on dried weight of roots) and a methanol extract (1300 g, 15.29% based on dried weight of roots).

3.1.2 Isolation

3.1.2.1 Isolation of Compound AG11 (Artogomezianol)

The methanol extract (100 g) was fractionated by vacuum liquid column chromatography using a sintered glass filter column of silica gel (No. 7734, 400 g). Elution was performed in a polarity gradient manner with mixtures of hexane and EtOAc (100:0 to 0:100). The eluates were collected 500 ml per fraction and examined by TLC (silica gel, EtOAc-hexane 2:1). Fractions (42 fractions) with similar chromatographic pattern were combined to yield thirteen fractions: fractions A-1 (26 mg), A-2 (9 mg), A-3 (67 mg), A-4 (9 mg), A-5 (32 mg), A-6 (48 mg), A-7 (641 mg), A-8 (338 mg), A-9 (956 mg), A-10 (23.5 g), A-11 (4.6 g), A-12 (12.7 g) and A-13 (56 g).

Fraction A-12 (12.7 g) was further purified on a vacuum liquid column (silica gel No. 7734, 240 g; chroloform and methanol with increasing polarity). Twenty fractions, approximately 300 ml each, were collected. The eluates were examined by TLC (silica gel, acetone-toluene 4.5 : 5.5). Combination of fractions showing similar chromatographic pattern gave fractions B-1 (7 mg), B-2 (175 mg), B-3 (580 mg), B-4 (9.2 g) and B-5 (1.4 g).

Fraction B-4 (240 mg) was equally divided into eight portions. Each was fractionated by gel filtration chromatography using a column of Sephadex LH20 (100 g, 2.5 x 80 cm) with methanol as the eluent. The eluates were collected 20 ml per fraction and examined by TLC (silica gel, acetone-toluene 4.5:5.5). Fractions 2-4 from each column were combined (124 mg) and further separated by preparative TLC (precoated silica gel 60 F_{254} plates, 1 mm, 15 x 20 cm, with acetone-toluene 4.5:5.5). Purification of the obtained residue on a Sephadex LH20 column with methanol gave 9 mg of compound AG11 as a yellow gum. This compound was later identified as a new dimeric stilbene, which was subsequently named artogomezianol [**441**] (R_f 0.23, silica gel, acetone-toluene 4.5:5.5).

3.1.2.2 Isolation of Compound AG12 (Andalasin A)

Fraction B-3 (580 mg) was purified by flash column chromatography (silica gel 60 No. 9385, 100 g; 70% EtOAc in hexane). Thirty-two fractions (50 ml per fraction) were collected and combined according to their TLC behavior (silica gel, MeOH-$CHCl_3$-H_2O 2:8:0.1). Fractions 5-7 (163 mg) were combined and further separated by gel filtration chromatography, using a column of Sephadex LH20 with gradient elution ($CHCl_3$-MeOH 50:50 to 0:100) to furnish compound AG12 as a yellow solid (22 mg, R_f 0.22, silica gel, methanol-chloroform 2:8). This compound was subsequently identified as andalasin A [**442**].

3.2 Extraction and Isolation of Compound from the Bark of *Millettia erythrocalyx*

3.2.1 Extraction

The dried, coarsely powdered stem bark of *Millettia erythrocalyx* (2 kg) were macerated with ethyl acetate (4 x 4 L), and MeOH (3 x 4 L) to give an EtOAc extract (37g, 1.85% based on dried weight of stem bark) and a MeOH extract (164 g, 8.2% based on dried weight of stem bark) after evaporation of the organic solvent.

The methanol extract was then partitioned between ethyl acetate (3.8 L) and water (1 L). The aqueous fraction was further shaken with butanol. The butanol layer was dried to yield 29 g of a butanol extract (1.45% based on dried weight of stem bark) whereas 90 g of an aqueous extract (4.5% based on dried weight of stem bark) was obtained after freeze-drying.

3.2.2 Isolation

3.2.2.1 Isolation of Compounds from Ethyl acetate Extract

The ethyl acetate extract (37 g) was separated by vacuum liquid column chromatography (silica gel No. 7734, 400 g). The eluates were collected 500 mL per fraction. Elution was performed in a polarity gradient manner with mixtures of hexane and ethyl acetate (100:0 to 0:100). Thirty-three fractions were collected. Fractions with similar TLC pattern (silica gel, EtOAc-hexane 1:1) were combined to yield 9 fractions: fractions C-1 (22 mg), C-2 (60 mg), C-3 (3.3 g), C-4 (1.5 g), C-5 (535 mg), C-6 (785 mg), C-7 (7.1 g), C-8 (4.8 g) and C-9 (8.8 g).

3.2.2.1.1 Isolation of Compound ME1 (Derricidin)

Fraction C-5 (535 mg) was re-chromatographed on a silica gel 60 (No. 9385, 120 g) column. Gradient elution (3% ethyl acetate in hexane to 10% ethyl acetate in hexane) was performed (50 mL per fraction) to give 7 fractions: fractions I (20 mg), II (126 mg), III (43 mg), IV (40 mg), V (128 mg), VI (26 mg) and VII (153 mg).

Fraction II (126 mg) was further purified on a silica gel 60 column with hexane-EtOAc gradient elution. Fractions 3-6 (48 mg) from this column were combined and re-purified on Sephadex LH20 with acetone as eluent to afford 25 mg of compound ME1 (yellow needles, R_{f} 0.40, silica gel, EtOAc-hexane 1:20). It was later identified as derricidin [**443**].

3.2.2.1.2 Isolation of Compounds ME2 (7-γ,γ-Dimethylallyloxyflavanone) and ME3 (2'-Hydroxy-3,4-methylenedioxy-4'-γ,γ-dimethylallyloxychalcone)

Fraction C-6 (785 mg) was subjected to column chromatography using silica gel 60 (No. 7734) as adsorbent. Mixtures of ethyl acetate and hexane (2:98 to 10:90) were used as mobile phase. Purification of fractions 20-26 (75 mg) by RP18 HPLC (Bisschoff HPLC 250 x 25 mm column, LiCrospher 100RP-18, 10 μm) with 70% acetonitrile in H_2O + 0.05%Trifluoro acetic acid (TFA) as eluent and UV-VIS detection (λ 225 nm) gave compounds ME2 (1.5 mg, R_{f} 0.13, silica gel, ethyl acetate-hexane 1:9) and ME3 (1 mg, R_{f} 0.14, silica gel, ethyl acetate-hexane 1:9). Compounds ME2 and ME3 were identified as 7-γ,γ-dimethylallyloxyflavanone [**444**] and 2'-hydroxy-3,4-methylenedioxy-4'-γ,γ-dimethylallyloxychalcone [**445**], respectively.

3.2.2.1.3 Isolation of Compounds ME4 (Lupeol) and ME5 (Ponganone I)

Fraction C-7 (7.1 g) was subjected to flash column chromatography (silica gel 60 No. 7734, gradient 10%-50% EtOAc in pet. ether). Six fractions were obtained: fractions D-1 (1.1 g), D-2 (4.5 g), D-3 (343 mg), D-4 (162 mg), D-5 (557 mg) and D-6 (242 mg).

Fraction D-2 (4.5 g) was further separated by repeated column chromatography (silica gel 60, No. 7734, EtOAc-pet. ether gradient elution) to afford 9 major fractions: fractions E-1 (230 mg), E-2 (247 mg), E-3 (2.4 g), E-4 (595 mg), E-5 (323 mg), E-6 (160 mg) and E-7 (453 mg).

Fraction E-3 (2.4 g) was re-separated on Sephadex LH20 (chloroform and methanol 1:1). The eluates (50 mL per fraction) were examined by TLC (silica gel, chloroform-hexane 1:1), and then combined according to their chromatographic patterns to yield 3 fractions: fractions E-3-1 (1.2 g), E-3-2 (1.0 g) and E-3-3 (236 mg).

Fraction E-3-2 (1.0 g) was separated on a silica gel 60 flash column with ethyl acetate-pet. ether gradient elution (10:90 to 20:80). Eluates (50 mL each) with similar TLC behavior (silica gel, EtOAc-pet. ether 2:8) were pooled to give nine fractions. Fraction 2 (223 mg) from this column was further separated on a silica gel 60 (No. 9385) column. Elution was performed with $CHCl_3$-toluene gradient (8:92 to 15:85, 50 mL per fraction) to give 16 fractions. Similar fractions were combined after examined by TLC (chloroform-toluene 1:4). The TLC chromatogram of fractions 2-3 and fractions 10-13 showed a single spot under UV light at 254 nm and fraction 15 exhibited a dark purple single spot with anisaldehyde detection. Fractions 2-3, after removal of solvent gave compound ME3 (1.4 mg, R_f 0.44, silica gel, $CHCl_3$-toluene 1:4), which was identified as 2'-hydroxy-3,4-methylenedioxy-4'-γ,γ-dimethylallyloxychalcone [**445**]. Fractions 10-13 gave compound ME5 (60 mg, R_f 0.27, silica gel, $CHCl_3$-toluene 1:4), which was later identified as ponganone I [**446**]. And compound ME4 (64 mg, R_f 0.18, silica gel, $CHCl_3$-toluene 1:4) was obtained from fraction 15 and later identified as lupeol [**169**].

3.2.2.1.4 Isolation of Compound ME6 (Karanjin)

Fraction E4 (595 mg) was separated on a Sephadex LH20 ($CHCl_3$ and MeOH 1:1) column. Eleven fractions (30 mL each) were collected and subsequently combined according to their TLC behavior (silica gel, ethyl acetate-hexane 1:1) to yield 3 main fractions: fractions E-4-1 (469 mg), E-4-2 (181 mg) and E-4-3 (7 mg).

Fraction E-4-2 (181 mg) was re-purified on a Sephadex LH20 column ($CHCl_3$ and MeOH 1:1). Fractions 7-9 (23 mg) from this column were combined and further separated by HPLC (normal phase column, Shimpack PREP-SIL), eluted with ethyl acetate-pet. ether (2:8) and detected with UV-VIS light to give compound ME6 as a yellow powder (1.7 mg, R_f 0.3, silica gel, ethyl acetate-pet. ether 2:8). It was subsequently identified as karanjin [**295**].

3.2.2.1.5 Isolation of Compound ME7 (Milletenone)

Fraction C-8 (4.8 g) was subjected to medium pressure liquid chromatography (MPLC) on a Sephadex LH20 column with acetone, methanol, 5%H_2O in methanol, 20%H_2O in methanol and acetone in H_2O, respectively, as mobile phase. Forty-five fractions (15 mL each) were collected and then combined according to their TLC behavior (silica gel, ethyl acetate-pet. ether 3:7).

Fraction C-8-2 (3.6 g) was further separated over a silica gel 60 (No. 9385) column (EtOAc-pet. ether (10:90 to 100:0) gradient elution). Fractions (41 fractions) showing similar chromatographic pattern were combined (TLC, silica gel, EtOAc-pet. ether 3:7) to yield 9 fractions: fractions F-1 (76 mg), F-2 (28 mg), F-3 (289 mg), F-4 (256 mg), F-5 (685 mg), F-5 (685 mg), F-6 (285 mg), F-7 (753 mg), F-8 (570 mg) and F-9 (696 mg).

Fraction F-5 (685 mg) was fractionated on a silica gel 60 column (ethyl acetate-pet. ether 1:9). Twelve fractions (30 mL each) were collected. Fractions 6-9 (345 mg) were combined and further purified on a Sephadex LH20 column (acetone) to give 30 mg of compound ME7 as yellow needles (R_f 0.38, silica gel, ethyl acetate-pet. ether 2:3). This compound was identified as milletenone [**280**].

3.2.2.1.6 Isolation of Compound ME8 (Ovalifolin)

Repeated column chromatography of fraction F-7 (753 mg) (silica gel 60, ethyl acetate-pet. ether 2:8) gave 92 mg of compound ME8 (R_f 0.46, silica gel, ethyl acetate-pet. ether 1:1). This compound was later identified as ovalifolin [**308**].

3.2.2.1.7 Isolation of Compound ME9 (Pongol methyl ether)

Fraction F-8 (570 mg) was separated by silica gel 60 column chromatography (gradient elution EtOAc-pet. ether 20:80 to 30:70) to afford 139 mg of compound ME9 as a yellowish powder (R_f 0.38, silica gel, EtOAc-pet. ether 2:3). This compound was identified as pongol methyl ether [**447**], which was isolated from natural sources for the first time in this study.

3.3.3.1.8 Isolation of Compound ME10 (Millettocalyxin B)

Fraction C-9 (8.8 g) was separated on a polyamide column, eluted with the gradient mixture of ethanol and water + 0.01% trifluoro acetic acid (TFA) (20:80 to 100:0). The eluates (49 fractions) were combined based on their TLC behavior (silica gel, EtOAc-hexane 6:4) to yield 5 fractions: fractions C-9-1 (420 mg), C-9-2 (6.2 g), C-9-3 (200 mg), C-9-4 (1.4 g) and C-9-5 (550 mg).

Fraction C-9-2 (6.2 g) was further separated on a Sephadex LH20 MPLC column (methanol). Fifty-one fractions (20 mL each) were collected, examined by TLC (silica gel, ethyl acetate-pet. ether 7:3) and combined to give 13 major fractions: fractions G-1 (156 mg), G-2 (1.6 g), G-3 (160 mg), G-4 (102 mg), G-5 (498 mg), G-6 (304 mg), G-7 (1.8 g), G-9 (615 mg), G-10 (60 mg), G-11 (71 mg), G-12 (41 mg) and G-13 (41 mg).

Fraction G-6 (304 mg) was subjected to silica gel 60 column chromatography, eluted with gradient mixtures of ethyl acetate and pet. ether (20:80 to 50:50). Eighteen fractions (30 mL per fraction) were collected, then combined according to their TLC patterns (silica gel, ethyl acetate-pet. ether 4:6) to give 5 fractions: fractions G-6-1 (2 mg), G-6-2 (16 mg), G-6-3 (39 mg), G-6-4 (235 mg) and G-6-5 (37 mg).

Compound ME8 (ovalifolin [**308**], 16 mg) was obtained as a yellow powder from fraction G-6-2 (R_f 0.46, silica gel, ethyl acetate-pet.ether 1:1).

Fraction G-6-4 (235 mg) was re-chromatographed over a silica gel 60 column (ethyl acetate-pet. ether 30:70 to 50:50 gradient elution) to afford 22 mg of compound ME10 (R_f 0.43, silica gel, ethyl acetate-pet. ether 1:1). This compound was later identified as a new flavone, with the trivial name millettocalyxin B [**448**].

3.2.2.1.9 Isolation of Compound ME11 (Milletenin C)

Fraction G-7 (1.8 g) was subjected to medium pressure liquid chromatography (MPLC) (silica gel, gradient mixtures of EtOAc-pet. ether 20:80 to 100:0). Fractions (138 fractions) with similar chromatographic patterns were combined (TLC: silica gel, ethyl acetate-pet. ether 1:1) to give 14 major fractions: fractions H-1 (2 mg), H-2 (2 mg), H-3 (1 mg), H-4 (25 mg), H-5 (27 mg), H-6 (50 mg), H-7 (502 mg), H-8 (235 mg), H-9 (162 mg), H-10 (192 mg), H-11 (30 mg), H-12 (31 mg), H-13 (10 mg) and H-14 (12 mg).

Compound ME5 (ponganone I [**446**], 2 mg) was obtained as a yellow powder from fraction H-1 (R_f 0.27, silica gel, chloroform-toluene 1:4).

Compound ME11 (31 mg) was obtained as a yellow powder from fraction H-12 (R_f 0.22, silica gel, ethyl acetate-pet. ether 3:2). It was later identified as milletenin C.

Fraction H-4 (25 mg) was further purified by flash column chromatography (silica gel, ethyl acetate-pet. ether 2:8) to give compound ME8 (6 mg, ovalifolin [**308**]) and compound ME9 (6.5 mg, Pongol methyl ether [**447**]), respectively.

3.2.2.1.10 Isolation of Compound ME12 (Millettocalyxin C)

Fraction H-8 (235 mg) was fractionated on a silica gel column (EtOAc-$CHCl_3$ 10:90) to yield compound ME10 (19 mg, millettocalyxin B [**448**], R_f 0.43, silica gel, EtOAc-pet. ether 1:1) and compound ME12 (34 mg, R_f 0.20, silica gel, EtOAc-pet. ether 1:1). Compound ME12 was identified as a new flavone, 2',5'-dimethyoxy-[2",3":7,8]-furanoflavone, namely millettocalyxin C [**449**].

3.2.2.1.11 Isolation of Compound ME13 (Millettocalyxin A)

Fraction G-9 (615 mg) was separated on a silica gel 60 column (mixtures of ethyl acetate-pet. ether 20:80 to 100:0). The eluates were combined on the basis of their TLC composition (silica gel, ethyl acetate-pet. ether 60:40) to yield 11 fractions: fractions I-1 (2 mg), I-2 (9 mg), I-3 (43 mg), I-4

(117 mg), I-5 (108 mg), I-6 (24 mg), I-7 (8 mg), I-8 (14 mg), I-9 (33 mg), I-10 (7 mg) and I-11 (32 mg).

Compound ME9 (9 mg, pongol methyl ether [**447**]) was obtained as a yellow powder from fraction I-2.

Compound ME13 (8 mg, R_f 0.25, silica gel, ethyl acetate-pet. ether 3:2) was obtained as a yellow powder from fraction I-7. This new compound was identified as 3',4'-methylenedioxy-7,2'-dimethoxyflavone, and given the name millettocalyxin A [**450**].

3.2.2.1.12 Isolation of Compound ME14 (3',4'-Methylenedioxy-7-methoxyflavone)

Fraction I-4 (117 mg) was separated by HPLC using an RP18 column (Bischoff HPLC, 250 x 25 mm column, LiCrospher 100 RP-18, 10 μm) with 50% acetonitrile in H_2O + 0.05%TFA as eluent (flow rate 4 mL/min.) and UV-VIS detection (λ 225 nm) to give 4 mg of compound ME14 (R_f 0.4, silica gel, ethyl acetate-pet. ether 2:3). It was subsequently identified as 3',4'-methylenedioxy-7-methoxyflavone [**279**].

3.2.2.1.13 Isolation of Compound ME15 (Pongaglabrone)

Fraction G-11 (71 mg) was subjected to flash column chromatography (silica gel, mixture of ethyl acetate-pet. ether 20:80 to 25:75), which resulted in the isolation of compound ME7 (4 mg, milletenone [**280**]) and compound ME15 (9 mg, R_f 0.27, silica gel, ethyl acetate-pet. ether 2:3). Compound ME15 was later identified as pongaglabrone [**314**].

3.2.2.1.14 Isolation of Compound ME16 (Prunetin)

Fraction G-12 (41 mg) was performed by RP 18 HPLC (Bischoff HPLC, 250 x 25 mm column, LiCrospher 100 RP-18, 10 μm). Elution was performed in a polarity gradient manner with 45% acetonitrile in H_2O + 0.05%TFA to 60% acetonitrile in H_2O + 0.05%TFA (4 mL/min) to give compound ME16 (11 mg, R_f 0.67, silica gel, ethyl acetate-pet. ether 3:2). It was identified as prunetin [**451**].

3.2.2.2 Isolation of Compound from Butanol Extract

The butanol extract (25 g) was separated by MPLC over a polyamide column (120 g), eluted with 5% ethanol in H_2O + 0.01%TFA, 25% ethanol in H_2O + 0.01%TFA, 50% ethanol in H_2O + 0.01%TFA, 75% ethanol in H_2O + 0.01%TFA, ethanol and 50% acetone in ethanol, respectively. The eluates (200 mL each) were examined by analytical RP18 HPLC (Knauer HPLC pump K1001, Merck LichroCART 125 x 4 mm, LiChrospher 100 RP18, 5 μm) and Knauer UV-detector K-2001 (λ 220 nm). Elution was performed in a polarity gradient manner with CH_3CN-H_2O + 0.005% TFA (5:95 to 40:60) with flow rate of 1 mL/min. Fractions (19 fractions) with similar chromatographic pattern were combined to give 9 fractions: fractions J-1 (12.5 g), J-2 (4.6 g), J-3 (924 mg), J-4 (423 mg), J-5 (773 mg), J-6 (4.7 g), J-7 (461 mg), J-8 (700 mg) and J-9 (361 mg).

3.2.2.2.1 Isolation of Compound ME17 (Vicenin II)

Fraction J-4 (423 mg) was divided into two portions. Each was purified by RP18 HPLC (Knauer HPLC pump K1001, Macherey-Nagel, SP 250/21 Nucleosil 100-7 C-18, 250 x 25 mm) with UV 254 nm detection. Elution was performed in a polarity gradient manner with CH_3CN-H_2O + 0.005% TFA as follows: 15-35% CH_3CN-H_2O + 0.005% TFA 60 min, 35-50% CH_3CN-H_2O + 0.005% TFA 10 min and 50-100% CH_3CN-H_2O + 0.005% TFA 10 min (flow rate 4 mL/min). A total of 65 mg of compound ME17 was obtained (Rt 11.2 min, Merck LichroCART LiCrosphern100 RP18, 5 μm, eluted with 8-25% CH_3CN-H_2O + 0.005% TFA 25 min with flow rate 1 mL/min). This compound was later identified as vicenin II [**452**].

3.2.2.2.2 Isolation of Compound ME18 (Dihydrophaseic acid-4'-*O*-β-D-glucopyranoside)

Fraction J-2 (3 g) was divided into ten portions. Each portion was purified by HPLC using an RP18 column (Knauer HPLC pump K1001, Macherey-Nagel, SP 250/21, Nucleosil 100-7 C-18, 250 x 25 mm) and UV 254 nm detection. Elution was performed in a polarity gradient manner with CH_3CN-H_2O + 0.005% TFA (15:85 to 35:65 60 min, 35:65 to 50:50 10 min and 50:50 to 0:100 10 min, flow rate 4 mL/min). The eluates were examined by analytical HPLC (RP18 column, Merck LichroCART 125 x 4 mm, Lichrospher 100 RP18, 5 μm) eluted with CH_3CN-H_2O + 0.005% TFA (8:92 to 25:75 25 min., flow rate 1 mL/min). Five major fractions were obtained: fractions K-1 (1.3 g), K-2 (83 mg), K-3 (97 mg), K-4 (130 mg) and K-5 (1 g).

Fraction K-4, after removal the solvent gave compound ME17 (130 mg, vicenin II [**452**]) as a yellow powder.

Fraction K-2 (83 mg) was separated by RP18 HPLC (Macherey-Nagel, SP 250/21, Nucleosil 100-7 C-18, 250 x 25 mm) eluted with CH_3CN-H_2O + 0.005% TFA (10:90 to 30:70 60 min., 30:70 to 45:55 10 min and 45:55 to 0:100 10 min with flow rate 4 mL/min) and detected with UV 254 nm (Knauer UV Detector K-2001). The target fraction was dried and further separated by HPLC using a CN column (Bischoff HPLC 250 x 25 mm, Spherisorb CN, 5 μm) with polarity gradient elution (CH_3CN-H_2O + 0.005% TFA: 0:100 to 20:80 60 min, 20:80 to 30:70 10 min and 30:70 to 0:100 10 min, with flow rate 4 mL/min) to afford 25 mg of compound ME18 (Rt 6 min, Merck LichroCART, Licrospher 100 RP18, 5 μm, eluted with 8-25% CH_3CN-H_2O + 0.005% TFA 25 min, flow rate 1 mL/min). This compound was subsequently identified as dihydrophaseic acid-4'-*O*-β-D-glucopyranoside [**453**].

3.2.2.2.3 Isolation of Compound ME19 (Isovitexin)

Fraction J-7 (461 mg) was divided into two portions. Each was separated by HPLC using a CN column (Bischoff HPLC 250 x 25 mm, Spherisorb CN, 5 μm) eluted with CH_3CN-H_2O + 0.005% TFA in a polarity gradient manner (0:100 to 30:70 60 min., 30:70 to 45:55 10 min. and 45:55 to 0:100 10 min, flow rate 4 mL/min). The target fractions from the two column were combined, dried and further purified on an HPLC diol column (Merck LicroCART 250-10, 250 x 10 mm, LiChrospher 100 Diol, 10 μm) eluted with 95% CH_3CN-H_2O + 0.005% TFA (4 mL/min) to give compound ME19 (3 mg) at the retention time 25 min. It was identified as isovitexin [**454**].

3.3 Extraction and Isolation of Compound from the Roots of *Millettia erythrocalyx*

3.3.1 Extraction

The dried powdered roots of *Millettia erythrocalyx* (8 kg) were extracted with hexane (2 x 20 L), ethyl acetate (2 x 20 L) and methanol (2 x 20 L), successively. The obtained extracts were evaporated to dryness to give a hexane extract (91 g, 1.14% based on dried weight of the roots), an ethyl acetate extract (87 g, 1.09% based on dried weight of the roots) and a methanol extract (429 g, 5.36% based on dried weight of the roots).

3.3.2 Isolation

3.2.2.1 Isolation of Compound from Hexane Extract

The hexane extract (91 g) was equally divided into two portions: portions A and B and initially fractionated by vacuum column chromatography (silica gel No. 7734, 150 g), eluted with pet. ether and ethyl acetate in a polarity gradient manner (95:5 to 0:100). The eluates (500 mL each) from the two columns were examined by TLC (ethyl acetate-pet. ether 1:2) and then combined to yield 5 major fractions: fractions L-1 (9.4 g), L-2 (6.9 g), L-3 (14.8 g), L-4 (36.4 g) and L-5 (21.6 g).

3.3.2.1.1 Isolation of Compound ME20 (1-(4-Hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one)

Fraction L-2 (6.9 g) was subjected to flash column chromatography (silica gel, mixtures of pet. ether-ethyl acetate 95:5 to 85:15). Eighteen fractions were collected (200 mL each), and then combined according to their TLC patterns (silica gel, 20% ethyl acetate in pet. ether). Fraction L-2-1 (1.2 g) was further separated by MPLC on a silica gel 60 column (toluene-pet. ether 30:70 to 100:0). A total of 46 fractions (100 mL each) were collected and combined based on their TLC behavior (silica gel, toluene) to give 14 fractions: fractions M-1 (58 mg), M-2 (86 mg), M-3 (191 mg), M-4 (105 mg), M-5 (11 mg), M-6 (15 mg), M-7 (44 mg), M-8 (15 mg), M-9 (79 mg), M-10 (116 mg), M-11 (40 mg), M-12 (390 mg), M-13 (42 mg) and M-14 (211 mg).

Fraction M-3 (191 mg) was separated by flash column chromatography over silica gel, eluted with 30% toluene in pet. ether. Fifteen fractions (100 mL each) were collected and examined by TLC (silica gel, toluene). Fractions 5-9 (13 mg) were combined, dried and re-chromatographed in a similar manner to give 7 mg of compound ME20 as a yellow powder (R_f 0.63, silica gel, toluene). It was later identified as 1-(4-hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one [**289**].

3.3.2.1.2 Isolation of Compound ME1 (Derricidin)

Separation of fraction M-7 (44 mg) on a silica gel column (40% toluene in pet. ether) gave compound ME1 (3.5 mg) as yellowish needles. This compound was later identified as derricidin [**443**].

3.3.2.1.3 Isolation of Compound ME21 (Purpurenone)

Fraction M-9 (79 mg) was purified by column chromatography (silica gel, toluene-pet. ether 30:70). Seventeen fractions (50 mL each) were collected and then examined by TLC (silica gel, toluene). Fractions 1-3 and fractions 6-7 were combined and dried to give compound ME20 (4 mg, 1-(4-hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one [**289**]) and compound ME21 (14 mg, R_f 0.44, silica gel, toluene), respectively. Compound ME21 was subsequently assigned as purpurenone [**455**].

3.3.2.1.4 Isolation of Compound ME22 (Pongaglabol)

Fraction M-12 (390 mg) was subjected to column chromatography (silica gel, toluene). The eluates (50 mL each) were collected and combined according to their TLC pattern (chloroform-toluene 1:2) to give 21 mg of compound ME22 (R_f 0.14, silica gel, chloroform-toluene 1:2). It was later identified as pongaglabol [**337**].

3.3.2.1.5 Isolation of Compound ME5 (Ponganone I)

Fraction L-3 (14.8 g) was separated by column chromatography (silica gel, mixture of ethyl acetate-pet. ether 7:93 to 10:90). Fraction L-3-2 (5.3 g) was further fractionated on a silica gel column (toluene). A total of 38 fractions (50 mL each) were collected and combined based on their TLC behavior (silica gel, 3% chloroform in toluene) leading to 11 major fractions: fractions N-1 (10 mg), N-2 (281 mg), N-3 (760 mg), N-4 (277 mg), N-5 (300 mg), N-6 (184 mg), N-7 (277 mg), N-8 (158 mg), N-9 (242 mg), N-10 (184 mg) and N-11 (1.1 g).

Compound ME5 (277 mg) was obtained as yellow crystals from fraction N-7. This compound was later identified as ponganone I [**446**].

3.3.2.1.6 Isolation of Compound ME23 (Pongamol)

Compound ME23 (54 mg) was obtained from fraction N-3 (760 mg) by separation over a silica gel 60 column (50-80% of toluene in pet. ether) (R_f 0.44, silica gel, toluene). It was identified as pongamol [**315**].

3.3.2.1.7 Isolation of Compounds ME24 (Ovalitenone) and ME25 ((-)-(2*S*)-6-Methoxy-[2",3":7,8]-furanoflavone)

Fraction L-4 (36.4 g) was fractionated by vacuum liquid chromatography (silica gel, pet. ether-ethyl acetate with increasing polarity). Fractions of 170 mL were collected. The separation was monitored by TLC (silica gel, ethyl acetate-pet. ether 3:7). Fractions 29-30 were combined and further separated on an MPLC silica gel column (gradient ethyl acetate-pet. ether 10:90 to 50:50). A total of thirty fraction were collected (150 mL per fraction) and then combined on the basis of their TLC behavior (silica gel, chloroform-toluene 1:4) to give 9 fractions: fractions O-1 (10 mg), O-2 (31 mg), O-3 (169 mg), O-4 (8.3 g), O-5 (5.8 g), O-6 (2 g), O-7 (3.4 g), O-8 (1.3 g) and O-9 (203 mg).

Fraction O-3 (169 mg) was subsequently separated by column chromatography (silica gel, toluene) to afford 26 mg of compound ME24 as a yellow powder and 16 mg of compound ME25 as colorless needles. Compound ME24 (R_f 0.24, silica gel, chloroform-toluene 1:2) was identified as ovalitenone [**312**]. Compound ME25 (R_f 0.20, silica gel, chloroform-toluene 1:1) was later identified as a new furanoflavone, (-)-(2*S*)-6-methoxy-[2",3":7,8]-furanoflavone [**456**].

3.3.2.1.8 Isolation of Compound ME7 (Milletenone)

Fraction O-4 (8.3 g) was separated by MPLC over a silica gel column, eluted with mixtures of EtOAc-pet. ether (5:95 to 15:85). Fifty-five fractions (250 mL each) were collected and combined based on their TLC behavior (silica gel, ethyl acetate-toluene 1:5) to give 19 fractions: fractions P-1 (16 mg), P-2 (6 mg), P-3 (253 mg), P-4 (52 mg), P-5 (43 mg), P-6 (40 mg), P-7 (165 mg), P-8 (152 mg), P-9 (182 mg), P-10 (71 mg), P-11 (341 mg), P-12 (578 mg), P-13 (2.0 g), P-14 (1.6 g), P-15 (368 mg), P-16 (127 mg), P-17 (122 mg), P-18 (31 mg) and P-19 (322 mg).

Compound ME23 (pongamol [**315**]) was obtained from fraction P-2 (6 mg) as a yellow powder after removal of the solvent. Compound ME24 (ovalitenone [**312**]) was obtained from fraction P-4 (52 mg) as a yellow powder. Compound ME7 was obtained from fraction P-7 (165 mg) as yellow needles after evaporation of the solvent. Compound ME7 was identified as milletenone [**280**].

3.3.2.1.9 Isolation of Compound ME26 (Ponganone V)

Fraction P-11 (341 mg) was separated on a silica gel 60 column, with gradient elution (chloroform-toluene 0:100 to 10:100). Sixteen fractions (50 mL per fraction) were collected and examined by TLC (silica gel, ethyl acetate-toluene 1:5). Fractions 10-13 were combined, dried and recrystallized from methanol to give 34 mg of compound ME26 as colorless needles (R_f 0.44, silica gel, ethyl acetate-toluene 1:5). It was subsequently identified as ponganone V [**457**].

3.3.2.1.10 Isolation of Compound ME27 (2,5-Dimethoxy-4-hydroxy-[2",3":7,8]-furanoflavan)

Fraction P-14 (1.6 g) was subjected to MPLC over silica gel (gradient elution with ethyl acetate-pet. ether 5:95 to 100:0) and then re-crystallized from methanol to give compound ME27 (183 mg) as colorless prisms (R_f 0.29, silica gel, chloroform-toluene 1:1). It was identified as a new furanoflavan, 2,5-dimethoxy-4-hydroxy-[2",3":7,8]-furanoflavan [**458**].

Repetitive chromatography of fraction O-5 (5.8 g) on an MPLC silica gel 60 column eluted with a polarity gradient elution (chloroform-toluene 0:100 to 50:50). Forty-four fractions (200 mL each) were collected. Compounds ME24 (19 mg, ovalitenone [**312**]) and ME7 (131 mg, milletenone [**280**]) were obtained from fractions 1-4 and fraction 11, respectively. Fraction 33 (400 mg) was dried and recrystallized from methanol to give compound ME27 (95 mg).

3.3.2.1.11 Isolation of Compound ME28 (3,4-Methylenedioxy-2',4'-dimethoxychalcone)

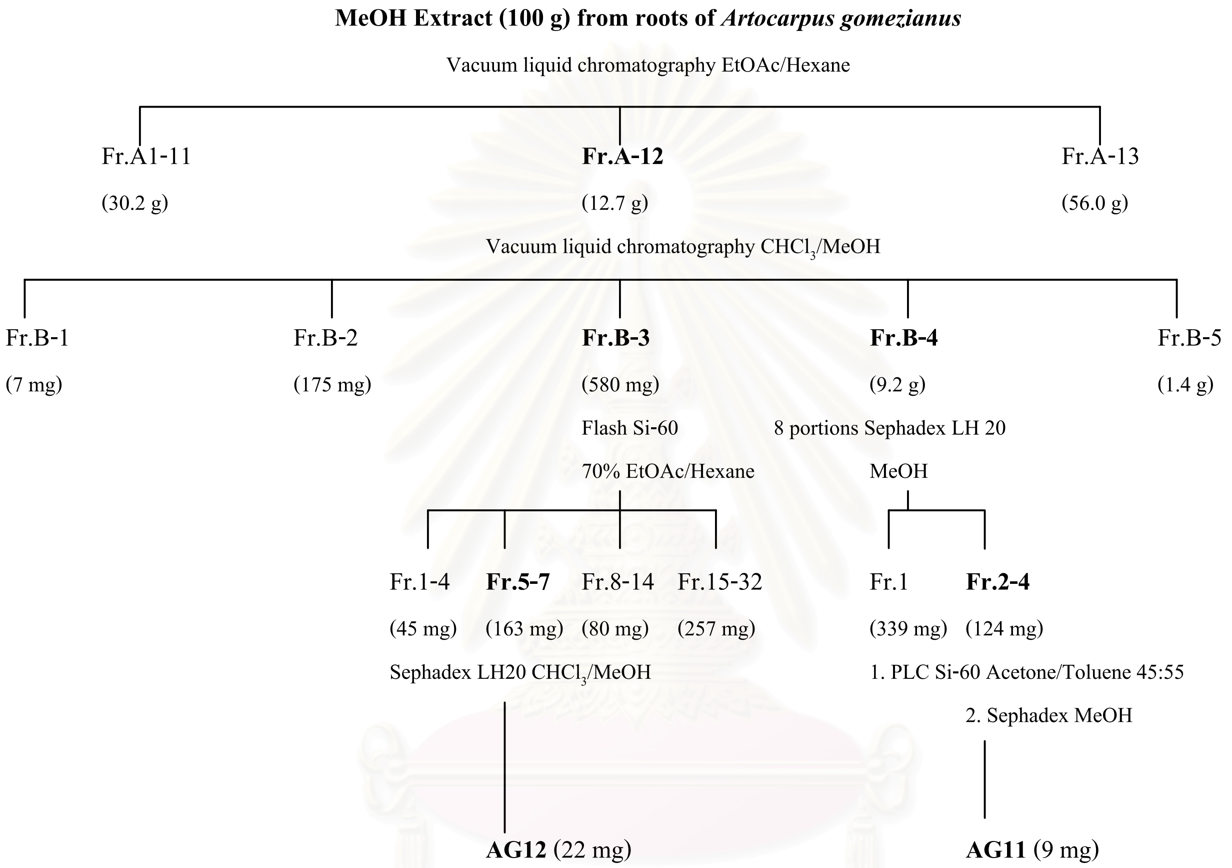
Separation of fraction O-6 (2.0 g) on a silica gel column with gradient elution (ethyl acetate-toluene) gave 16 fractions: Q-1 (10 mg), Q-2 (20 mg), Q-3 (13 mg), Q-4 (47 mg), Q-5 (155 mg), Q-6 (44 mg), Q-7 (43 mg), Q-8 (9 mg), Q-9 (8 mg), Q-10 (14 mg), Q-11 (26 mg), Q-12 (35 mg), Q-13 (64 mg), Q-14 (505 mg), Q-15 (668 mg) and Q-16 (471 mg).

Fractions Q-2 and Q-5, after drying, gave compound ME24 (ovalitenone [**312**], 20 mg) and compound ME7 (milletenone [**280**], 155 mg).

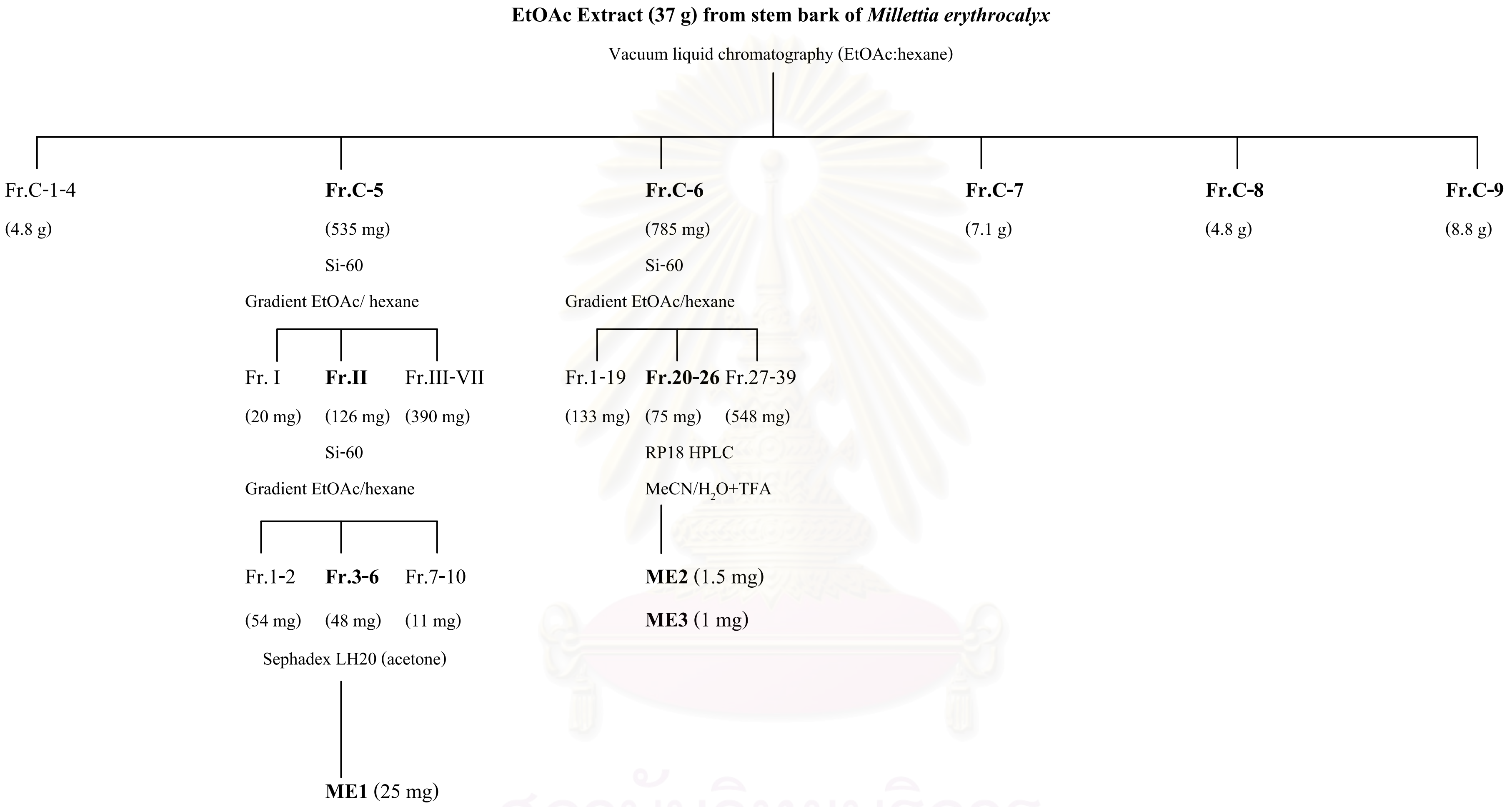
Fraction Q-11 (26 mg) was re-chromatographed on a silica gel 60 column (toluene) to give 9 mg of compound ME28 as a yellow powder (R_f 0.53, silica gel, EtOAc-toluene 1:5). This compound was later identified as 3,4-methylenedioxy-2',4'-dimethoxychalcone [**459**], which was isolated from natural sources for the first time in this study.

3.3.2.1.12 Isolation of Compound ME29 (Lanceolatin B)

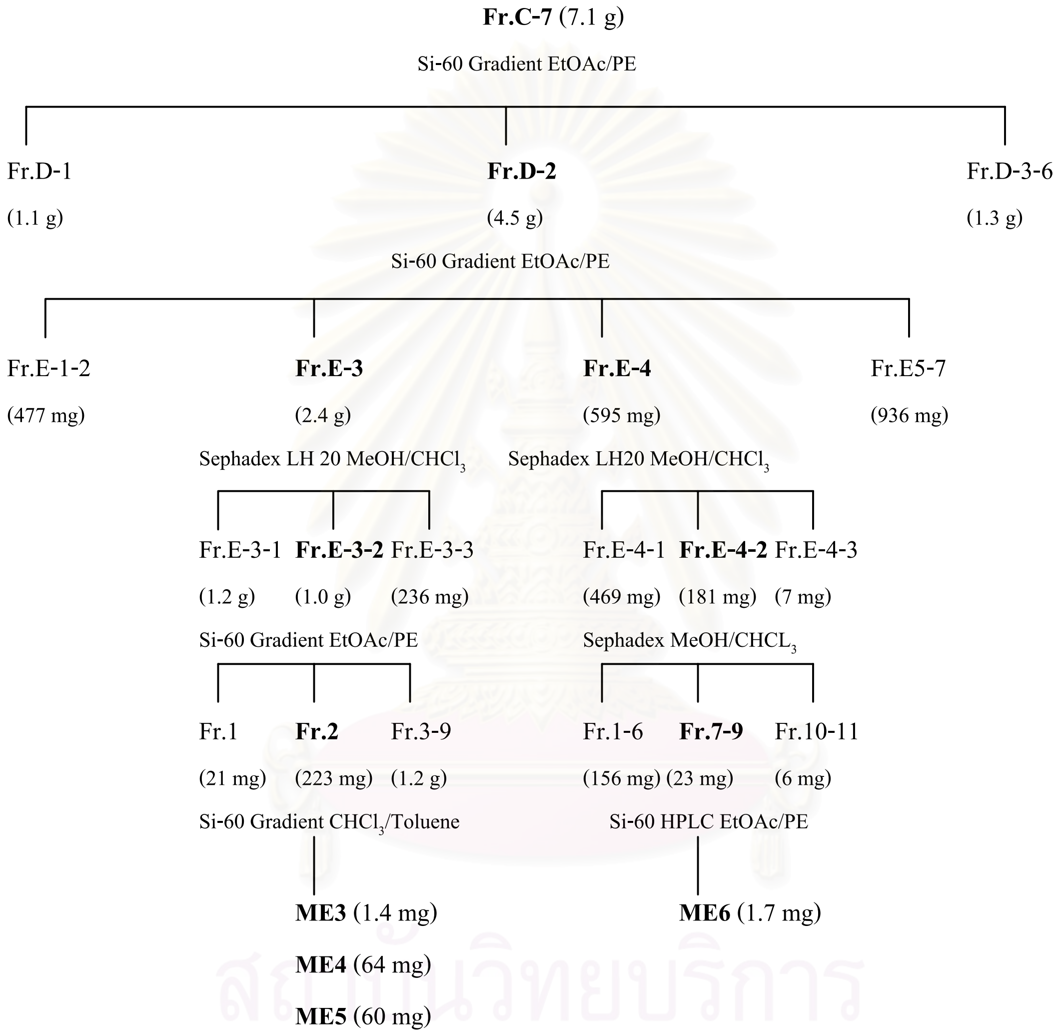
Fraction Q-15 (668 mg) was separated on an MPLC silica gel column (5% ethyl acetate in toluene). Fifteen fractions (50 mL each) were collected. Fraction 8, after drying, was recrystallized from methanol to give 16 mg of compound ME29 (R_f 0.38, silica gel, ethyl acetate-toluene 1:5). It was later elucidated as lanceolatin B [**278**].



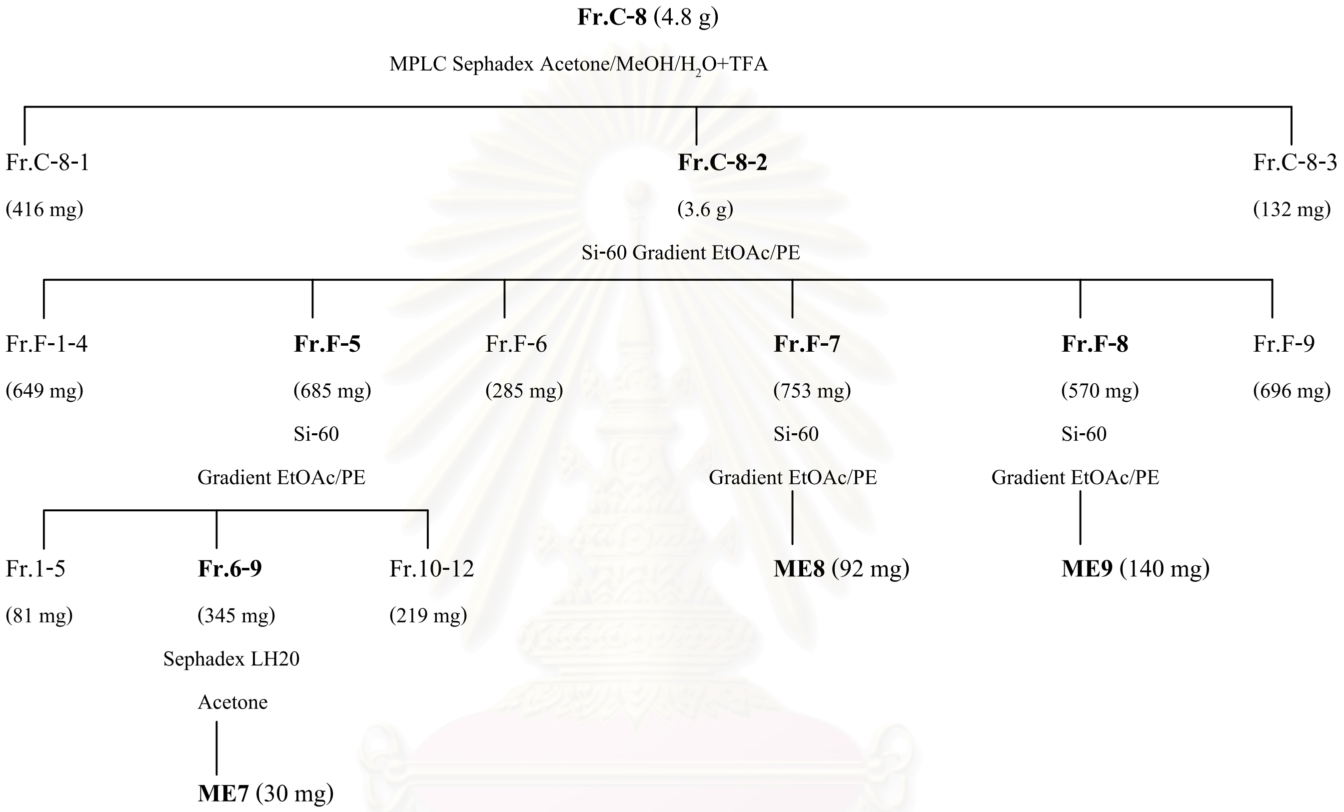
Scheme 1 Separation of the MeOH extract of the roots of *Artocarpus gomezianus*



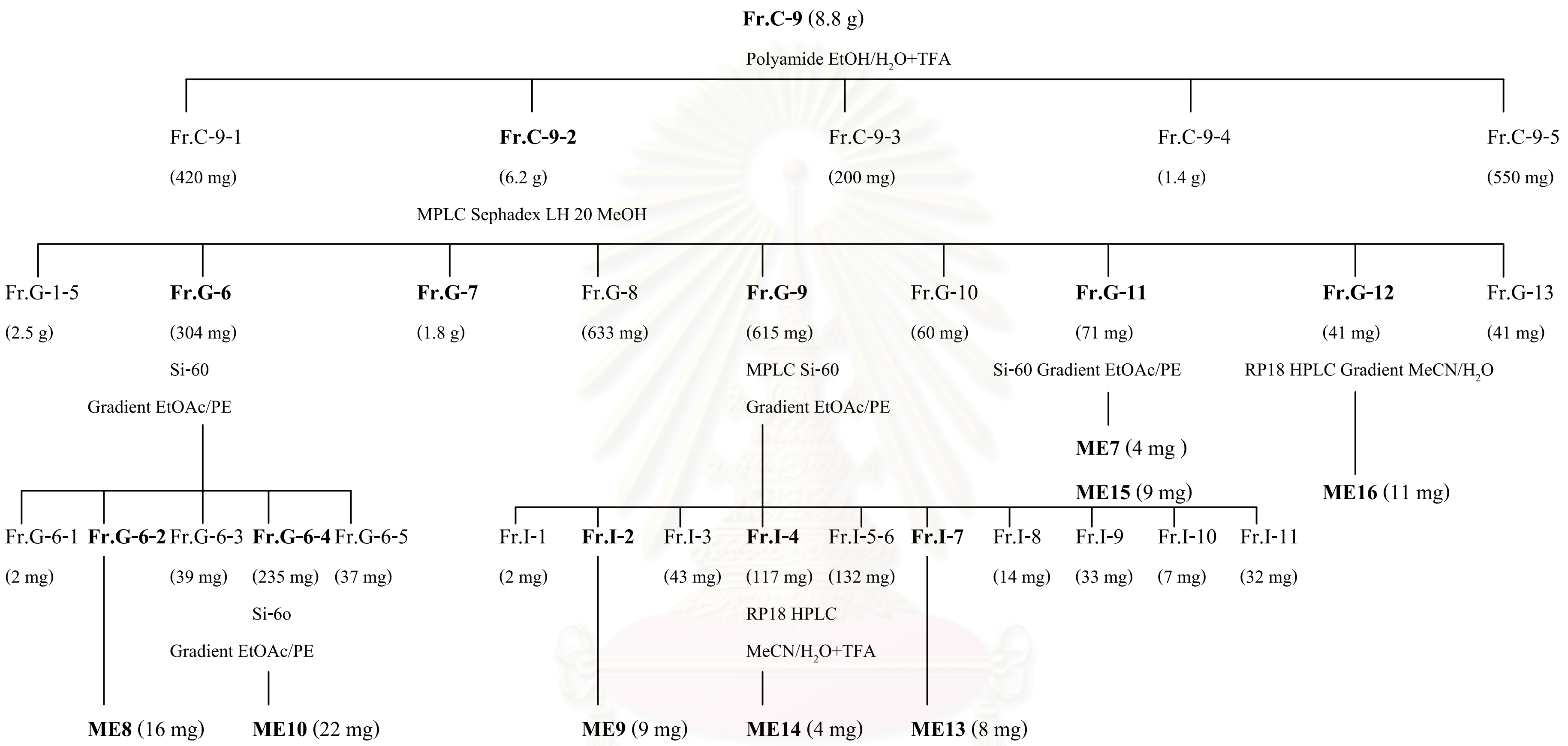
Scheme 2 Separation of the EtOAc extract of the stem bark of *Millettia erythrocalyx*



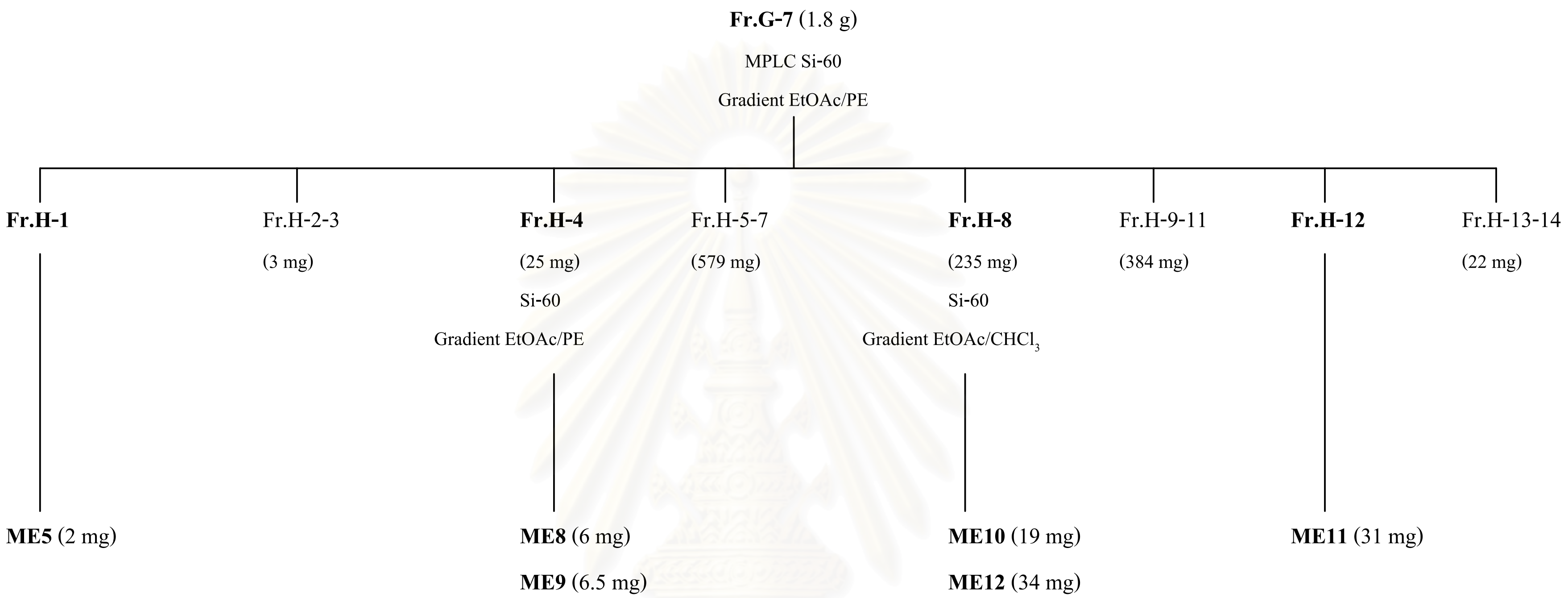
Scheme 3 Separation of fraction C-7 from the EtOAc extract of the stem bark of *M. erythrocalyx*



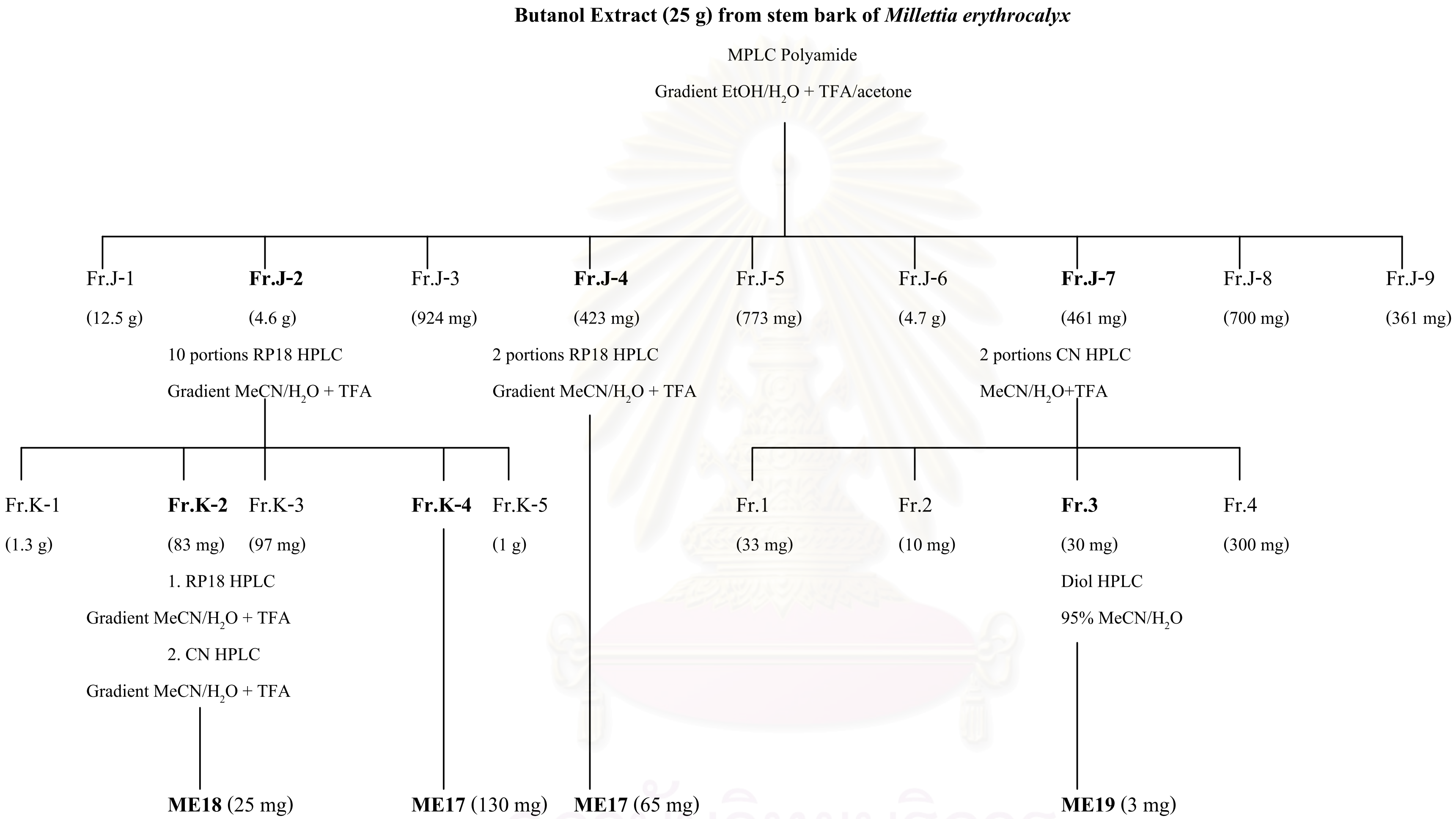
Scheme 4 Separation of fraction C-8 from the EtOAc extract of the stem bark of *M. erythrocalyx*



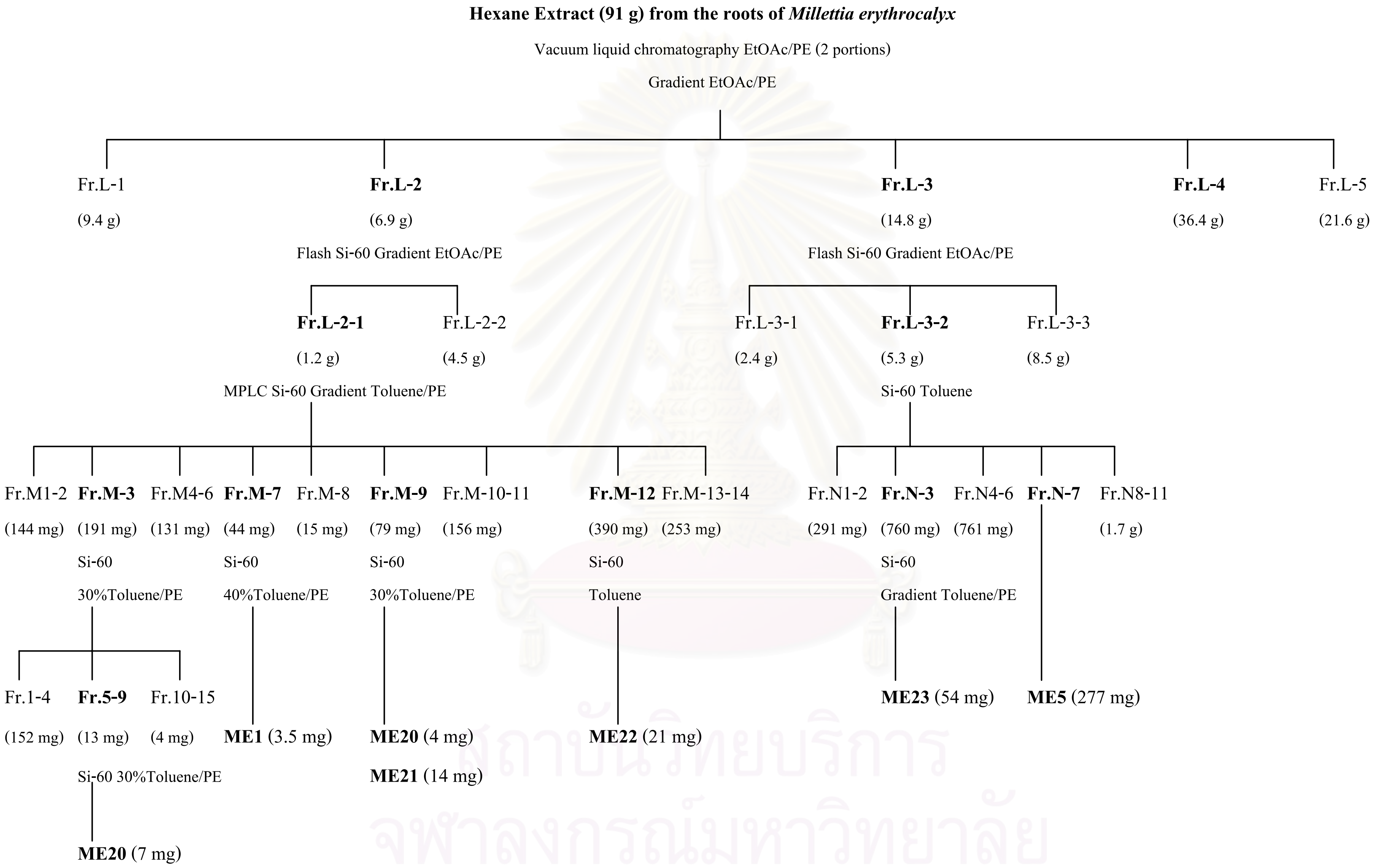
Scheme 5 Separation of fraction C-9 from the EtOAc extract of the stem bark of *M. erythrocalyx*



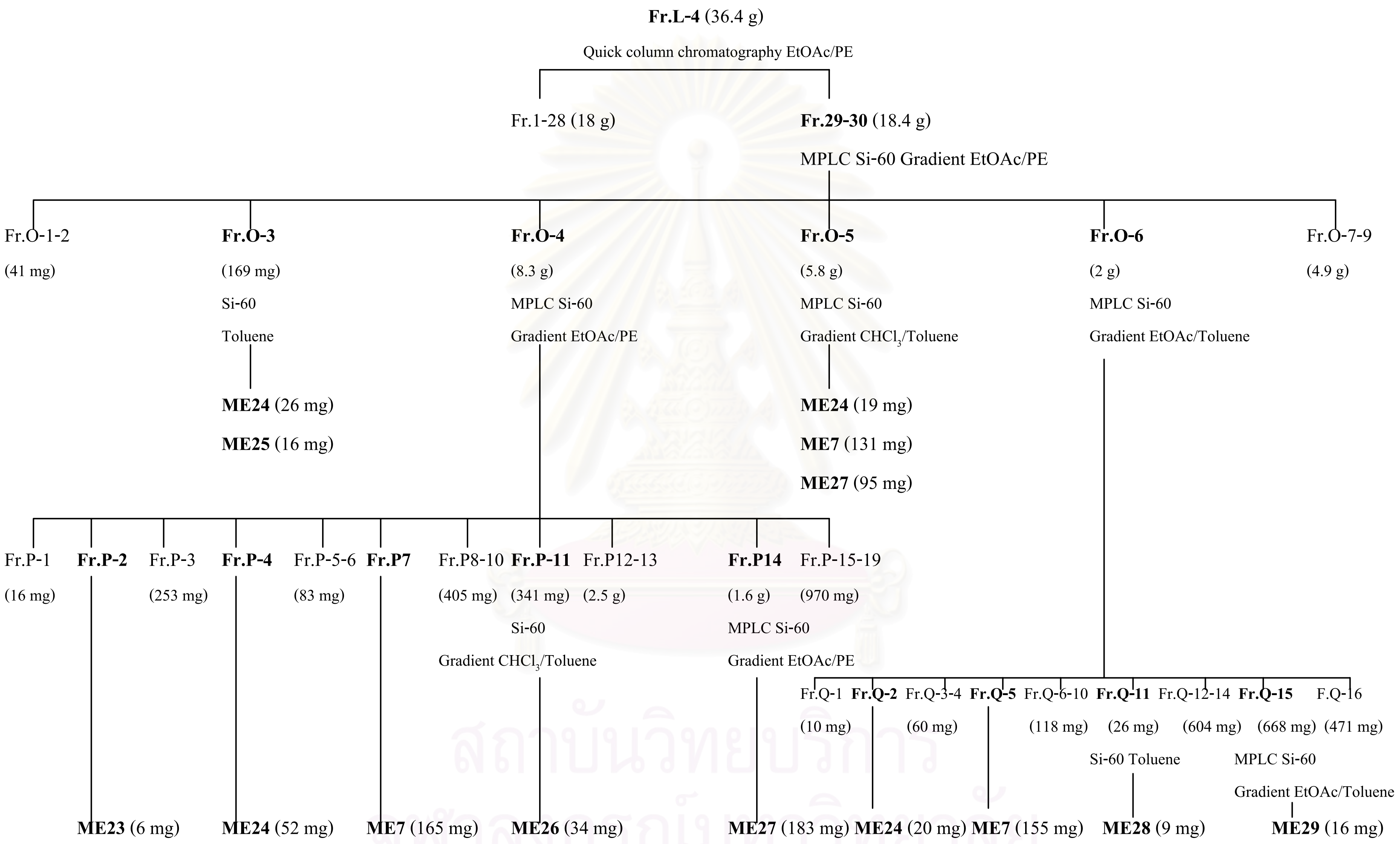
Scheme 6 Separation of fraction G-7 from the EtOAc extract of the stem bark of *M. erythrocalyx*



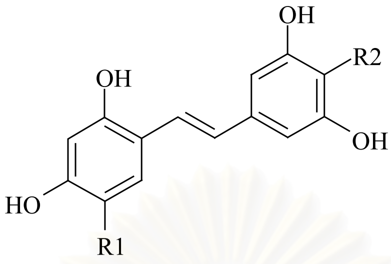
Scheme 7 Separation of the butanol extract of the stem bark of *M. erythrocalyx*



Scheme 8 Separation of the hexane extract of the roots of *M. erythrocalyx*



Scheme 9 Separation of fraction L-4 from the hexane extract of the roots of *M. erythrocalyx*



	R1	R2
AG11 [**441**]	X	H
AG12 [**442**]	H	X

X =

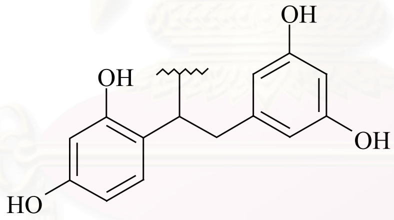
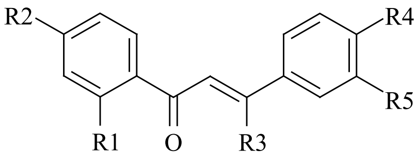
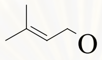
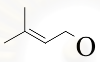
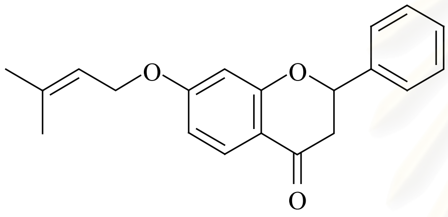
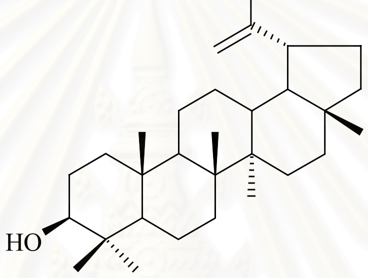


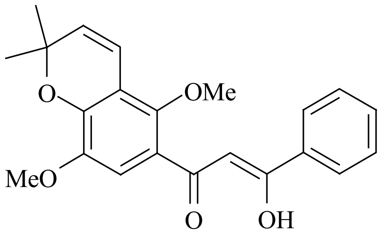
Figure 3 Structures of compounds isolated from the roots of *Artocarpus gomezianus*

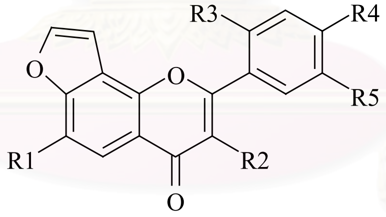


	R1	**R2**	**R3**	**R4**	**R5**
ME1 [**443**]	OH		H	H	H
ME3 [**445**]	H		H	$-OCH_2O-$	
ME7 [**280**]	OMe	OMe	OH	$-OCH_2O-$	


ME2 [**444**]


ME4 [**169**]


ME5 [**446**]



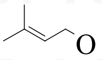
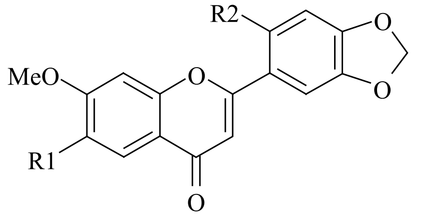
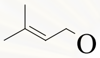
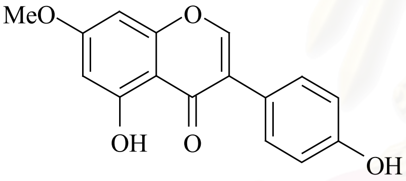
	R1	**R2**	**R3**	**R4**	**R5**
ME6 [**295**]	H	OMe	H	H	H
ME8 [**308**]		H	H	H	H
ME9 [**447**]	H	H	H	H	OMe
ME12 [**449**]	H	H	OMe	H	OMe
ME15 [**314**]	H	H	H	$-OCH_2O-$	

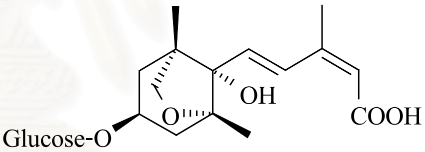
Figure 4 Structures of compounds isolated from the stem bark of *Millettia erythrocalyx*



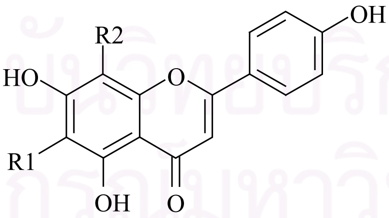
	R1	R2
ME10 [**448**]		H
ME11 [**298**]	OMe	H
ME13 [**450**]	H	OMe
ME14 [**279**]	H	H



ME16 [**451**]

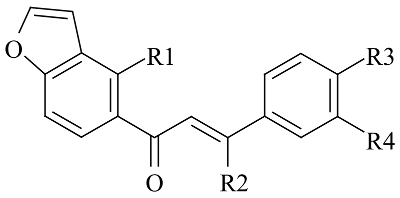


ME18 [**453**]

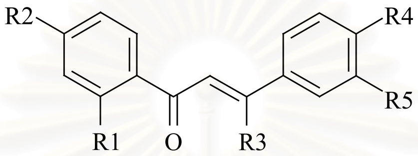


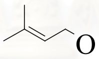
	R1	R2
ME17 [**452**]	Glucose	Glucose
ME19 [**454**]	Glucose	H

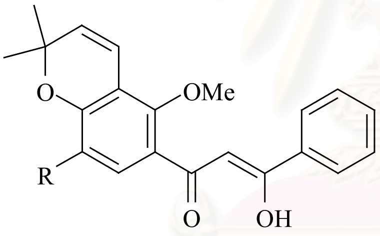
Figure 4 Structures of compounds isolated from the stembark of *Millettia erythrocalyx* (continued)



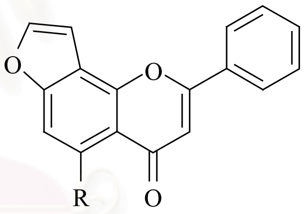
	R1	**R2**	**R3**	**R4**
ME20 [**289**]	OH	H	H	H
ME23 [**315**]	OMe	OH	H	H
ME24 [**312**]	OMe	OH	-OCH$_2$O-	



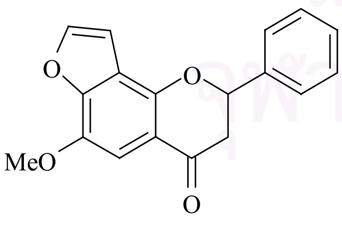
	R1	**R2**	**R3**	**R4**	**R5**
ME1 [**443**]	OH		H	H	H
ME7 [**280**]	OMe	OMe	OH	-OCH$_2$O-	
ME28 [**459**]	OMe	OMe	H	-OCH$_2$O-	



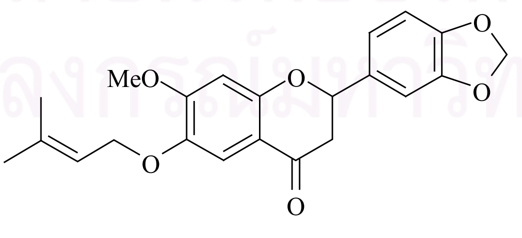
	R
ME21 [**455**]	H
ME5 [**446**]	OMe



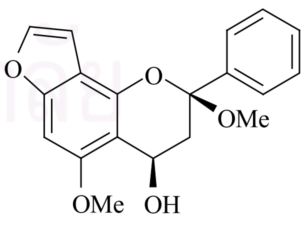
	R
ME22 [**337**]	OH
ME29 [**278**]	H



ME25 [**456**]



ME26 [**457**]



ME27 [**458**]

Figure 5 Structures of compounds isolated from the roots of *Millettia erythrocalyx*

4. Physical and Spectra data of Isolated Compounds

4.1 Compound AG11 (Artogomezianol)

Compound AG11 was obtained as yellowish amorphous solid, soluble in methanol (9 mg, 1.0 x 10^{-4}% based on dried weight of roots).

HRFABMS : $[M-H]^{-}$ *m/z* 487.1431 (calcd for $C_{28}H_{23}O_{8}$ 487.1391)

$[\alpha]^{28}_{D}$: -1.4 ° (*c* 0.48, MeOH)

UV : λ_{max} nm (log ε), in methanol; Figure 10

220 (2.35), 286 (1.93), 338 (1.96)

IR : ν_{max} cm^{-1}, Film; Figure 11

3750, 3680, 3273, 1605, 1511, 1450, 1166, 1057, 882, 852, 585

^{1}H NMR : δ ppm, 500 MHz, in DMSO-d_6; Figure 12, Table 6

^{13}C NMR : δ ppm, 125 MHz, in DMSO-d_6; Figure 13, Table 6

4.2 Compound AG12 (Andalasin A)

Compound AG12 was obtained as yellowish amorphous solid, soluble in methanol (22 mg, 2.5 x 10^{-4}% based on dried weight of roots).

HRFABMS : $[M-H]^{-}$ *m/z* 487.1419 (calcd for $C_{28}H_{23}O_{8}$ 487.1393)

$[\alpha]^{28}_{D}$: +1.8 ° (*c* 0.48, MeOH)

UV : λ_{max} nm (log ε), in methanol; Figure 18

218 (2.14), 284 (1.67), 330 (1.72)

IR : ν_{max} cm^{-1}, Film; Figure 19

3792, 3699, 3292, 1607, 1512, 1455, 1290, 1158, 1000, 83

^{1}H NMR : δ ppm, 500 MHz, in DMSO-d_6; Figure 20, Table 7

^{13}C NMR : δ ppm, 125 MHz, in DMSO-d_6; Figure 21, Table 7

4.3 Compound ME1 (Derricidin)

Compound ME1 was obtained as yellow needles, soluble in chloroform (25 mg, 1.25 x 10^{-3}% based on dried weight of stem bark and 3.5 mg, 4 x 10^{-5}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 28

308 (M^{+}, 34), 240 (100), 163 (94), 137 (68), 103 (39), 69 (96), 41 (87)

UV : λ_{max} nm (log ε), in methanol; Figure 26

251 (2.59), 321 (3.09), 344 (3.07)

IR : ν_{max} cm^{-1}, KBr disc; Figure 27

1632, 1611, 1574, 1416, 1359, 1217, 1137, 991, 966

^{1}H NMR **:** δ ppm, 500 MHz, in chloroform-*d*; Figure 29, Table 8

^{13}C NMR **:** δ ppm, 125 MHz, in chloroform-*d*; Figure 30, Table 8

4.4 Compound ME2 (7-γ,γ-Dimethylallyloxyflavanone)

Compound ME2 was obtained as a yellow powder, soluble in chloroform (1.5 mg, 7.5 x 10^{-5}% based on dried weight of stem bark).

EIMS **:** *m/z* (% relative intensity); Figure 38

308 (M^{+}, 6), 240 (100), 163 (46), 136 (42), 104 (34), 69 (66), 28 (45)

UV **:** λ_{max} nm (log ε), in methanol; Figure 36

274 (3.16), 309 (2.94)

IR **:** ν_{max} cm^{-1}, KBr disc; Figure 37

3364, 3035, 1681, 1607, 1445, 1369, 1336, 1254, 1219, 1163, 757, 700

^{1}H NMR **:** δ ppm, 300 MHz, in chloroform-*d*; Figure 39, Table 9

^{13}C NMR **:** δ ppm, 75 MHz, in chloroform-*d*; Figure 40, Table 9

4.5 Compound ME3 (2'-Hydroxy-3,4-methylenedioxy-4'-γ,γ-dimethylallyloxychalcone)

Compound ME3 was obtained as yellowish powder, soluble in chloroform (2.4 mg, 1.2 x10^{-4}% based on dried weight of stem bark).

EIMS **:** *m/z* (% relative intensity); Figure 46

352 (M^{+}, 10), 321 (8), 284 (60), 148 (44), 135 (70), 69 (88), 41 (100)

UV **:** λ_{max} nm (log ε), in methanol; Figure 44

258 (2.10), 313 (2.12), 373 (2.52)

IR **:** ν_{max} cm^{-1}, KBr disc; Figure 45

3435, 2922, 2853, 1635, 1573, 1503, 1493, 1450, 1379, 1248

^{1}H NMR **:** δ ppm, 500 MHz, in chloroform-*d* ; Figure 47, Table 10

4.6 Compound ME4 (Lupeol)

Compound ME4 was obtained as colorless needles, soluble in chloroform (64 mg, 3.2 x 10^{-3}% based on dried weight of stem bark).

IR **:** ν_{max} cm^{-1}, KBr disc; Figure 52

3322, 3068, 1456, 1383, 1262, 1190, 1103, 945, 804

^{1}H NMR **:** δ ppm, 300 MHz, in chloroform-*d*; Figure 53, Table 11

^{13}C NMR **:** δ ppm, 75 MHz, in chloroform-*d*; Figure 55, Table 11

4.7 Compound ME5 (Ponganone I)

Compound ME5 was obtained as yellow needles, soluble in chloroform (62 mg, 3.1×10^{-3}% based on dried weight of stem bark and 277 mg, 3.46×10^{-3}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 58

366 (M^+, 26), 352 (54), 335 (100), 247 (6), 230 (10), 105 (32), 77 (20)

UV : λ_{max} nm (log ε), in methanol; Figure 56

225 (3.11), 292 (2.84), 369 (3.20)

IR : ν_{max} cm^{-1}, KBr disc; Figure 57

2930, 1590, 1566, 1470, 1445, 1374, 1273, 1224, 1162, 1028

^{1}H NMR : δ ppm, 300 MHz, in acetone-d_6; Figure 59, Table 12

^{13}C NMR : δ ppm, 75 MHz, in acetone-d_6; Figure 60, Table 12

4.8 Compound ME6 (Karanjin)

Compound ME6 was obtained as a yellow powder, soluble in chloroform (1.7 mg, 8.5×10^{-5}% based on dried weight of stem bark).

EIMS : *m/z* (% relative intensity); Figure 67

292 (M^+, 92), 291 (100), 221 (32), 160 (76), 132 (22), 105 (32), 28 (19)

UV : λ_{max} nm (log ε), in methanol; Figure 65

215 (3.01), 259 (2.82), 303 (2.64)

IR : ν_{max} cm^{-1}, KBr disc; Figure 66

3059, 2965, 1628, 1529, 1464, 1444, 1372, 1342, 1260, 1166, 1037, 802, 759

^{1}H NMR : δ ppm, 500 MHz, in chloroform-d; Figure 68, Table 13

4.9 Compound ME7 (Milletenone)

Compound ME7 was obtained as yellow needles, soluble in chloroform (34 mg, 1.7×10^{-3}% based on dried weight of stem bark and 427 mg, 5.34×10^{-3}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 75

328 (M^+, 48), 297 (70), 165 (100), 164 (18), 138 (26), 65 (12), 28 (8)

UV : λ_{max} nm (log ε), in methanol; Figure 73

274 (2.94), 372 (3.45)

IR : ν_{max} cm^{-1}, KBr disc; Figure 74

3086, 2938, 1607, 1462, 1252, 1214, 835

^{1}H NMR : δ ppm, 500 MHz, in acetone-d_6; Figure 76, Table 14

^{13}C NMR : δ ppm, 125 MHz, in acetone-d_6; Figure 77, Table 14

4.10 Compound ME8 (Ovalifolin)

Compound ME8 was obtained as a yellow powder, soluble in chloroform (114 mg, 5.7 x 10^{-3}% based on dried weight of stem bark).

EIMS : *m/z* (% relative intensity); Figure 82

346 (M$^+$, 3), 278 (100), 176 (32), 102 (4), 69 (11), 18 (50)

UV : λ_{max} nm (log ε), in methanol; Figure 80

219 (3.11), 268 (2.96), 302 (2.74)

IR : ν_{max} cm^{-1}, KBr disc; Figure 81

2972, 1639, 1604, 1530, 1453, 1249, 1213, 1072, 769

^{1}H NMR : δ ppm, 500 MHz, in chloroform-*d*; Figure 83, Table 15

^{13}C NMR : δ ppm, 125 MHz, in chloroform-*d*; Figure 84, Table 15

4.11 Compound ME9 (Pongol methyl ether)

Compound ME9 was obtained as a yellow powder (154.5 mg, 7.72 x 10^{-3}% based on dried weight of stem bark), soluble in chloroform.

HREIMS : [M$^+$] at *m/z* 292.07252 (calcd for $C_{18}H_{12}O_4$ 292.07355)

EIMS : *m/z* (% relative intensity); Figure 90

292 (M$^+$, 99), 264 (44), 160 (100), 132 (79), 76 (59), 28 (74)

UV : λ_{max} nm (log ε), in methanol; Figure 88

210 (3.40), 263 (3.11), 295 (2.97)

IR : ν_{max} cm^{-1}, KBr disc; Figure 89

3437, 1641, 1530, 1437, 1406, 1216, 1072, 1051

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 91, Table 16

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 92, Table 16

4.12 Compound ME10 (Millettocalyxin B)

Compound ME10 was obtained as a yellow powder, soluble in chloroform (41 mg, 2.05 x 10^{-3}% based on dried weight of stem bark).

HREIMS : [M$^+$] at *m/z* 380.11738 (calcd for $C_{22}H_{20}O_6$ 380.12598)

EIMS : *m/z* (% relative intensity); Figure 99

380 (M$^+$, 1.7), 312 (100), 166 (20), 146 (12), 69 (18), 28 (36)

UV : λ_{max} nm (log ε), in methanol; Figure 97

329 (3.12), 240 (3.33)

IR : ν_{max} cm^{-1}, KBr disc; Figure 98

3438, 1632, 1506, 1452, 1336, 1265, 1109

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure100, Table 17

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 101, Table 17

4.13 Compound ME11 (3',4'-Methylenedioxy-6,7-dimethoxyflavone)

Compound ME11 was obtained as a yellow powder, soluble in chloroform (31 mg, 1.55x10^{-3}% based on dried weight of stem bark).

EIMS : *m/z* (% relative intensity); Figure 110

326 (M^{+}, 100), 307 (24), 180 (26), 165 (22), 109 (8), 28 (76)

UV : λ_{max} nm (log ε), in methanol; Figure 108

211(2.15), 243 (2.96), 333 (2.96)

IR : ν_{max} cm^{-1}, KBr disc; Figure 109

3082, 1636, 1505, 1476, 1453, 1334, 1262, 1086, 751

^{1}H NMR : δ ppm, 300 MHz, in DMSO-d_6; Figure 111, Table 18

^{13}C NMR : δ ppm, 75 MHz, in DMSO-d_6; Figure 112, Table 18

4.14 Compound ME12 (Millettocalyxin C)

Compound ME12 was obtained as a yellow powder, soluble in chloroform (34 mg, 1.7 x 10^{-3}% based on dried weight of stem bark).

HREIMS : [M^{+}] at *m/z* 322.08367 (calcd for $C_{19}H_{14}O_5$ 322.08414)

EIMS : *m/z* (% relative intensity); Figure 119

322 (M^{+}, 84), 162 (46), 161 (33), 160 (16), 147 (36), 119 (13), 76 (17)

UV : λ_{max} nm (log ε), in methanol; Figure 117

249 (3.59), 295 (3.28)

IR : ν_{max} cm^{-1}, KBr disc; Figure 118

3438, 1637, 1595, 1464, 1409, 1362, 1238, 1205, 1072, 741

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 120, Table 19

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 121, Table 19

4.15 Compound ME13 (Millettocalyxin A)

Compound ME13 was obtained as a yellow powder, soluble in chloroform (8 mg, 4.0 x 10^{-4}% based on dried weight of stem bark).

HREIMS : [M^+] at *m/z* 326.08335 (calcd for $C_{18}H_{14}O_6$ 326.07904)

EIMS : *m/z* (% relative intensity); Figure 128

326 (M^+, 100), 295 (38), 283 (26), 281 (8), 176 (40), 151 (36)

UV : λ_{max} nm (log ε), in methanol; Figure 126

300 (2.97), 354 (2.98)

IR : ν_{max} cm^{-1}, KBr disc; Figure 127

1616, 1567, 1504, 1259, 1199, 1169, 1093, 837

^{1}H NMR : δ ppm, 300 MHz, in acetone-d_6; Figure 129, Table 20

^{13}C NMR : δ ppm, 75 MHz, in acetone-d_6; Figure 130, Table 20

4.16 Compound ME14 (3',4'-Methylenedioxy-7-methoxyflavone)

Compound ME14 was obtained as a yellow powder, soluble in chloroform (4 mg, 2.0 x 10^{-4}% based on dried weight of stem bark).

EIMS : *m/z* (% relative intensity); Figure 136

296 (M^+, 100), 268 (26), 253 (48), 146 (72), 122 (14), 79 (14), 28 (26)

UV : λ_{max} nm (log ε), in methanol; Figure 134

207 (2.47), 237 (3.27), 332 (3.19)

IR : ν_{max} cm^{-1}, KBr disc; Figure 135

1610, 1479, 1450, 1240, 1202, 1164, 1134, 1034

^{1}H NMR : δ ppm, 300 MHz, in acetone-d_6; Figure 137, Table 21

^{13}C NMR : δ ppm, 75 MHz, in acetone-d_6; Figure 138, Table 21

4.17 Compound ME15 (Pongaglabrone)

Compound ME15 was obtained as a yellow powder, soluble in chloroform (9 mg, 4.5 x 10^{-4}% based on dried weight of stem bark).

EIMS : *m/z* (% relative intensity); Figure 143

306 (M^+, 100), 160 (66), 146 (90), 104 (12), 76 (24), 28 (31)

UV : λ_{max} nm (log ε), in methanol; Figure 141

204 (2.44), 241 (3.30), 328 (3.19)

IR : ν_{max} cm^{-1}, KBr disc; Figure 142

2921, 2851, 1640, 1592, 1502, 1449, 1347, 1256, 745

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 144, Table 22

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 145, Table 22

4.18 Compound ME16 (Prunetin)

Compound ME15 was obtained as a yellow powder, soluble in chloroform (11 mg, 5.5 x 10^{-4}% based on dried weight of stem bark).

EIMS : *m/z* (% relative intensity); Figure 152
284 (M^+, 100), 166 (20), 138 (16), 118 (12), 95 (12), 28 (32)
UV : λ_{max} nm (log ε), in methanol; Figure 150
261 (3.08)
IR : ν_{max} cm^{-1}, KBr disc; Figure 151
3380, 1665, 1614, 1570, 1515, 1440, 1358, 1258, 1196, 1053
^{1}H NMR : δ ppm, 300 MHz, in acetone-d_6 ; Figure 153, Table 23
^{13}C NMR : δ ppm, 75 MHz, in acetone-d_6 ; Figure 154, Table 23

4.19 Compound ME17 (Vicenin II)

Compound ME17 was obtained as a yellow powder, soluble in pyridine (195 mg, 9.75 x 10^{-3}% based on dried weight of stem bark).

ESIMS : $[M+Na]^+$ *m/z* 617.5 (positive ion mode); Figure 161
$[M-H]^-$ *m/z* 592.8 (negative ion mode); Figure 162
UV : λ_{max} nm (log ε), in methanol; Figure 159
215 (2.98), 270 (2.74), 325 (2.66)
IR : ν_{max} cm^{-1}, KBr disc; Figure 160
3382, 1628, 1577, 1437, 1362, 1210, 1101, 1082
^{1}H NMR : δ ppm, 300 MHz, in pyridine-d_5; Figure 163-165, Table 24
^{13}C NMR : δ ppm, 75 MHz, in pyridine-d_5; Figure 166, Table 24

4.20 Compound ME18 (Dihydrophaseic acid-4'-*O*-β-D-glucopyranoside)

Compound ME18 was obtained as gum, soluble in methanol (25 mg, 1.25 x 10^{-3}% based on dried weight of stem bark).

ESIMS : $[M+Na]^+$ *m/z* 467.4 (positive ion mode); Figure 178
$[M-H]^-$ *m/z* 443.4 (negative ion mode); Figure 177
UV : λ_{max} nm (log ε), in methanol; Figure 175
257 (2.27)
IR : ν_{max} cm^{-1}, KBr disc; Figure 176
3374, 2934, 1688, 1603, 1380, 1164, 1076

^{1}H NMR : δ ppm, 300 MHz, in methanol-d_4; Figure 179-180, Table 25

^{13}C NMR : δ ppm, 75 MHz, in methanol-d_4; Figure 181, Table 25

4.21 Compound ME19 (Isovitexin)

Compound ME19 was obtained as a yellow powder, soluble in methanol (3 mg, 1.5 x 10^{-4}% based on dried weight of stem bark).

ESIMS : $[M+H]^+$ *m/z* 433.2 (positive ion mode); Figure 192

$[M-H]^-$ *m/z* 431.4 (negative ion mode); Figure 191

UV : λ_{max} nm (log ε), in methanol; Figure 189

211 (2.69), 269 (2.35), 329 (2.34)

IR : ν_{max} cm^{-1}, KBr disc; Figure 190

3369, 1654, 1612, 1569, 1363, 1283, 1245, 1179, 1092

^{1}H NMR : δ ppm, 300 MHz, in pyridine-d_5 ; Figure 193, Table 26

^{13}C NMR : δ ppm, 75 MHz, in pyridine-d_5; Table 26

4.22 Compound ME20 (1-(4-Hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one)

Compound ME20 was obtained as yellow needles, soluble in chloroform (11 mg, 1.37 x 10^{-4}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 200

264 (M^+, 56), 187 (24), 161 (26), 160 (100), 103 (16), 77 (24), 51 (12)

UV : λ_{max} nm (log ε), in methanol; Figure 198

214 (3.02), 249 (2.84), 263 (2.84)

IR : ν_{max} cm^{-1}, KBr disc; Figure 199

2956, 2925, 1641, 1599, 1470, 1378, 1298, 1131, 793

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 201, Table 27

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 202, Table 27

4.23 Compound ME21 (Purpurenone)

Compound ME21 was obtained as yellow needles, soluble in chloroform (14 mg, 1.75 x 10^{-4}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 209

336 (M^+, 64), 321 (100), 305 (96), 217 (26), 201 (66), 175 (54), 105 (58), 77 (64)

UV : λ_{max} nm (log ε), in methanol; Figure 207

207 (2.40), 359 (3.70)

IR : ν_{max} cm^{-1}, KBr disc; Figure 208
3061, 2975, 1636, 1592, 1460, 1280, 1219, 1166, 1115, 1027, 778

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 210, Table 28

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 211, Table 28

4.24 Compound ME22 (Pongaglabol)

Compound ME22 was obtained as a yellow powder, soluble in chloroform (21 mg, 2.62 x 10^{-4}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 218
278 (M$^+$, 100), 250 (18), 176 (94), 139 (32), 120 (46), 102 (30), 92 (60)

UV : λ_{max} nm (log ε), in methanol; Figure 216
217 (3.22), 280 (3.21)

IR : ν_{max} cm^{-1}, KBr disc; Figure 217
2924, 1666, 1614, 1428, 1318, 1249, 1141, 1108, 798

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 219, Table 29

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 220, Table 29

4.25 Compound ME23 (Pongamol)

Compound ME23 was obtained as a yellow powder, soluble in chloroform (60 mg, 7.5 x 10^{-4}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 226
294 (M$^+$, 30), 263 (100), 175 (70), 160 (24), 105 (36), 77 (38), 51 (14)

UV : λ_{max} nm (log ε), in methanol; Figure 224
237 (2.63), 347 (2.60)

IR : ν_{max} cm^{-1}, KBr disc; Figure 225
1599, 1565, 1297, 1263, 1219, 1061, 973, 775

^{1}H NMR : δ ppm, 500 MHz, in chloroform-*d*; Figure 227, Table 30

^{13}C NMR : δ ppm, 125 MHz, in chloroform-*d*; Figure 228, Table 30

4.26 Compound ME24 (Ovalitenone)

Compound ME24 was obtained as a yellow powder, soluble in chloroform (97 mg, 1.21 x 10^{-3}% based on dried weight of roots).

HREIMS : [M$^+$] at *m/z* 338.07878 (calcd for $C_{19}H_{14}O_6$ 338.07904)

EIMS : *m/z* (% relative intensity); Figure 234

338 (M^+, 38), 307 (100), 175 (82), 150 (32), 149 (56), 121 (16), 89 (14), 69 (22)

UV : λ_{max} nm (log ε), in methanol; Figure 232
237 (3.01), 361 (2.95)

IR : ν_{max} cm^{-1}, KBr disc; Figure 233
3440, 3078, 3295, 1600, 1473, 1292, 1066, 1038

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 235, Table 31

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 236, Table 31

4.27 Compound ME25 ((-)-(2*S*)-6-Methoxy-[2",3":7,8]-furanoflavanone)

Compound ME25 was obtained as colorless needles, soluble in chloroform (16 mg, 2.0 x 10^{-4}% based on dried weight of roots).

HREIMS : [M^+] at *m/z* 294.08423 (calcd for $C_{18}H_{14}O_4$ 294.08920)

EIMS : *m/z* (% relative intensity); Figure 244
294 (M^+, 48), 190 (100), 119 (30), 103 (6), 77 (10), 28 (12)

$[\alpha]^{28}_{D}$:-55.8 ° (*c* 0.1; MeOH)

CD : $[\theta]_{215}$ –4821, $[\theta]_{226.5}$ +1538, $[\theta]_{233}$ –166, $[\theta]_{244}$ +4492, $[\theta]_{281}$ –9871, $[\theta]_{312}$ –12850, $[\theta]_{350}$ +12398; (*c* 0.1, MeOH); Figure 245

UV : λ_{max} nm (log ε), in methanol; Figure 242
234 (3.44), 247 (3.32), 341 (2.65)

IR : ν_{max} cm^{-1}, KBr disc; Figure 243
1681, 1483, 1378, 1225, 1212, 1108, 1082, 779

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 246, Table 32

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 247, Table 32

4.28 Compound ME26 (Ponganone V)

Compound ME26 was obtained as colorless needles, soluble in chloroform (34 mg, 4.25 x 10^{-4}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 254
382 (M^+, 4), 314 (90), 166 (76), 148 (100), 69 (36), 41 (32), 28 (26)

UV : λ_{max} nm (log ε), in methanol; Figure 252
240 (2.21), 277 (1.97), 338 (1.72)

IR : ν_{max} cm^{-1}, KBr disc; Figure 253
1677, 1611, 1503, 1449, 1252, 1179, 1036, 992

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 255, Table 33

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 256, Table 33

4.29 Compound ME27 (2,5-Dimethoxy-4-hydroxy-[2'',3'':7,8]-furanoflavan)

Compound ME27 was obtained as colorless crystals, soluble in chloroform (278 mg, 3.47 x 10^{-3}% based on dried weight of roots).

HREIMS : [M^{+}] at *m/z* 326.11934 (calcd for $C_{19}H_{18}O_{5}$ 326.11542)

EIMS : *m/z* (% relative intensity); Figure 263

326 (M^{+}, 20), 277 (65), 192 (100), 174 (50), 146 (60), 134 (10), 105 (32), 77 (36)

$[\alpha]^{28}_{D}$: +42.1 ° (*c* 0.3; MeOH)

CD : $[\theta]_{209.5}$ –1556, $[\theta]_{223}$ +212, $[\theta]_{233}$ –873, $[\theta]_{248.5}$ +1703, $[\theta]_{282}$ +863; (*c* 0.3; MeOH); Figure 264

UV : λ_{max} nm (log ε), in methanol; Figure 261

214 (3.15), 250 (2.68)

IR : ν_{max} cm^{-1}, KBr disc; Figure 262

3533, 3447, 3059, 1631, 1606, 1441, 1315, 1278, 1169, 1113, 1095, 994

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 265, Table 34

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 266, Table 34

4.30 Compound ME28 (3,4-Methylenedioxy-2',4'-dimethoxychalcone)

Compound ME28 was obtained as yellow powder, soluble in chloroform (9 mg, 1.12 x 10^{-4}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 273

312 (M^{+}, 98), 297 (30), 284 (45), 165 (100), 147 (26), 135 (74), 122 (44), 89 (80)

UV : λ_{max} nm (log ε), in methanol; Figure 271

351 (2.91)

IR : ν_{max} cm^{-1}, KBr disc; Figure 272

1653, 1602, 1502, 1440, 1419, 1359, 1252, 1214, 1032, 758

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 274, Table 35

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 275, Table 35

4.31 Compound ME29 (Lanceolatin B)

Compound ME29 was obtained as a colorless neddles, soluble in chloroform (16 mg, 2.0 x 10^{-4}% based on dried weight of roots).

EIMS : *m/z* (% relative intensity); Figure 282

262 (M^+, 77), 234 (12), 192 (66), 160 (100), 76 (28), 28 (38)

UV : λ_{max} nm (log ε), in methanol; Figure 280

215 (2.97), 263 (2.84), 297 (2.72)

IR : ν_{max} cm^{-1}, KBr disc; Figure 281

1645, 1605, 1494, 1404, 1361, 1215, 1116, 767

^{1}H NMR : δ ppm, 300 MHz, in chloroform-*d*; Figure 283, Table 36

^{13}C NMR : δ ppm, 75 MHz, in chloroform-*d*; Figure 284, Table 36

5. Determination of Tyrosinase Inhibitory Activity

In this study, tyrosinase inhibitory activity is determined by the dopachrome method using L-DOPA as the substrates (Iida *et al*., 1995). Dopachrome is one of the intermediate substances in the melanin biosynthesis. The red color of dopachome can be detected by visible light. In this experiment a microplate reader (BIO-RAD, model 450) with 492 nm interference filter was used for detection. The potential tyrosinase inhibitor would show minimal dopachrome absorption. This method was modified from the methods of Masamoto (Masamoto *et al*., 1980), Iida (Iida *et al*., 1995) and Morita (Morita *et al*., 1994).

5.1 Preparation of the Reaction Mixture

5.1.1 Preparation of 20 mM Phosephate buffer (pH 6.8)

Solution A: $NaH_2PO_4 \cdot 2H_2O$ (312 mg) was dissolved in 100 mL of H_2O.

Solution B: Na_2HPO_4 (284 mg) was dissolved in 100 mL of H_2O.

Then, solutions A and B were mixed until pH 6.8 was reached.

5.1.2 Preparation of 0.85 mM L-DOPA

L-DOPA (0.8 mg) was dissolved in 5 ml of 20 mM phosphate buffer (pH 6.8).

5.1.3 Preparation of Tyrosinase Solution

Tyrosinase enzyme (0.5 mg) was dissolved in 5 ml of 20 mM phosphate buffer (pH 6.8)

5.1.4 Preparation of the Test Sample

One mg of the test compound was dissolved in 3 ml ethanol (or suitable solvent) and diluted with ethanol until a suitable range of concentrations (mg/mL) was obtained. The concentration was express as μM in the final calculation. For example, the concentration of compound AG11 (MW 488) at 1 mg/3 mL was equal to 683 μM (1 mg/3 mL x 488 = 683 μM). For each well, 20 μL of test solution was added to the reaction mixture to furnish the total volume of 200 μL. The final concentration was calculated by the formula below.

$$N_1V_1 = N_2V_2$$

N_1 = Beginning concentration (μM)
V_1 = Beginning volume (μL)
N_2 = Final concentration (μM)
V_2 = Final volume (μL)

Thus, final concentration of AL1 solution = 683 μM x 20 μL / 200 μL
= 68.3 μM

5.2 Measurement of Activity

The reaction mixture (200 μL) was measured in four wells (A, B, C and D). In each well, the substance was added in the order of mixing, as follows;

A (control)	20 μL of mushroom tyrosinase solution (48 unit/mL)
	140 μL of 20 mM phosphate buffer (pH 6.8)
	20 μL of ethanol
B (blank of A)	160 μL of 20 mM phosphate buffer (pH 6.8)
	20 μL of ethanol
C (test sample)	20 μL of mushroom tyrosinase solution (48 unit/mL)
	140 μL of 20 mM phosphate buffer (pH 6.8)
	20 μL of test sample in ethanol
D (blank of C)	160 μL of 20 mM phosphate buffer (pH 6.8)
	20 μL of test sample in ethanol

After each well was mixed and preincubated at 25 °C for 10 minutes, 20 μL of 0.85 μM L-DOPA was added, and the mixture was incubated at 25 °C for 20 min. The absorbance of each well was measured at 492 nm with the microplate reader both before and after incubation.

5.3 Calculation of the Percent Inhibition of Tyrosinase Enzyme

The percent inhibition of tyrosinase reaction was calculated as follows.

$$\text{\% Tyrosinase inhibition} = \left[\frac{(A-B)-(C-D)}{A-B} \right] \text{x } 100$$

A : The difference of optical density before and after incubation at 492 nm without test sample

B : The difference of optical density before and after incubation at 492 nm without test sample and enzyme

C : The difference of optical density before and after incubation at 492 nm with test sample

D : The difference of optical density before and after incubation at 492 nm with test sample, but without enzyme

5.4 Calculation of IC_{50}

After the % tyrosinase inhibition of the test solution in each concentration was obtained, a graph showing concentration against % tyrosinase inhibition was plotted. The IC_{50} (concentration at 50% tyrosinase inhibition) of each pure compound was then obtained from the graph.

6. Determination of Anti-Herpes Simplex Activity

6.1 Antiviral Activity Assay (Lipipun, *et al.*, 2000; Abou-karam and Shier, 1990)

Antiviral activity against HSV-1 (KOS) and HSV-2 (186) was evaluated using the plaque reduction assay (Inactivation). Briefly, virus (30 PFU/25 μL) was mixed with 25 μL of complete medium containing various concentrations of test compound and then incubated at 37 °C for 1 hr. After incubation, the mixtures were added into Vero cell (6 x 10^5 cells/well) in 96-well microtiter plates and incubated at 37 °C for 2 hr. The overlay medium containing the various concentrations of test compound was added to the Vero cells and incubated at 37 °C in humidified CO_2 incubator for 2 days. Then, virus growth inhibition was evaluated by counting the virus plaque forming on Vero cells compared with the controls. The cells also were stained with 1% crystal violet in 10% formalin for 1 hr. The percent plaque inhibition was determined. Acyclovir was used as positive control.

For ED_{50} evaluation (effective dose at 50% inhibition of virus growth), the graph between the values of each concentration and its % plaque inhibition was plotted. The ED_{50} of the pure compound was then obtained from the graph.

6.2 Cytotoxicity Test

Cytotoxicity was evaluated by incubating Vero cell monolayers with complete medium containing various dilutions of extract for 72 hr at 37 °C. Then the cell cytotoxicity was examined by microscopic observation. The maximal non-cytotoxic concentration of the extract was used for the antiviral activity study.

7. Determination of Free Radical Scavenging Activity

7.1 TLC Screening Assay (Takao *et al*., 1994)

The samples were spotted and developed on a TLC plate with suitable developing solvent. After drying, the TLC plate was sprayed with 0.2% solution of 1,1-diphenyl-2-picrylhydrazyl (DPPH) in methanol. After 30 min, active components appear as yellow spots against purple background.

7.2 Free Radical Scavenging Activity Assay (Braca *et a*l., 2002)

7.2.1 Preparation of the Test Sample

Test samples were prepared as previously described in section 5.1.4.

7.2.2 Preparation of the DPPH Solution (100 μM)

Two mg of DPPH was dissolved in 100 mL of EtOH, and the solution was subsequently stirred for 30 min.

7.2.3 Measurement of Activity

The test sample (20 μL) was added to 180 μL of DPPH solution (100 μM) in a 96-well microtiter plate. The reaction mixture was incubated at 37 °C for 30 min and then the absorbance of each well was measured at 510 nm. The DPPH solution was used as negative control and quercetin as reference compound.

7.2.4 Calculation of Percentage of Free Radical Scavenging Activity

The percentage of scavenging activity was calculated as follows.

$$\%\ \text{DPPH reduction} = \left[\frac{A - B}{A} \right] \times 100$$

A: The absorbance of DPPH solution after incubation at 510 nm

B: The absorbance of the reaction mixture after incubation at 510 nm

For IC_{50} evaluation of pure compounds, a graph showing concentration versus %DPPH reduction was plotted. The IC_{50} was then calculated from the graph.

CHAPTER IV

RESULTS AND DISCUSSION

The methanol extract (100 g) from the roots of *Artocarpus gomezianus* Wall. ex Tréc. was separated using several chromatographic techniques to afford two pure compounds (AG11 and AG12).

The dried stem bark of *Millettia erythrocalyx* Gagnep. (2 kg) was extracted with ethyl acetate and methanol to give an ethyl acetate extract (37 g) and a methanol extract (164 g). The methanol extract was then partitioned with ethyl acetate and water. The aqueous fraction was shaken with butanol to give a butanol extract (29 g) and an aqueous extract (90 g). By repetitive chromatography, nineteen compounds (ME1-ME19) were obtained from the ethyl acetate and butanol extracts.

The dried roots of *Millettia erythrocalyx* Gagnep. (8 kg) were extracted with hexane, ethyl acetate and methanol, successively, to give a hexane extract (91 g), an ethyl acetate extract (87 g) and a methanol extract (429 g), respectively. The hexane extract was further purified using several chromatographic techniques to yield thirteen pure compounds (ME1, ME5, ME7, and ME20-ME29).

The structures of all isolates were determined by interpretation of their UV, IR, NMR and MS data, and further confirmed by comparison with literature values.

1. Structure Determination of Isolated Compounds

1.1 Structure Determination of Compound AG11

Compound AG11 was obtained as a yellowish amorphous solid. A molecular formula of $C_{28}H_{24}O_8$ was deduced from its $[M-H]^-$ ion at *m/z* 487.1431 (calcd for $C_{28}H_{23}O_8$ 487.1393) in the HRFABMS. The UV absorptions (Figure 10) at 286 and 338 nm were characteristics of a stilbene skeleton (Gorham, 1995). The IR spectrum (Figure 11) showed the presence of aromatic and olefinic structures (1605 and 1511 cm^{-1}) with hydroxyl groups (3750-3273 cm^{-1}).

The ^1H NMR spectrum of AG11 (Figure 12 and Table 6) displayed 13 aromatic and olefinic protons at δ 7.20-5.90, together with three aliphatic protons at δ 4.70 (1H) and 2.95 (2H), reminiscent of a dimeric structure comprising a stilbene unit and a dihydrostilbene moiety linked by a C-C (sp^2-sp^3) bond (Lin *et al*., 1992). In the ^1H NMR spectrum, the stilbene monomeric part displayed proton signals similar to those of oxyresveratrol ([**174**]; 2,4,3',5'-tetrahydroxystilbene) (Sritularak, 1998), except for the absence of the H-5 resonance, with two *trans*-olefinic proton signals at δ 7.08 (1H, d, *J* = 16.2 Hz, H-α) and 6.63 (1H, d, *J* = 16.2 Hz, H-β) and three aromatic protons resonances at δ 6.31 (2H, d, *J* =

2.1 Hz, H-2' and H-6') and 6.05 (1H, dd, *J* = 2.1, 2.1 Hz, H-4'). In support of this, C-5 of this compound was shown in the ^{13}C NMR (Figure 13) and DEPT spectra to be a quaternary sp^{2} carbon at δ 122.5, in contrast with that of oxyresveratrol [**174**], which appeared as a methine sp^{2} carbon at δ 107.4. These obsearvations suggested that this stilbene monomer was derived from oxyresveratrol [**174**]. The argument was further corroborated by the upfield shifts for the carbons in the *ortho* (2.8 and 0.9 ppm for C-4 and C-6, respectively) and *para* positions (2.4 ppm for C-2) in relation to C-5, as compared with their counterparts in oxyresveratrol [**174**].

The ^{1}H-^{1}H COSY spectrum of compound AG11 revealed the following signals for the dihydrostilbene substituent: six aromatic protons at δ 6.85 (1H, d, *J* = 8.5 Hz, H-6"), 6.21 (1H, d, *J* = 2.4 Hz, H-3"), 6.10 (1H, dd, *J* = 8.5, 2.4 Hz, H-5"), 6.09 (2H, d, *J* = 2.1 Hz, H-2''' and H-6'''), 5.90 (1H, dd, *J* = 2.1, 2.1 Hz, H-4'''); one methine proton at δ 4.70 (1H, dd, *J* = 7.9, 7.9 Hz, H-α'); and two methylene protons at δ 2.95 (2H, d, J = 7.9 Hz, H$_{2}$-β'). The ^{13}C NMR, DEPT, and HMQC spectra (Figure 14) displayed the C-α' and C-β' resonances at δ 36.7 and 40.0, respectively. From these spectral data, it could be inferred that the second monomer was a dihydro derivative of oxyresveratrol [**174**], i.e., dihydrooxyresveratrol, in which one of its aliphatic carbons (C-α') was connected to C-5 of the first monomer. Confirmation of the proposed structure was obtained from the long range C-H couplings observed in the HMBC spectrum (Figures 15-17). The C-6 carbon showed three-bond correlations with H-α and H-α', confirming the involvement of C-5 and C-α' of the first and the second units in the interstilbenoid C-C linkage. The other important three-bond couplings were found between H-α' and H-6", and H-6''' and C-β'. In addition, two-bond couplings between H-α' and C-5, and H-α' and C-β', were displayed. Compound AG11 is a new compound, and have been given the name artogomezianol [**441**] (Likhitwitayawuid and Sritularak, 2001).

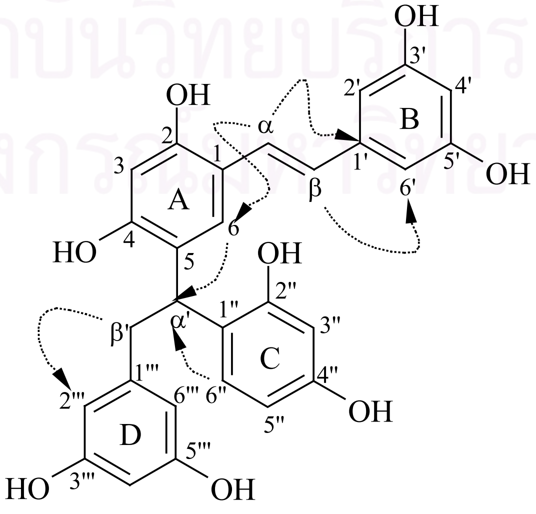


Figure 6 Selected HMBC correlations of compound AG11

Table 6 NMR Spectral data of compound AG11 as compared with oxyresveratrol (DMSO-d_6)

position	Compound AG11		Oxyresveratrol		HMBC (correlation with ^{1}H)
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	
1	-	114.3 (s)	-	115.4 (s)	H-3 and H-β
2	-	153.7 (s)	-	156.1 (s)	H-6 and H-α
3	6.30 (s)	102.6 (d)	6.33 (d, 2.4)	102.7 (d)	-
4	-	155.4 (s)	-	158.5 (s)	H-6
5	-	122.5 (s)	6.25 (dd, 8.4, 2.4)	107.4 (d)	H-3, H-β' and H-α'*
6	7.20 (s)	126.4 (d)	7.34 (d, 8.4)	127.3 (d)	H-α and H-α'
α	7.08 (d, 16.2)	124.0 (d)	7.15 (d, 16.5)	123.3 (d)	H-6
β	6.63 (d, 16.2)	124.2 (d)	6.77 (d, 16.5)	124.7 (d)	H-2' and H-6'
1'	-	140.2 (s)	-	140.1 (s)	H-α and H-β*
2'	6.31 (d, 2.1)	103.9 (d)	6.35 (d, 1.8)	104.2 (d)	H-β, H-6' and H-4'
3'	-	157.8 (s)	-	158.5 (s)	H-4'*
4'	6.05 (dd, 2.1, 2.1)	101.3 (d)	6.08 (br s)	101.5 (d)	H-2' and H-6'
5'	-	157.8 (s)	-	158.5 (s)	H-4'*
6'	6.31 (d, 2.1)	103.9 (d)	6.35 (d, 1.8)	104.2 (d)	H-β, H-2' and H-4'
1''	-	121.6 (s)			H-3'', H-5'', H-β' and H-α'*
2''	-	155.6 (s)[a]			H-6'' and H-α'
3''	6.21 (d, 2.4)	102.5 (d)			H-5''
4''	-	155.9 (s)[a]			H-6''
5''	6.10 (dd, 8.5, 2.4)	105.6 (d)			H-3''
6''	6.85 (d, 8.5)	128.8 (d)			H-α'
α'	4.70 (dd, 7.9, 7.9)	36.7 (d)			H-6, H-6'' and H-β'*
β'	2.95 (d, 7.9)	40.0 (t)			H-2''', H-6''' and H-α'*
1'''	-	143.8 (s)			H-α'
2'''	6.09 (d, 2.1)	107.0 (d)			H-6''', H-4''' and H-α'
3'''	-	158.5 (s)			H-2'''* and H-4'''*
4'''	5.90 (dd, 2.1, 2.1)	99.9 (d)			H-2''' and H-6'''
5'''	-	158.5 (s)			H-4''' * and H-6'''*
6'''	6.09 (d, 2.1)	107.0 (d)			H-2''', H-4''' and H-β'

[a] Interchangeable assignments.

*Two-bond coupling.

1.2 Structure Determination of Compound AG12

Compound AG12 was obtained as an amorphous solid. The UV spectrum (Figure 18) showed absorptions similar to those of AG11 at 218, 284 and 330 nm, suggestive of a stilbene nucleus. The IR spectrum (Figure 19) exhibited absorption bands at 1512 cm^{-1} (olefinic), 1607 (aromatic), and 3792 cm^{-1} (hydroxyl). It has a molecular formula of $C_{28}H_{24}O_8$, as indicated by the $[M-H]^-$ ion at *m/z* 487.1419 (calcd for $C_{28}H_{23}O_8$ 487.1393) in the HRFABMS, suggesting that it was an isomeric stilbene dimer of AG11.

The ^{1}H NMR spectrum (Table 7 and Figure 20) showed eleven aromatic protons at δ 5.89-7.31 and two *trans*-olefinic protons at δ 7.07 (1H, d, J = 16.4 Hz, H-α) and 6.66 (1H, d, J = 16.4 Hz, H-β), along with three aliphatic protons at δ 4.83 (1H), 3.42 (1H) and 3.03 (1H), indicating the connection of the oxyresveratrol [**174**] and the dihydrooxyresveratrol units by a C-C (sp^2-sp^3) bond. The ^{1}H-^{1}H COSY spectrum (Figure 22) exhibited proton signals for the dihydrooxyresveratrol moiety, with the presence of six aromatic protons at δ 7.07 (1H, d, J = 8.4 Hz, H-6''), 6.19 (1H, d, J = 2.4 Hz, H-3''), 6.06 (1H, dd, J = 8.4, 2.4 Hz, H-5''), 6.07 (2H, d, J = 2.1 Hz, H-2''' and H-6'''), 5.89 (1H, t, J = 2.1 Hz, H-4'''); one methine proton at δ 4.83 (1H, dd, J = 9.7, 5.7 Hz, H-α'); and two methylene protons at δ 3.03 (1H, dd, J = 13.4, 5.7 Hz, H-β') and 3.42 (1H, dd, J = 13.4, 9.7 Hz, H-β').

The ^{13}C NMR (Figure 21) and HSQC (Figure 23) spectra showed the C-α' and C-β' resonances at δ 35.5 and 38.0, respectively. AG12 differed from AG11 by the ^{1}H NMR signals for oxyresveratrol unit. In AG12, the dihydrooxyresveratrol unit was linked to ring B, of the oxyresveratrol moiety, as evident from the ABM proton spin system at δ 6.30 (1H, d, J = 2.3 Hz, H-3), δ 6.22 (1H, dd, J = 8.5, 2.3 Hz, H-5) and δ 7.31 (1H, d, J = 8.5 Hz, H-6). The disappearance of the H-4' signal, together with the presence of a broad singlet signal at δ 6.36 (2H, H-2' and H-6') indicated the two monomers should be linked at C-4' and C-α'. This was supported by the three-bond correlations of C-4' with H_2-β' and C-3' and C-5' with H-α' in the HMBC spectrum (Figures 24-25). The other important ^3J-couplings were observed between H-α' and C-6'', and H-2''' (H-6''') and C-β'. Furthermore, two-bond correlations of H-α' with C-4', and of H-α' and C-β', were also displayed.

Through analysis of the above spectral data and comparison with earlier reported ^{1}H and ^{13}C NMR data (Syah *et al*., 2000), compound AG12 was identified as andalasin A [**442**], a compound previously reported as a new stilbene dimer from *Morus macroura*.

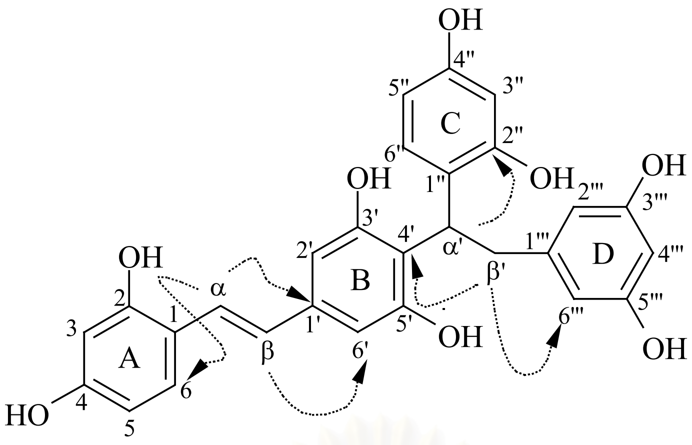


Figure 7 Selected HMBC correlations of compound AG12

Table 7 NMR Spectral data of compound AG12 (DMSO-d_6) and andalasin A (acetone-d_6)

position	Compound AG12		Andalasin A		HMBC (correlation with ^{1}H)
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	^{13}C	
1	-	115.4 (s)	-	117.1	H-3, H-5 and H-β
2	-	155.9 (s)	-	156.7	H-6 and H-α
3	6.30 (d, 2.3)	102.6 (d)	6.40 (d, 2.3)	103.4	H-5, HO-2 and HO-4
4	-	158.1 (s)	-	158.9	H-6 and HO-4*
5	6.22 (dd, 8.5, 2.3)	107.3 (d)	6.35 (dd, 8.3, 2.3)	108.3	H-3 and HO-4
6	7.31 (d, 8.5)	126.9 (d)	7.35 (d, 8.3)	128.0	H-α
α	7.07 (d, 16.4)	122.4 (d)	7.23 (d, 16.4)	123.7	H-6
β	6.66 (d, 16.4)	124.3 (d)	6.77 (d, 16.4)	125.6	H-2', H-6' and H-α*
1'	-	136.6 (s)	-	138.5	H-α and H-β*
2'	6.36 (br s)	104.8 (d)	6.53 (br s)	106.4	H-6' and H-β
3'	-	156.3 (s)	-	156.7	H-α'
4'	-	115.4 (s)	-	116.9	H-2', H-6', H-β' and H-α'*
5'	-	156.3 (s)	-	156.7	H-α'
6'	6.36 (br s)	104.8 (d)	6.53 (br s)	106.4	H-2' and H-β
1''	-	121.5 (s)	-	121.8	H-3'', H-5'', H-β' and H-α'*

Table 7 (continued)

position	Compound AG12		Andalasin A		HMBC (correlation with ^{1}H)
	^{1}H (mult., J in Hz)	^{13}C (mult.)	^{1}H (mult., J in Hz)	^{13}C	
2''	-	155.2 (s)	-	157.1	H-6'' and H-α'
3''	6.19 (d, 2.4)	102.3 (d)	6.28 (d, 2.5)	103.3	H-5'' and HO-4''
4''	-	155.8 (s)	-	156.1	H-6'' and HO-4''*
5''	6.06 (dd, 8.4, 2.4)	105.6 (d)	6.29 (dd, 8.2, 2.5)	107.3	H-3'' and HO-4''
6''	7.07 (d, 8.4)	129.7 (d)	7.48 (d, 8.2)	130.7	H-α'
α'	4.83 (dd, 9.7, 5.7)	35.5 (d)	4.86 (dd, 8.5, 6.8)	36.4	H-6'' and H-β'*
β'	3.42 (dd, 13.4, 9.7)	38.0 (t)	3.67 (dd, 13.7, 8.5)	38.2	H-2''', H-6''' and H-α'*
	3.03 (dd, 13.4, 5.7)	-	3.38 (dd, 13.7, 6.8)	-	-
1'''	-	144.6 (s)	-	145.2	H-α'
2'''	6.07 (d, 2.1)	107.0 (d)	6.26 (d, 2.2)	108.2	H-6''', H-4''' and H-β'
3'''	-	157.7 (s)	-	158.8	H-2'''* and H-4'''*
4'''	5.89 (t, 2.1)	99.9 (d)	6.07 (dd, 2.2, 2.2)	100.9	H-2''' and H-6'''
5'''	-	157.7 (s)	-	158.8	H-4'''* and H-6'''*
6'''	6.07 (d, 2.1)	107.0 (d)	6.26 (d, 2.2)	108.2	H-2''', H-4''' and H-β'
HO-2	9.51 (s)	-			
HO-4	9.33 (s)	-			
HO-3', 5'	9.08 (s)	-			
HO-4''	8.83 (s)	-			
HO-3''', 5'''	8.80 (s)	-			

*Two-bond coupling.

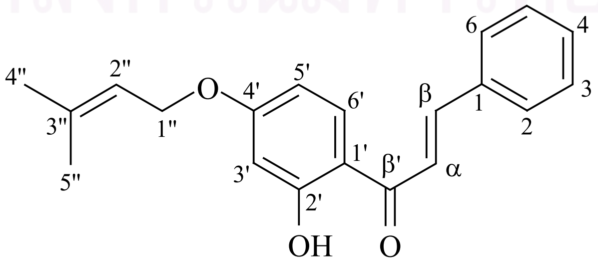
1.3 Structure Determination of Compound ME1

Compound ME1 showed [M$^+$] at *m/z* 308 in the EI mass spectrum (Figure 28), corresponding to the molecular formula $C_{20}H_{20}O_3$. The UV absorptions at 321 and 344 nm (Figure 26) and the IR absorptions at 1632 (conjugated C=O) and 1611 (conjugated C=C) cm^{-1} (Figure 27) were suggestive of a chalcone skeleton (Markham, 1982).

The ^{1}H NMR spectrum (Table 8 and Figure 29) revealed the presence of two *trans* oriented olefinic protons at δ 7.62 and 7.93 (each d, *J* = 15.4 Hz) and a chelated phenolic proton at δ 13.5, indicating the existence of a chalcone structure with a hydroxyl group at C-2'.

The ^{13}C NMR (Table 8 and Figure 30) and HSQC (Figure 33) spectra showed 20 carbon signals, corresponding to two methyls, one methylene, eleven methines, and six quaternary carbons. The ^{1}H NMR spectrum also exhibited a set of prenyloxyl signals at δ 4.61 (d, *J* = 6.7 Hz, H-1"), 5.53 (t, *J* = 6.7 Hz, H-2"), 1.86 (s, Me-4") and 1.81 (s, Me-5"). The presence of an AA'BB'C spin system at δ 7.69 (2H, m, H-2 and H-6) and δ 7.47 (3H, m, H-3, H-4 and H-5) together with the fragment ion at *m/z* 103 indicated an unsubstituted ring B (Drewes, 1974). In the EIMS, the molecular ion through the loss of the prenyl group with H transfer gave a fragment ion at *m/z* 240, and this ion subsequently underwent α-cleavage to give an ion at *m/z* 137, suggesting the presence of the prenoxyl group on ring A (Drewes, 1974). On ring A, an ABM splitting system consisting of a broad singlet at δ 6.53 (H-3'), a broad doublet at δ 6.54 (*J* = 8.2 Hz, H-5'), and a doublet at δ 7.87 (*J* = 8.2 Hz, H-6'), together with the HMBC correlation (Figures 34-35) of H-6' with C-β' (δ 191.7) placed the prenyloxy group at C-4'. This was confirmed by the ROESY interactions (Figure 32) of H-1" with H-3' and H-5'. The three-bond correlations were also found between C-β' and H-β, C-4' and H-6' and H-1", and C-1 and H-3 (H-5) and H-α in the HMBC spectrum (Table 8).

Based on the above spectral evidence, compound ME1 was identified as derricidin [**443**], a chalcone first isolated from the root bark of *Derris sericea* (do Nascimento and Mors, 1972).



[**443**]

Table 8 NMR Spectral data of compound ME1 and derricidin ($CDCl_3$)

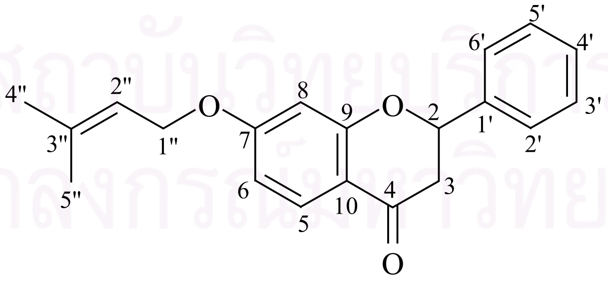
position	Compound ME1		Derricidin	HMBC (correlation with ^{1}H)
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	
1	-	134.8 (s)	-	H-3, H-5, H-α and H-β*
2	7.69 (m)	128.5 (d)	6.40-7.90	H-6, H-4 and H-β
3	7.47 (m)	128.9 (d)	6.40-7.90	H-5
4	7.47 (m)	130.6 (d)	6.40-7.90	H-2 and H-6
5	7.47 (m)	128.9 (d)	6.40-7.90	H-3
6	7.69 (m)	128.5 (d)	6.40-7.90	H-2, H-4 and H-β
1'	-	113.9 (s)	-	HO-2' and H-6'*
2'	-	165.6 (s)	-	H-6', H-3'* and HO-2'*
3'	6.53 (br s)	101.7 (d)	6.40	H-5' and HO-2'
4'	-	165.5 (s)	-	H-6'
5'	6.54 (br d, 8.2)	108.3 (d)	6.40	H-3'
6'	7.87 (d, 8.2)	131.2 (d)	7.90	-
α	7.62 (d, 15.4)	120.3 (d)	7.20	H-β*
β	7.93 (d, 15.4)	144.3 (d)	7.80	H-2, H-6 and H-α*
β'	-	191.7 (s)	-	H-6', H-β and H-α*
1"	4.61 (d, 6.7)	65.2 (t)	4.42 (2H)	-
2"	5.53 (t, 6.7)	118.6 (d)	5.50	-
3"	-	139.1 (s)	-	H-1", H-4" and H-5"
4"	1.86 (s)	25.7 (q)	1.78	H-5"
5"	1.81 (s)	18.2 (q)	1.78	H-4"
2'-OH	13.5 (s)	-	-	-

*Two-bond coupling

1.4 Structure Determination of Compound ME2

Compound ME2 was obtained as a yellow powder. The EIMS spectrum (Figure 38) showed a molecular ion [M^+] at *m/z* 308, corresponding to $C_{20}H_{20}O_3$. The IR spectrum exhibited absorption bands for carbonyl (1681 cm^{-1}) and ether (1254, 1219 cm^{-1}) functionalities (Figure 37). The UV absorptions at 274 and 309 nm (Figure 36) were indicative of a flavanone skeleton (Markham, 1982).

The ^{1}H NMR signals (Table 9 and Figure 39) at δ 2.88 (dd, J = 16.9, 3.0 Hz, H-3eq), δ 3.09 (dd, J = 16.9, 13.0 Hz, H-3ax) and δ 5.52 (d, J = 13.0, 3.0 Hz, H-2), and ^{13}C NMR resonances (δ 80.0 for C-2, δ 44.4 for C-3, and δ 190.0 for C-4) further confirmed a flavanone structure. The ^{1}H NMR data also revealed the presence of a γ,γ-dimethylallyloxy group with signals at δ 1.78, 1.85 (3H each, s, Me-4" and Me-5"), δ 4.59 (2H, d, J = 6.8 Hz, H-1"), and 5.52 (1H, t, J = 5.2 Hz, H-2") which correlated to the ^{13}C NMR (Table 9 and Figure 40) signals at δ 25.8, 18.2, 65.3, and 118.7, respectively. Ring B was unsubstituted as evidenced by the fragment ion at *m/z* 103 resulting from *retro*-Diels-Alder cleavage of ring C (Drewes, 1974). The presence of the fragment ion at *m/z* 240 due to the elimination of the prenyl group from the molecule and the ion at *m/z* 163 resulting from RDA cleavage ring C after losing of the prenyl group suggested the attachment of the γ,γ-dimethylallyloxy group on ring A (Drewes, 1974). An ABM proton splitting system at δ 6.55 (d, J = 2.2 Hz, H-8), δ 6.67 (dd, J = 8.8, 2.2 Hz, H-6), and δ 7.91 (d, J = 8.8 Hz, H-5) was observed. The HMBC correlation between C-4 and H-5 indicated that the γ,γ-dimethylallyloxy group should be placed at C-7. The HMBC spectrum also demonstrated a ^3J correlation peak for each of these pairs of H-C atoms: H-2 and C-4; H-2' (H-6') and C-2; H-5 and C-7, and H-1" and C-7, as shown in Table 9. Compound ME2 was identified as 7-γ,γ-dimethylallyloxyflavanone [**444**] (Islam, Gupta and Krishnamurti, 1981).



[**444**]

Table 9 NMR Spectral data of compound ME2 and 7-γ,γ-dimethylallyloxyflavanone ($CDCl_3$)

position	Compound ME2		7-γ,γ-Dimethylallyl -oxyflavanone	HMBC (correlation with ^{1}H)
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	
2	5.52 (d, 13.0, 3.0)	80.0 (d)	5.28-5.50	H-2', H-6' and H-3*
3	3.09 (dd, 16.9, 13.0)	44.4 (t)	2.80-3.10	-
	2.88 (dd, 16.9, 3.0)	44.4 (t)	2.80-3.10	-
4	-	190.0 (s)	-	H-5, H-2 and H-3*
5	7.91 (d, 8.8)	128.7 (d)	7.80	-
6	6.67 (dd, 8.8, 2.2)	110.8 (d)	6.40-6.62	H-8
7	-	165.0 (s)	-	H-5 and H-8*
8	6.55 (d, 2.2)	101.6 (d)	6.40-6.62	H-6
9	-	163.0 (s)	-	H-5 and H-8*
10	-	115.0 (s)	-	H-8
1'	-	138.8 (s)	-	H-3, H-3', H-5' and H-2'*
2'	7.52 (m)	126.2 (d)	7.35	H-4' and H-6'
3'	7.52 (m)	128.7 (d)	7.35	H-5'
4'	7.52 (m)	128.7 (d)	7.35	H-2' and H-6'
5'	7.52 (m)	128.7 (d)	7.35	H-3'
6'	7.52 (m)	126.2 (d)	7.35	H-2' and H-4'
1"	4.59 (d, 6.8)	65.3 (t)	4.5	-
2"	5.52 (t, 5.2)	118.7 (d)	5.28-5.50	H-1"*
3"	-	138.8 (s)	-	H-1", H-4"*, H-5"* and H-2"*
4"	1.85 (s)	25.8 (q)	1.84	H-5"
5"	1.78 (s)	18.2 (q)	1.76	H-4"

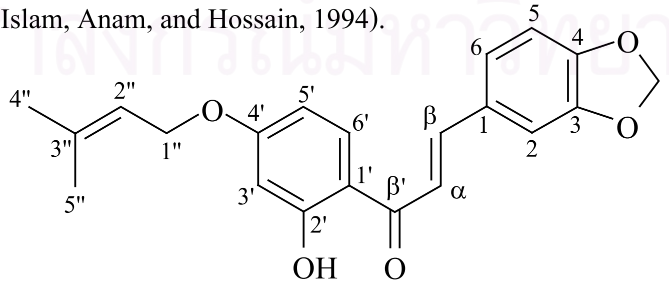
*Two-bond coupling

1.5 Structure Determination of Compound ME3

Compound ME3 was isolated as a yellow powder. It showed the [M$^+$] ion at *m/z* 352 in the EIMS (Figure 46), analyzed for $C_{21}H_{20}O_5$. The UV maxima at 258, 313 and 373 nm (Figure 44) and the IR absorptions at 3435 (hydroxyl), 1635 (conjugated carbonyl) and 1573 (conjugated C=C) cm^{-1} (Figure 45) were suggestive of a chalcone skeleton (Markham, 1982).

In the ^{1}H NMR spectrum (Table 10 and Figure 47), a set of *trans*-olefinic protons at δ 7.42 and 7.85 (each d, J = 15.2 Hz, H-α and H-β) and a chelated phenolic proton at δ 13.6 assignable to 2'-OH were observed. The ^{13}C NMR (Table 10) and HSQC (Figure 49) spectra showed 21 carbon signals, corresponding to two methyls, two methylenes, nine methines, and seven quaternary carbons. Two substituents were attached to the 2'-hydroxychalcone nucleus, as indicated by signals for a γ,γ-dimethylallyloxy group [δ 1.81, 1.86 (6H, 2 x Me), δ 4.61 (2H, d, J = 6.7 Hz, H-1''), and δ 5.53 (1H, t, J = 5.2 Hz, H-2'')] and a methylenedioxy group at δ 6.10 (2H, s) in the ^{1}H NMR spectrum. The methylenedioxy group was located at C-3 and C-4 on ring B, as supported by the fragment ion at *m/z* 148 in the EIMS (Drewes, 1974) and the presence of an ABM spin system at δ 7.22 (1H, br s, H-2), δ 7.19 (1H, br d, J= 8.0 Hz, H-6) and δ 6.90 (1H, d, J = 8.0 Hz, H-5) in the ^{1}H-^{1}H COSY spectrum (Figure). In the EI mass spectrum, the [M$^+$] through the loss of the prenyl group with H transfer gave a fragment ion at *m/z* 284, and this ion subsequently underwent α-cleavage to yield an ion at *m/z* 135. The presence of another set of ABM spin system at δ 6.52 (1H, br s, H-3'), δ 6.53 (1H, dd, J = 8.6, 2.5 Hz, H-5'), and δ 7.85 (1H, d, J = 8.6 Hz, H-6') suggested the location of the prenyloxy group at C-4' (Drewes, 1974). This was confirmed by the HMBC correlations (Figures 50-51) of C-β' (δ 191.5) with H-6', and C-4' (δ 165.9) with H-1'' and H-6'.

From all of the above spectral data, compound ME3 was identified as 2'-hydroxy-3,4-methylenedioxy-4'-γ,γ-dimethylallyloxychalcone [**445**]. This compound has been found as a natural compound for the first time in this study although its synthesis and ^{1}H and ^{13}C NMR data have been earlier reported (Islam, Anam, and Hossain, 1994).



[**445**]

Table 10 NMR Spectral data of compound ME3 ($CDCl_3$)

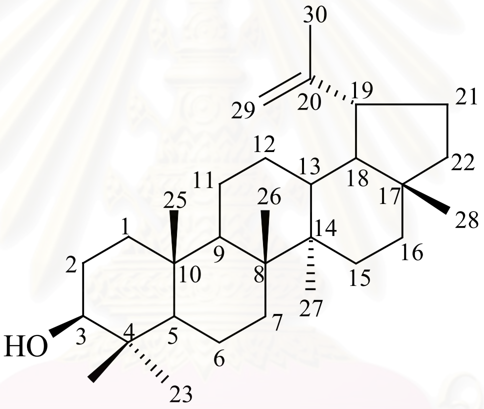
position	Compound ME3		HMBC (correlation with ^{1}H)
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	
1	-	129.2 (s)	H-α and H-5
2	7.22 (br s)	106.5 (d)	H-β
3	-	148.2 (s)	H-5, H-2* and $-OCH_2O-$
4	-	150.0 (s)	H-2 and $-OCH_2O-$
5	6.90 (d, 8.0)	108.5 (d)	-
6	7.19 (br d, 8.0)	125.5 (d)	H-2 and H-β
1'	-	114.0 (s)	H-3' and H-5'
2'	-	165.0 (s)	H-6'
3'	6.52 (br s)	101.5 (d)	H-5'
4'	-	165.9 (s)	H-6', H-1'', H-3' and H-5'
5'	6.53 (dd, 8.6, 2.5)	108 0 (d)	H-3'
6'	7.85 (d, 8.6)	130.6 (d)	-
α	7.42 (d, 15.2)	118.1 (d)	-
β	7.85 (d, 15.2)	144.2 (d)	H-2 and H-6
β'	-	191.5 (s)	H-6', H-β and H-α*
1''	4.61 (d, 6.7)	64.0 (t)	-
2''	5.53 (t, 5.2)	118.2 (d)	H-4'', H-5'' and H-1''*
3''	-	139.2 (s)	H-1'', H-4''* and H-5''*
4''	1.86 (s)	26.0 (q)	H-5''
5''	1.81 (s)	18.1 (q)	H-4''
$-OCH_2O-$	6.10 (s)	101.5 (t)	-

*Two-bond coupling

1.6 Structure Determination of Compound ME4

Compound ME4 was obtained as colorless needles. The IR spectrum exhibited absorption bands at 3322 (OH stretching) and 1456 and 1383 (CH bending) cm^{-1} (Figure 52).

The ^1H NMR spectrum (Figure 53) displayed signals for seven methyl groups at δ 0.77, 0.80, 0.84, 0.96, 0.98, 1.04 and 1.69. Signals for several methine and methylene protons appeared at δ 0.90-1.80. In addition, a proton signal at δ 3.20 (dd, J = 10.5, 5.4 Hz, H-3), a multiplet proton signal at δ 2.39 (H-19) and two broad singlet proton signals at δ 4.58 and δ 4.70 (H-29) were also observed. The ^{13}C NMR (Figure 55) and DEPT spectra (Figure 54) showed 30 carbon signals, corresponding to seven methyls, eleven methylenes, six methines and six quaternary carbons. These ^1H and ^{13}C NMR data which were in good agreement with those reported for lupeol [**169**] as shown in Table 11 (Reynolds, McLean, and Poplawski, 1986).



[**169**]

Table 11 NMR Spectral data of compound ME4 and lupeol ($CDCl_3$)

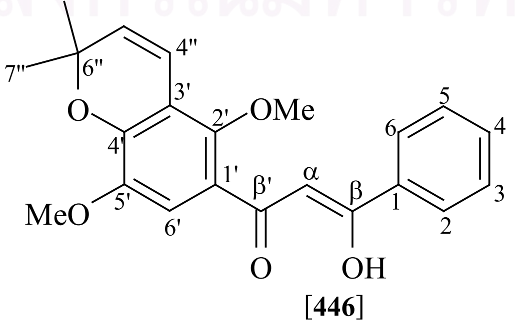
position	Compound ME4		Lupeol	
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H	^{13}C
1a	0.90-1.80	38.7 (t)	1.68	38.6
1b	0.90-1.80		0.91	
2a	0.90-1.80	27.3 (t)	1.61	27.3
2b	0.90-1.80		1.54	
3	3.20 (dd, 10.5, 5.4)	78.9 (d)	3.18 (dd)	78.9
4	-	38.8 (s)	-	38.8
5	0.90-1.80	55.3 (d)	0.69	55.2
6a	0.90-1.80	18.3 (t)	1.54	18.2
6b	0.90-1.80		1.39	
7	0.90-1.80	34.3 (t)	1.41	34.2
8	-	40.8 (s)	-	40.7
9	0.90-1.80	50.4 (d)	1.28	50.3
10	-	37.1 (s)	-	37.1
11a	0.90-1.80	20.9 (t)	1.42	20.9
11b	0.90-1.80		1.25	
12a	0.90-1.80	25.1 (t)	1.68	25.0
12b	0.90-1.80		1.07	
13	0.90-1.80	38.0 (d)	1.67	38.0
14	-	42.8 (s)	-	42.7
15a	0.90-1.80	27.4 (t)	1.71	27.4
15b	0.90-1.80		1.01	
16a	0.90-1.80	35.3 (t)	1.49	35.5
16b	0.90-1.80		1.38	
17	-	42.9 (s)	-	42.9
18	0.90-1.80	48.3 (d)	1.37	48.2
19	2.39 (m)	47.9 (d)	2.39	47.9
20	-	150.9 (s)	-	150.8
21a	1.93 (m)	29.8 (t)	1.93	29.8
21b	0.90-1.80		1.33	
22a	0.90-1.80	40.0 (t)	1.42	39.9
22b	0.90-1.80		1.20	
23	0.98 (s)	28.0 (q)	0.98	27.9
24	0.77 (s)	15.3 (q)	0.77	15.3
25	0.84 (s)	16.1 (q)	0.84	16.1
26	1.04 (s)	15.9 (q)	1.04	15.9
27	0.96 (s)	14.5 (q)	0.97	14.5
28	0.80 (s)	18.0 (q)	0.79	17.9
29	4.70 (br s)	109.3 (t)	4.69	109.3
	4.58 (br s)		4.56	
30	1.69 (s)	19.3 (q)	1.69	19.2

1.7 Structure Determination of Compound ME5

Compound ME5 was obtained as yellow needles. The EIMS (Figure 58) exhibited the molecular ion at *m/z* 366, corresponding to the molecular formula $C_{22}H_{22}O_5$. The UV absorptions at 225, 292 and 369 nm (Figure 56) and the IR bands for conjugated carbonyl (1590 cm^{-1}) and conjugated C=C (1566 cm^{-1}) functionalities (Figure 57) suggested a chalcone structure having a chelated hydroxyl group (Markham, 1982).

This was supported by a ^1H NMR signal at δ 7.39 (H-α) and three ^{13}C NMR signals at δ 96.0 (C- α), 184.5 (C-β) and 186.0 (C-β') (Table12 and Figure 59), due to keto-enol tautomerism effect. The ^1H NMR spectrum revealed the presence of two methoxyls [δ 3.82 and 3.95 (each 3H, s)], a dimethylpyran ring [δ 6.73 (1H, d, *J* = 9.9 Hz, H-4"), δ 5.93 (1H, d, *J* = 9.9 Hz, H-5"), and δ 1.52 (6H, 2 x Me)]. The proton singlet at δ 7.47, assignable to H-6' of ring A, showed long-range (^3J) coupling with the carbonyl carbon (δ 186.0, C-β') in the HMBC spectrum (Figures 63-64). Furthermore, a two-proton multiplet centered at δ 8.08 (H-2 and H-6) and a three-proton multiplet centered at δ 7.63 (H-3, H-4 and H-5) indicated unsubstitution for ring B. In the EIMS, a fragment ion at *m/z* 351 was formed through the elimination of a methyl group from the dimethylpyran ring and the loss of a methoxyl from [M^+] gave the base peak at *m/z* 335. An α-cleavage ion peak at *m/z* 247 suggested the placement of the two methoxyls and the dimethylpyran ring on ring A (Drewes, 1974). The first methoxyl could be placed at C-2' according to its NOESY correlation peak with H-α and the HMBC correlation of C-2' with H-6'. The second methoxyl was located at C-5', as shown by its NOESY interaction with H-6'. The position of the dimethylpyran ring on ring A was unambigously determined on the basis of the NOESY spectrum (Figure 62), which showed correlation contours between H-4" and MeO-2'. This was confirmed by the HMBC correlations of C-3' (δ 116.2) with H-5" and C-4' with H-6'.

From all of the above spectroscopic data in comparison with reported values (Tanaka *et al.*, 1991), compound ME5 was identified as ponganone I [**446**]. This compound was first isolated from the root bark of *Pongamia pinnata* (Tanaka *et al.*, 1991).



[**446**]

Table 12 NMR Spectral data of compound ME5 (acetone-d_6) and Ponganone I ($CDCl_3$)

position	Compound ME5		Ponganone I		HMBC
	^{1}H (mult., J in Hz)	^{13}C (mult.)	^{1}H (mult., J in Hz)	^{13}C	(correlation with ^{1}H)
1	-	136.5 (s)	-	135.6	H-α and H-2*
2	8.08 (m)	127.9 (d)	7.97 (m)	127.1	H-4, H-6, H-3* and H-5*
3	7.63 (m)	129.8 (d)	7.50 (m)	128.7	H-5
4	7.63 (m)	133.2 (d)	7.50 (m)	127.1	H-2 and H-6
5	7.63 (m)	129.8 (d)	7.50 (m)	128.7	H-3
6	8.08 (m)	127.9 (d)	7.97 (m)	127.1	H-2, H-4, H-3* and H-5*
1'	-	121.2 (s)	-	120.7	H-α and H-6'*
2'	-	151.5 (s)	-	150.5	H-6' and H-4"
3'	-	116.2 (s)	-	116.0	H-5", MeO-3', and H-4"*
4'	-	147.8 (s)	-	148.6	H-6' and H-4"
5'	-	146.3 (s)	-	145.3	H-6'*
6'	7.47 (s)	112.2 (d)	7.39 (s)	112.2	MeO-6'
α	7.39 (s)	96.0 (d)	7.28 (s)	97.0	-
β	-	184.5 (s)	-	184.0	H-2, H-6 and H-α*
β'	-	186.0 (s)	-	185.3	H-6' and H-α*
4"	6.73 (d, 9.9)	116.5 (d)	6.63 (d, 10.0)	116.8	-
5"	5.93 (d, 9.9)	132.0 (d)	5.72 (d, 10.0)	130.8	-
6"	-	77.8 (s)	-	77.3	H-4" and H-5"*
7"	1.52 (s)	26.8 (q)	1.50 (s)	28.0	H-5"
8"	1.52 (s)	26.8 (q)	1.50 (s)	28.0	H-5"
MeO-5'	3.95 (s)	56.0 (q)	3.91 (s)	56.4	-
MeO-2'	3.82 (s)	62.1 (q)	3.77 (s)	62.8	-

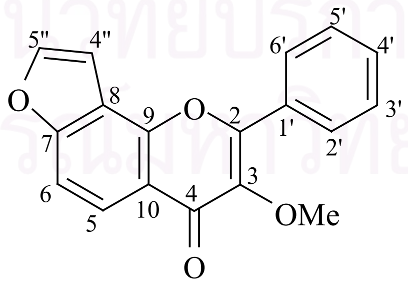
*Two-bond coupling

1.8 Structure Determination of Compound ME6

Compound ME6, a yellow powder, showed a [M^+] at *m/z* 292 in the EIMS (Figure 67), suggesting the molecular formula $C_{18}H_{12}O_4$. The IR bands at 1628 (conjugated carbonyl) and 1260 and 1166 (ether) cm^{-1} (Figure 66) and the UV absorptions at 215, 259 and 303 nm (Figure 65) were suggestive of a furanoflavone skeleton (Mbafor *et al*., 1995).

The ^1H NMR spectrum (Table 13 and Figure 68) displayed signals for a furan ring at δ 7.20 and δ 7.78 (each d, *J* = 2.1 Hz, H-4'' and H-5''), a methoxyl group at δ 3.98 (3H, s). An AA'BB'C spin system for the unsubstituted ring B at δ 8.16 (2H, m, H-2' and H-6') and δ 7.58 (3H, m, H-3', H-4' and H-5') was observed. In the EIMS, the fragment ion at *m/z* 160 due to RDA cleavage of ring C placed the furan ring on ring A (Drewes, 1974). The presence of an AB spin system at δ 7.57 (d, *J* = 8.8 Hz, H-6) and 8.22 (d, *J* = 8.8 Hz, H-5) together with the HMBC correlation between the carbonyl carbon (C-4, δ 175.3) and H-5 placed the furan ring in an angular position at C-7 and C-8. This was confirmed by the three-bond correlations of C-7 (δ 158.1) with H-5, H-4'' and H-5''. Finally, the methoxyl group should be located at C-3.

The NOE difference data, in which irradiation of a methoxyl signal (δ 3.98) enhanced the signal of H-2' and H-6' (δ 8.16), confirmed the position of the methoxyl at C-3. The assignment of ^{13}C NMR spectra was carried out by ^1H-^{13}C correlations of HSQC (Figure 71) and HMBC (Figure 72) experiments, however the signal for C-9 could not detected (Table 13). Therefore, ME6 was identified as 3-methoxy [2'',3'':7,8]-furanoflavone or karanjin [**295**] (Gupta and Krishnamurti, 1976b). This compound has been found in several plants such as *Lonchocarpus latifolius* (Magalhães *et al*., 2000) and *Pongamia glabra* (Malik, Sharma, and Seshadri, 1977).



[**295**]

Table13 NMR Spectral data of compound ME6 ($CDCl_3$)

position	^1H (mult., *J* in Hz)	^{13}C (mult.)	HMBC (correlation with ^1H)
2	-	155.0	H-2' and H-6'
3	-	141.8	MeO-3
4	-	175.3	H-5
5	8.22 (d, 8.8)	122.0	-
6	7.57 (d, 8.8)	110.0	-
7	-	158.1	H-5, H-4" and H-5"
8	-	114.1	H-6, H-5" and H-4"*
9	-	Not detected	-
10	-	120.0	H-6
1'	-	119.7	H-3' and H-5'
2'	8.16 (m)	128.5	H-4' and H-6'
3'	7.58 (m)	128.8	H-5'
4'	7.58 (m)	130.6	H-2' and H-6'
5'	7.58 (m)	128.8	H-3'
6'	8.16 (m)	128.5	H-2' and H-4'
4"	7.20 (d, 2.1)	104.5	-
5"	7.78 (d, 2.1)	146.0	-
MeO-3	3.98 (s)	60.1	-

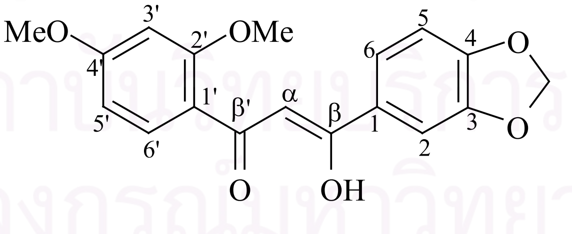
*Two-bond coupling

1.9 Structure Determination of Compound ME7

Compound ME7 was isolated as yellow needles. The EI mass spectrum (Figure 75) showed the [M^+] ion peak at *m/z* 328, corresponding to $C_{18}H_{16}O_6$. Compound ME7 should be a chelated hydroxy chalcone as indicated by the UV absorptions at 274 and 372 nm (Figure 73) and the IR bands of H-bonded hydroxyl (3086 cm^{-1}) and conjugated carbonyl (1607 cm^{-1}) (Figure 74) (Markham, 1982).

The ^1H NMR spectrum (Table 14 and Figure 76) showed an olefinic proton (δ 7.23, H-α) which correlated with a carbon at δ 96.8 in the HSQC spectrum (Figure 78). Two carbons which were affected by keto-enol tautomerism also appeared at δ 185.9 and 181.9 (C-β and C-β'). This suggested that compound ME7 was a hydroxychalcone derivative in a *Z*-configuration (Parmar *et al.*, 1989). Three substituents were attached to the β-hydroxychalcone nucleus, as indicated by the signals for two methoxyls at δ 3.98 and 4.09 (each 3H, s) and for a methylenedioxy group at δ 6.18 (2H, s) in the ^1H NMR spectrum. The two methoxyl groups were attached on ring A and a methylenedioxy group was assigned on ring B, as supported by the fragment ions at *m/z* 165 and 164 due to α-cleavage in the EI mass spectrum (Drewes, 1974). For ring A, the ABM splitting system consisting of two doublets at δ 6.71 (J = 2.0 Hz, H-3') and δ 7.98 (J = 8.5 Hz, H-6') and a doublet of doublets at δ 6.70 (J = 8.5, 2.0 Hz, H-5'), together with the HMBC correlation (Figure 79) of H-6' with C-β', suggested the location of the two methoxyls on *ortho*- and *para*-positions in relation to C-1' of ring A.

Compound ME7 was identified as milletenone [**280**] by analysis of the above spectral data and confirmed by comparison with previously published data (Khan and Zaman, 1974). This compound was first isolated from *Millettia ovalifolia* (Khan and Zaman, 1974).



[**280**]

Table 14 NMR Spectral data of compound ME7 (acetone-d_6)

position	^{1}H (mult., J in Hz)	^{13}C (mult.)	HMBC (correlation with ^{1}H)
1	-	130.8 (s)	H-5 and H-α
2	7.47 (d, 1.5)	106.9 (d)	H-6
3	-	148.6 (s)	H-5, H-2* and -OCH$_2$O-
4	-	151.6 (s)	H-2, H-5* and -OCH$_2$O-
5	6.99 (d, 8.5)	108.4 (d)	-
6	7.66 (dd, 8.5, 1.5)	123.1 (d)	H-2
1'	-	116.8 (s)	H-α, H-3' and H-5'
2'	-	161.2 (s)	H-6', MeO-2', and H-3'*
3'	6.71 (d, 2.0)	98.8 (d)	H-5'
4'	-	164.8 (s)	H-6', MeO-4', H-3'* and H-5'*
5'	6.70 (dd, 8.5, 2.0)	106.1 (d)	H-3'
6'	7.98 (d, 8.5)	131.8 (d)	-
α	7.23 (s)	96.8 (d)	-
β	-	185.9 (s)	H-2, H-6 and H-α*
β'	-	181.9 (s)	H-6' and H-α*
MeO-2'	3.98 (s)	55.4 (q)	-
MeO-4'	4.09 (s)	55.7 (q)	-
-OCH$_2$O-	6.18 (s)	102.4 (t)	-

*Two-bond coupling.

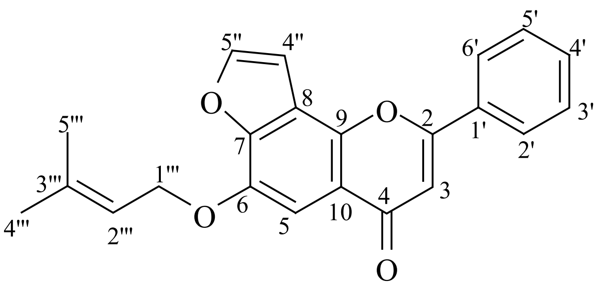
1.10 Structure Determination of Compound ME8

Compound ME8 showed $[M^+]$ at *m/z* 346, analyzed for $C_{22}H_{18}O_4$. The IR spectrum (Figure 81) exhibited absorption bands due to the presence of conjugated carbonyl (1639 cm^{-1}) and aromatic (1530 cm^{-1}) functionalities but no band for hydroxyl groups. The UV absorptions at 219, 268 and 302 nm (Figure 80) and the ^1H NMR signal at δ 6.93 (1H, s, H-3) were characteristics of a furanoflavone derivative (Mbafor *et al*., 1995).

The ^{13}C (Table 15 and Figure 84) and HSQC spectra (Figure 86) showed 22 carbon signals, indicating two methyls, ten methines, one methylene and seven quaternary carbons. The presence of an AA'BB'C spin system in ^1H NMR spectrum at δ 8.00 (2H, m, H-2' and H-6') and δ 7.59 (3H, m, H-3', H-4' and H-5') suggested an unsubstituted B ring. The ^1H NMR spectrum (Table 15 and Figure 83) also revealed the presence of signals for a γ,γ-dimethylallyoxy group [δ 4.84 (2H, d, J = 6.9 Hz, H-1'''), δ 5.67 (1H, t, J = 6.9 Hz, H-2''') and δ 1.86 (6H, s, Me-4''' and Me-5''')]. An aromatic singlet proton at δ 7.59 was assigned to H-5 by its HMBC correlation with C-4 (δ 178.4). Furthermore, two one-proton doublets at δ 7.25 and δ 7.81 (J = 1.8 Hz) could be assigned to the H-4'' and H-5'' protons of the furan ring, respectively.

In the EIMS, prominent fragment ions were observed at *m/z* 278, 176 and 102. The fragment ion at *m/z* 102 resulting from RDA cleavage confirmed the presence of an unsubstituted B ring (Drewes, 1974). The $[M^+]$ through the elimination of the prenyl group with H transfer gave a fragment ion at *m/z* 278, and this ion subsequently underwent RDA cleavage of ring C to yield an ion at *m/z* 176, thereby confirming the presence of a furan ring and a prenyloxy unit on ring A (Drewes, 1974). The HMBC correlations of H-5 with C-4 and C-7 indicated the location of a furan ring at C-7 and C-8 and the correlations of H-5 with C-4 and C-6 (2-bond correlation) suggested the attachment of the prenyloxy group at C-6.

From the above ^1H and ^{13}C NMR data, together with the information from ^1H-^1H COSY (Figure 85), HSQC (Figure 86) and HMBC (Figure 87) experiments, compound ME8 was identified as ovalifolin [**308**], a flavone first reported from the leaves of *Millettia ovalifolia* (Khan and Zaman, 1974).



[**308**]

Table 15 NMR Spectral data of compound ME8 and ovalifolin ($CDCl_3$)

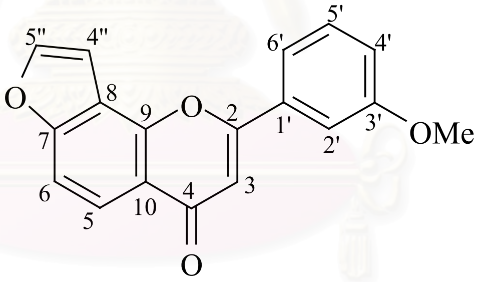
position	Compound ME8		Ovalifolin	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
2	-	162.2 (s)	-	H-2', H-6' and H-3*
3	6.93 (s)	107.7 (d)	6.89 (s)	-
4	-	178.4 (s)	-	H-5 and H-3*
5	7.59 (s)	101.2 (d)	7.56 (s)	-
6	-	143.9 (s)	-	H-5*
7	-	148.8 (s)	-	H-5, H-4" and H-5"
8	-	119.2 (s)	-	H-5" and H-4"*
9	-	146.0 (s)	-	H-5 and H-4"
10	-	120.3 (s)	-	H-3
1'	-	132.2 (s)	-	H-3, H-3' and H-5'
2'	8.00 (m)	126.4 (d)	7.96 (m)	H-6' and H-4'
3'	7.59 (m)	129.4 (d)	7.56 (m)	H-5'
4'	7.59 (m)	131.7 (d)	7.56 (m)	H-2' and H-6'
5'	7.59 (m)	129.3 (d)	7.56 (m)	H-3'
6'	8.00 (m)	126.4 (d)	7.96 (m)	H-2' and H-4'
4"	7.25 (d, 1.8)	104.9 (d)	7.22 (d, 2.0)	-
5"	7.81 (d, 1.8)	146.2 (d)	7.78 (d, 2.0)	-
1'''	4.84 (d, 6.9)	66.4 (t)	4.82 (br d, 7.0)	-
2'''	5.67 (t, 6.9)	119.0 (d)	5.63 (m)	H-4''' and H-5'''
3'''	-	139.6 (s)	-	H-1''', H-4''' and H-5'''
4'''	1.86 (s)	26.1 (q)	1.85 (s)	H-5'''
5'''	1.84 (s)	18.6 (q)	1.81 (s)	H-4'''

*Two-bond coupling

1.11 Structure Determination of Compound ME9

Compound ME9, a pale yellow powder, was analyzed for $C_{18}H_{12}O_4$ from its $[M^+]$ at *m/z* 292.07252 (calcd for 292.07355) in HREIMS. The UV absorptions at 210, 263 and 295 nm (Figure 88) and the IR bands at 1641 (conjugated carbonyl), 1530 (aromatic) and 1216 and 1072 (ether) cm^{-1} (Figure 89) were suggestive of a furanoflavone skeleton (Mbafor *et al*., 1995).

In the EIMS, the fragment ions at *m/z* 160 and 132 in the EIMS suggested that the furan ring should be located on ring A and the methoxyl group on ring B (Drewes, 1974). The HMBC correlations (Figure 96) of H-5 with C-4 and C-7 indicated the location of the furan ring on C-7 and C-8. For ring B, the methoxyl was situated at the *m*-position in relation to C-1', as shown by its NOESY interactions (Figure 94) with the protons at δ 7.53 (1H, br d, J = 3.6 Hz, H-2') and δ 7.15 (1H, dd, J = 7.8, 2.1 Hz, H-4'). Although compound ME9 has been obtained synthetically by methylation of pongol (Roy and Khanna, 1979), this is the first time that it has been found as a naturally occurring compound (Sritularak *et al*., 2002a). Regarding the NMR properties of compound ME9, it should be noted that prior to this investigation only partial ^1H NMR data have been available (Roy and Khanna, 1979), and no ^{13}C NMR study has been reported.



[**447**]

Table16 NMR Spectral data of compound ME9 ($CDCl_3$) and pongol methyl ether (DMSO-d_6)

position	Compound ME9		Pongol methyl ether	HMBC
	^1H (mult., J in Hz)	^{13}C (mult.)	^1H (mult., J in Hz)	(correlation with ^1H)
2	-	162.5 (s)	-	H-2', H-6' and H-3*
3	6.92 (s)	108.3 (d)	7.32 (s)	-
4	-	178.2 (s)	-	H-5 and H-3*
5	8.22 (d, 9.0)	121.8 (d)	8.6 (d, 10.0)	-
6	7.61 (d, 9.0)	110.2 (d)	7.7-7.9 (m)	-
7	-	158.4 (s)	-	H-5, H-4'' and H-5''
8	-	117.2 (s)	-	H-6 and H-5''
9	-	150.8 (s)	-	H-5
10	-	119.4 (s)	-	H-3 and H-6
1'	-	133.2 (s)	-	H-3 and H-5'
2'	7.53 (br d, 3.6)	111.9 (d)	8.0-8.5 (m)	H-4' and H-6'
3'	-	160.1 (s)	-	H-5' and MeO-3'
4'	7.15 (dd, 7.8, 2.1)	116.9 (d)	8.0-8.5 (m)	H-2' and H-6'
5'	7.52 (dd, 7.8, 7.8)	130.2 (d)	8.0-8.5 (m)	-
6'	7.60 (br d, 7.8)	118.6 (d)	8.0-8.5 (m)	H-2' and H-4'
4''	7.26 (d, 2.1)	104.2 (d)	7.7-7.9 (m)	H-4''*
5''	7.82 (d, 2.1)	145.8 (d)	8.0-8.5 (m)	H-5''*
MeO-3'	3.98 (s)	55.5 (q)	3.97 (s)	-

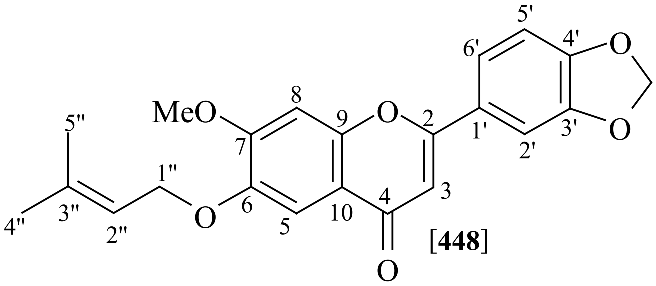
*Two-bond coupling.

1.12 Structure Determination of Compound ME10

Compound ME10, a pale yellow powder, exhibited a molecular ion [M^+] peak at *m/z* 380.11738 in the HREIMS, indicating a molecular formula of $C_{22}H_{20}O_6$ (calcd 380.12598). The IR bands at 1632 (conjugated C=O) and 1265 and 1109 (C-O stretch) cm^{-1} (Figure 98) and the UV absorptions at 240 and 329 nm (Figure 97) were characteristic of a flavone skeleton (Markham, 1982).

The ^1H NMR spectrum (Table 17 and Figure 100) confirmed the existence of the flavone nucleus (H-3, δ 6.69 Hz) and also displayed two sharp proton singlets at δ 7.59 and δ 6.99, assignable to the two *para*-coupled aromatic protons H-5 and H-8 of ring A. The assignment of H-5 was based on its long range (^3J) coupling to the carbonyl carbon (C-4, δ 177.6) observed in the HMBC spectrum (Figures 105-107). In the ^1H NMR spectrum, in addition to the signals for a γ,γ-dimethylallyloxy group [δ 1.81, 1.84 (6H, 2 x Me), δ 4.71 (2H, d, J = 6.6 Hz, H-1''), and δ 5.60 (1H, t, J = 6.6 Hz, H-3'')], two singlets at δ 6.10 (2H) and δ 4.03 (3H) were observed for a methylenedioxy and a methoxyl substituents, respectively. The methylenedioxy was placed on *m*- and *p*-positions in relation to C-1' for ring B, as a result of the fragment ion at *m/z* 146 in the EIMS and the ^1H NMR ABM spin system at δ 7.37 (1H, br s, H-2'), δ 7.49 (1H, br d, J = 8.4 Hz, H-6'), and δ 6.95 (1H, d, J = 8.4 Hz, H-5'). This led to the placement of the methoxyl and the γ,γ-dimethylallyloxy units on ring A. In the EIMS, the [M^+] through the loss of the prenyl group with H transfer gave a fragment ion at *m/z* 312, and this ion subsequently underwent *retro*-Diels-Alder cleavage of ring C to yield an ion at *m/z* 166, thereby confirming the presence of the prenoxyl unit on ring A (Drewes, 1974). The methoxyl was placed at C-7 according to its NOESY correlation peak with H-8 (Figure 103), leaving the γ,γ-dimethylallyloxy unit to be located at C-6. This was substantiated by the NOESY interaction of H_2-1'' with H-5. The HMBC spectrum confirmed the proposed structure of compound ME10, demonstrating a ^3J correlation peak for each pair of these H-C atoms: H-5 and C-4; H-8 and C-6; H_2-1'' and C-6; H-2' and C-2; H-6' and C-2.

The structure of ME10 was assigned as 3',4'-methylenedioxy-6-γ,γ-dimethylallyloxy-7-methoxyflavone, and has been given the trivial name millettocalyxin B [**448**] (Sritularak *et al.*, 2002a).



[**448**]

Table17 NMR Spectral data of compound ME10 ($CDCl_3$)

position	^1H (mult., *J* in Hz)	^{13}C (mult.)	HMBC (correlation with ^1H)
2	-	162.4 (s)	H-2', H-6' and H-3*
3	6.69 (s)	106.1 (d)	-
4	-	177.6 (s)	H-5 and H-3*
5	7.59 (s)	105.6 (d)	-
6	-	146.9 (s)	H-8 and H-5*
7	-	154.8 (s)	H-5 and H-8*
8	6.99 (s)	99.7 (d)	-
9	-	152.1 (s)	H-5 and H-8*
10	-	117.1 (s)	H-3, H-8 and H-5*
1'	-	126.0 (s)	H-3 and H-3'
2'	7.37 (br s)	106.2 (d)	H-6'
3'	-	148.4 (s)	H-5', H-2'*, and $-OCH_2O-$
4'	-	150.3 (s)	H-2', H-6', H-5'* and $-OCH_2O-$
5'	6.95 (d, 8.4)	108.7 (d)	-
6'	7.49 (br d, 8.4)	121.1 (d)	H-2'
1"	4.71 (d, 6.6)	66.1 (t)	-
2"	5.60 (t, 6.3)	119.0 (d)	H-4" and H-5"
3"	-	138.7 (s)	H-1", H-4" and H-5"
4"	1.84 (s)	25.9 (q)	H-5"
5"	1.81 (s)	18.3 (q)	H-4"
$-OCH_2O-$	6.10 (s)	101.9 (t)	-
MeO-7	4.03 (s)	56.4 (q)	-

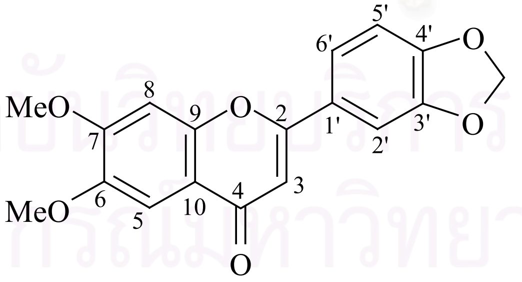
*Two-bond coupling.

1.13 Structure Determination of Compound ME11

Compound ME11 was obtained as a yellow powder. It showed a molecular [M^+] ion peak at *m/z* 326 in EIMS (Figure 110), suggesting a molecular formula of $C_{18}H_{14}O_6$. The IR bands at 1636 (conjugated carbonyl), 1595 (conjugated C=C) and 1453 (CH_2 bending) cm^{-1} (Figure 109) and the UV absorptions at 243 and 333 nm (Figure 108) were indicative of a flavone nucleus (Markham, 1982).

The ^1H NMR spectrum showed a singlet proton signal of H-3 (δ 6.86), confirming the flavone skeleton. It also exhibited two sharp proton singlets at δ 7.33 and δ 7.37, assignable to the *p*-coupled aromatic protons H-5 and H-8 of ring A (Table 18 and Figure 111). The assignment of H-5 was based on its HMBC correlation to C-4 (δ 171.4). The ^1H NMR spectral data, furthermore, revealed the presence of two methoxyl groups at δ 3.84 and δ 3.90 (each 3H, s) and a methylenedioxy unit at δ 6.14 (2H, s). The ions at *m/z* 180 and 146 resulting from *retro*-Diels-Alder cleavage of ring C in the EIMS suggested the location of the two methoxyls on ring A and the methylenedioxy on ring B (Drewes, 1974). The 7-OMe protons (δ 3.90) showed NOE interaction with H-8 (δ 7.37) and HMBC correlation with C-7 (δ 154.3). The 6-OMe (δ 3.84) protons exhibited an NOE cross peak with H-5 (δ 7.33) and also showed HMBC correlation with C-6 at δ 147.5. For ring B, the presence of an ABM spin system at δ 7.64 (1H, br s, H-2'), δ 7.08 (1H, d, *J* = 8.7 Hz, H-5') and δ 7.65 (1H, br d, *J* = 8.7 Hz, H-6') in the ^1H NMR spectrum, located the methylenedioxy group at C-3' and C-4' positions.

Based on the above spectral evidence, compound ME11 was identified as 3',4'-methylenedioxy-6,7-dimethoxyflavone or milletenin C [**298**] (Parma, Gupta and Sharma, 1989). This compound was first isolated from leaves of *Millettia ovalifolia* (Khan and Zaman, 1974).



[**298**]

Table 18 NMR Spectral data of compound ME11 (DMSO-d_6) and milletenin C ($CDCl_3$)

position	Compound ME11		Milletenin C		HMBC
	^{1}H (mult., J in Hz)	^{13}C (mult.)	^{1}H (mult., J in Hz)	^{13}C	(correlation with ^{1}H)
2	-	161.6 (s)	-	162.42	H-2', H-6' and H-3*
3	6.86 (s)	105.4 (d)	6.62 (s)	101.79	-
4	-	176.2 (s)	-	177.37	H-5 and H-3*
5	7.33 (s)	103.7 (d)	7.60 (s)	121.06	-
6	-	147.5 (s)	-	126.12	H-8, H-5* and MeO-6
7	-	154.3 (s)	-	162.42	H-5, H-8* and MeO-7
8	7.37 (s)	100.9 (d)	6.96 (s)	99.77	-
9	-	151.7 (s)	-	147.75	H-5 and H-8*
10	-	116.5 (s)	-	104.72	H-3, H-8 and H-5*
1'	-	125.3 (s)	-	121.06	H-3 and H-5'
2'	7.64 (br s)	106.2 (d)	7.34 (d, 1.5)	106.20	H-6'
3'	-	148.3 (s)	-	147.75	H-5', H-2'* and $-OCH_2O-$
4'	-	150.3 (s)	-	147.75	H-2', H-5'* and $-OCH_2O-$
5'	7.08 (d, 8.7)	108.8 (d)	6.93 (d, 8.0)	106.26	-
6'	7.65 (br d, 8.7)	121.3 (d)	7.44 (dd, 8.0, 1.5)	108.68	H-2'
MeO-6	3.84 (s)	55.6 (q)	3.98 (s)	56.37	-
MeO-7	3.90 (s)	56.0 (q)	4.00 (s)	56.37	-
$-OCH_2O-$	6.14 (s)	102.1 (t)	6.05 (s)	101.79	-

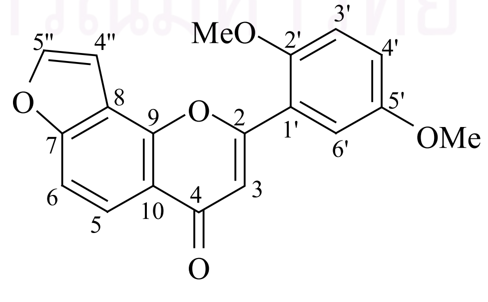
*Two-bond coupling.

1.14 Structure Determination of Compound ME12

Compound ME12, a yellow powder, showed a molecular ion [M^+] at *m/z* 322.08367 in the HREIMS, corresponding to the molecular formula $C_{19}H_{14}O_5$ (calcd 322.08414). The IR bands at 1637 (conjugated C=O), 1595 (conjugated C=C) and 1205 and 1072 (C-O stretching) cm^{-1} (Figure 118) and the UV absorptions at 249 and 295 nm (Figure 117) were indicative of a furanoflavone (Mbafor *et al.*, 1995).

This was supported by the ^{1}H and ^{13}C NMR signals (Table19 and Figure 120-121) for H-3/C-3 at δ 7.23 (1H, s)/ δ 113.2 and for a furan ring at δ 7.16 (1H, d, *J* = 2.0 Hz, H-4")/ δ 104.3 (C-4") and δ 7.75 (1H, d, *J* = 2 Hz, H-5")/ δ 145.7 (C-5"). Furthermore, the presence of two methoxyls was revealed by the proton resonance at δ 3.91 (3H, s) and δ 3.95 (3H, s) and the carbon signals at δ 56.0 (q) and δ 56.2 (q). In the EIMS, the fragment ions at *m/z* 160 and 162 resulting from *retro*-Dials-Alder cleavage of the [M^+] suggested the placement of the furan ring on ring A and the two methoxyls on ring B (Drewes, 1974). The appearance of H-5 and H-6 as doublets at δ 8.16 (d, *J* = 9 Hz) and δ 7.54 (*J* = 9 Hz) and the HMBC correlations of H-5 with C-4 and C-7 indicated that the furan ring should be fused in an angular position at C-7 (oxygenated) and C-8. Interactions through ^3J coupling of C-7 with H-4" and H-5", and of C-8 with H-5", were also observed. To determine the locations of the two methoxyls on ring B, a NOESY experiment (Figure 122) was carried out. The NOE interactions of the methoxyl at δ 3.95 with H-3 and with the proton at δ 6.99 (d, *J* = 9.0 Hz, H-3') placed this methoxyl at C-2'. The other methoxyl at δ 3.91 could be located at C-5' according to its NOE effects with the proton at δ 7.04 (dd, *J* = 9.0, 3.0 Hz, H-4') and δ 7.50 (d, *J* = 3.0 Hz, H-6'). A three-bond correlation was also found between H-6' and C-2 in the HMBC spectrum (Figures 124-125).

On the basis of the above spectroscopic studies, compound ME12 was thus identified as a new compound, 2',5'-dimethoxy[2",3":7,8]-furanoflavone and has been given the trivial name millettocalyxin C [**449**] (Sritularak *et al.*, 2002a).



[**449**]

Table19 NMR Spectral data of compound ME12 ($CDCl_3$)

position	^1H (mult., *J* in Hz)	^{13}C (mult.)	HMBC (correlation with ^1H)
2	-	159.8 (s)	H-6' and H-3*
3	7.23 (s)	113.2 (d)	-
4	-	178.7 (s)	H-5
5	8.16 (d, 9.0)	121.8 (d)	-
6	7.54 (d, 9.0)	110.0 (d)	-
7	-	158.3 (s)	H-5, H-4" and H-5"
8	-	117.2 (s)	H-6 and H-5"
9	-	151.0 (s)	H-5
10	-	119.3 (s)	H-3 and H-6
1'	-	121.4 (s)	H-3 and H-3'
2'	-	152.5 (s)	H-4', H-6' and MeO-2'
3'	6.99 (d, 9.0)	113.2 (d)	-
4'	7.04 (dd, 9.0, 3.0)	117.3 (d)	H-6'
5'	-	153.6 (s)	H-3' and MeO-5'
6'	7.50 (d, 3.0)	114.7 (d)	H-4'
4"	7.16 (d, 2.0)	104.3 (d)	-
5"	7.75 (d, 2.0)	145.7 (d)	-
MeO-2'	3.95 (s)	56.2 (q)	-
MeO-5'	3.91 (s)	56.0 (q)	-

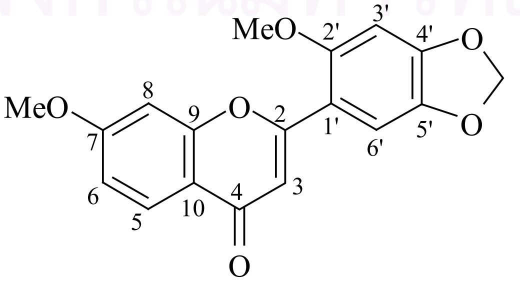
*Two-bond coulping.

1.15 Structure Determination of Compound ME13

Compound ME13 was obtained as a pale yellow powder. The molecular formula was determined as $C_{18}H_{14}O_6$ by HREIMS of its [M^+] ion at *m/z* 326.08335 (calcd 326.07904). The IR spectrum showed absorption bands of a conjugated double bond at 1616 and 1567 cm^{-1} (Figure 127). The UV absorptions at 300 and 354 nm (Figure 126) and the ^1H NMR signal at δ 6.94 (1H, s, H-3) were indicative of a flavone skeleton (Markham, 1982).

The ^{13}C NMR (Table 20 and Figure 130) and HSQC spectra (Figure 132) showed 18 carbon signals, corresponding to two methoxyls, one methylene, six methines, and nine quaternary carbons. Three substituents were attached to the flavone nucleus, as indicated by signals for two methoxyls at δ 4.02 (6H, s) and for a methylenedioxy group at δ 6.15 (2H, s) in the ^1H NMR spectrum (Table 20 and Figure 129). The first methoxyl could be placed on ring A, while the second methoxyl and the methylenedioxy were assigned to ring B, as supported by the fragment ions at *m/z* 150 and 176 due to *retro*-Diels-Alder cleavage of ring C in the mass spectrum (Drewes, 1974). For ring A, the ABM splitting system consisting of two doublets at δ 7.26 (J = 2.4 Hz, H-8) and 8.03 (J = 8.8 Hz, H-5) and a double doublet at δ 7.06 (J = 8.8, 2.4 Hz, H-6), together with the HMBC correlation of H-5 with C-4 (δ 177.6), suggested the location of the first methoxyl at C-7. For ring B, the appearance of two aromatic proton singlets at δ 6.95 and 7.52 indicated their *para*-correlation, placing the second methoxyl at C-2' and the methylenedioxy moiety at C-4' and C-5'. This was confirmed by the three-bond correlation between H-6' (δ 7.52) and C-2 (δ 160.8) in the HMBC spectrum (Figure 133). A NOESY experiment (Figure 131) revealed interactions of H-8 with MeO-7 and of H-3' with MeO-2'. A NOESY cross-peak between H-6 and MeO-7 was also observed.

Based on the above spectral evidence, compound ME13 was identified as a new flavone, 4',5'-methylenedioxy-7,2'-dimethoxyflavone and has been named millettocalyxin A [**450**] (Sritularak *et al.*, 2002a).



[**450**]

Table 20 NMR Spectral data of compound ME13 (acetone-d_6)

position	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	HMBC (correlation with ^{1}H)
2	-	160.8 (s)	H-6' and H-3*
3	6.94 (s)	111.7 (d)	-
4	-	177.6 (s)	H-5 and H-3*
5	8.03 (d, 8.8)	127.1 (d)	-
6	7.06 (dd, 8.8, 2.4)	114.9 (d)	H-8
7	-	165.0 (s)	H-5 and MeO-7
8	7.26 (d, 2.4)	101.4 (d)	-
9	-	158.9 (s)	H-5 and H-8*
10	-	118.4 (s)	H-3, H-6 and H-8
1'	-	113.5 (s)	H-3 and H-3'
2'	-	156.1 (s)	H-6', H-3'* and MeO-2'
3'	6.95 (s)	96.1 (d)	-
4'	-	151.9 (s)	H-6', H-3'* and -OCH$_2$O-
5'	-	142.7 (s)	H-3', H-6'* and -OCH$_2$O-
6'	7.52 (s)	108.2 (d)	-
-OCH$_2$O-	6.15 (s)	103.1 (t)	-
MeO-7	4.02 (s)	56.4 (q)	-
MeO-2'	4.02 (s)	57.1 (q)	-

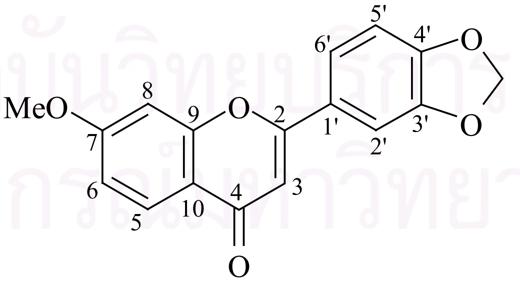
*Two-bond coupling.

1.16 Structure Determination of Compound ME14

Compound ME14, a pale yellow powder, exhibited a molecular ion [M^+] peak at *m/z* 296 in the EIMS (Figure 136), corresponding to $C_{17}H_{12}O_5$. The presence of a flavone skeleton was evident from the UV absorptions at 237 and 332 nm (Figure 134), the IR bands at 1610 (conjugated C=O), 1590 (conjugated C=C) and 1450 (CH_2 bending) cm^{-1} (Figure 135), and a typical singlet proton of H-3 at δ 6.73 (Markham, 1982).

The ^{13}C NMR (Table 21, Figure 138) and HSQC spectral data (Figure 139) showed 17 carbon signals, corresponding to one methoxyl, seven methines, one methylene and eight quaternary carbons. Comparison of the ^{13}C NMR data with those of compound ME11 indicated that ME14 differed from ME11 only by lacking a methoxyl group. The ^{1}H NMR spectrum (Table 21 and Figure 137) exhibited the signals for a methoxyl at δ 4.0 (3H, s) and a methylenedioxy at δ 6.20 (2H, s). The methoxyl should be placed on ring A, as indicated by the ions at *m/z* 122 and *m/z* 268 (Drewes, 1974). The presence of two doublets at δ 7.29 (*J* = 2.3 Hz, H-8) and δ 8.04 (*J* = 8.8 Hz, H-5) and a doublet doublet at δ 7.09 (*J* = 8.8, 2.3 Hz, H-6), together with the HMBC correlation (Figure 140) of H-5 with C-4 (δ 177.5), supported the placement of the methoxyl at C-7. The methylenedioxy group was placed on ring A, as suggested by the fragment ion at *m/z* 146. The second ABM splitting system at δ 7.61 (d, *J* = 1.8 Hz, H-2'), 7.08 (d, *J* = 8.9, H-5') and 7.71 (dd, *J* = 8.9, 1.8 Hz, H-6'), along with the ^{3}J correlation peak of H-2' with C-2 (δ 163.5), indicated the location of a methylenedioxy group at C-3' and C-4'.

Based on the above spectral data, this compound was identified as 3',4'-methylenedioxy-7-methoxyflavone [**279**]. Its ^{1}H NMR data are in good agreement with earlier published data (Mahmoud and Waterman, 1985).



[**279**]

Table 21 NMR Spectral data of compound ME14 (acetone-d_6) and 3',4'-methylenedioxy-7-methoxyflavone (pyridine-d_5)

position	Compound ME14		3',4'-Methylenedioxy-7-methoxyflavone	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
2	-	163.5 (s)	-	H-3*
3	6.73 (s)	106.8 (d)	7.08 (s)	-
4	-	177.5 (s)	-	H-5 and H-3*
5	8.04 (d, 8.8)	127.3 (d)	8.35 (d, 9.0)	-
6	7.09 (dd, 8.8, 2.3)	115.2 (d)	7.05 (dd, 9.0, 2.0)	H-8
7	-	165.4 (s)	-	H-5, H-8* and MeO-7
8	7.29 (d, 2.3)	101.5 (d)	7.15 (d, 2.0)	-
9	-	159.0 (s)	-	H-8*
10	-	118.6 (s)	-	H-3 and H-8
1'	-	126.8 (s)	-	H-3 and H-5'
2'	7.61 (d, 1.8)	106.9 (d)	7.60 (d, 2.0)	-
3'	-	149.7 (s)	-	H-5' and -OCH_2O-
4'	-	151.7 (s)	-	H-2', H-5'* and -OCH_2O-
5'	7.08 (d, 8.9)	109.4 (d)	6.99 (d, 8.0)	-
6'	7.71 (dd, 8.9, 1.8)	122.2 (d)	7.54 (dd, 8.0, 2.0)	H-2'
-OCH_2O-	6.20 (s)	103.0 (t)	6.08 (s)	-
MeO-7	4.0 (s)	56.1 (q)	3.80 (s)	-

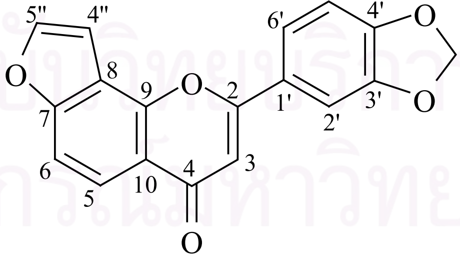
*Two-bond coupling

1.17 Structure Determination of Compound ME15

The EIMS of compound ME15 showed a $[M^+]$ ion at *m/z* 306 (Figure 143), corresponding to the elemental composition $C_{18}H_{10}O_5$. The IR spectrum exhibited the absorption bands for conjugated carbonyl (1640 cm^{-1}) and conjugated unsaturation (1592 cm^{-1}) (Figure 142). The UV absorptions at 241 and 328 nm (Figure 141) and the ^{1}H NMR signal at δ 6.82 (1H, s, H-3) were suggestive of a furanoflavone nucleus (Table 22 and Figure 144) (Mbafor *et al*., 1995)

In addition, ^{1}H and ^{13}C NMR signals (Table 22 and Figure 145) for a furan ring were observed at δ 7.24 (1H, d, J = 2.1, H-4")/δ 104.4 (C-4") and δ 7.82 (1H, d, J = 2.1 Hz, H-5")/ δ146.1 (C-5"). The ^{1}H NMR spectrum also revealed the presence of a methylenedioxy group at δ 6.14 (2H, s). The fragment ion at *m/z* 160 and 146 due to *retro*-Diels-Alder cleavage of ring C in the EIMS suggested the location of the furan ring on ring A and the methylenedioxy group on ring B (Drewes, 1974). The furan ring could be fused in an angular position at C-7 and C-8, as supported by the presence of two doublets (H-5 and H-6) at δ 8.20 and δ 7.60 (each d, J = 8.7 Hz) and the HMBC correlation of H-5 with C-4 (δ 178.4) (Figures 148-149). Furthermore, the HMBC correlations of C-7 (δ 158.6) with H-4" and H-5" and C-8 (δ 117.3) with H-5", were also observed. The methylenedioxyl was placed at C-3' and C-4', as evident from the ABM spin system at δ 7.44 (d, J = 1.8 Hz, H-2'), δ 7.01 (d, J = 8.1 Hz, H-5') and δ 7.57 (dd, J = 8.1, 1.8 Hz, H-6').

On the basis of the above spectroscopic data, together with the information from HSQC (Figure 147) and ^{1}H-^{1}H COSY (Figure 146) experiments, compound ME15 was identified as pongaglabrone [**314**]. Its ^{1}H NMR data are in agreement with literature values (Garcez *et al*., 1988).



[**314**]

Table 22 NMR Spectral data of compound ME15 and pongaglabrone ($CDCl_3$)

position	Compound ME15		Pongaglabrone	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
2	-	162.6 (s)	-	H-2', H-6' and H-3*
3	6.82 (s)	107.3 (d)	6.80 (s)	-
4	-	178.4 (s)	-	H-5 and H-3*
5	8.20 (d, 8.7)	122.0 (d)	8.22 (d, 8.0)	-
6	7.60 (d, 8.7)	110.4 (d)	7.59 (d, 8.0)	-
7	-	158.6 (s)	-	H-4" and H-5"
8	-	117.3 (s)	-	H-6 and H-5"
9	-	150.9 (s)	-	H-5
10	-	119.5 (s)	-	H-3 and H-6
1'	-	126.0 (s)	-	H-3 and H-5'
2'	7.44 (d, 1.8)	106.5 (d)	7.44 (d, 2.0)	H-6'
3'	-	148.8 (s)	-	H-5', H-2'* and -OCH$_2$O-
4'	-	150.8 (s)	-	H-2', H-6', H-5'* and -OCH$_2$O-
5'	7.01 (d, 8.1)	109.1 (d)	7.00 (d, 8.0)	-
6'	7.57 (dd, 8.1, 1.8)	121.6 (d)	7.59 (dd, 8.0, 2.0)	H-2'
4"	7.24 (d, 2.1)	104.4 (d)	7.20 (d, 2.0)	H-5"
5"	7.82 (d, 2.1)	146.1 (d)	7.82 (d, 2.0)	H-4"
-OCH$_2$O-	6.14 (s)	102.2 (t)	6.14 (s)	-

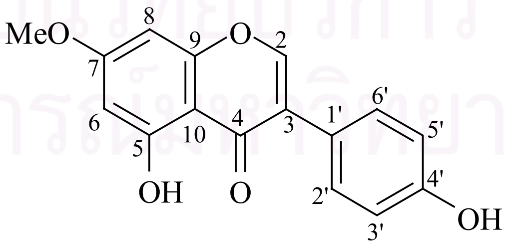
*Two-bond coupling

1.18 Structure Determination of Compound ME16

Compound ME16 was isolated as a yellow powder. Its EIMS (Figure 152) showed a molecular ion peak at *m/z* 284, analyzed for $C_{16}H_{12}O_5$. Compound ME16 was an isoflavone containing a chelated hydroxyl, as shown by the IR bands at 3380 (hydroxyl), 1665 (conjugated C=O), 1614 (conjugated C=C) and 1570 (C=C aromatic) cm^{-1} (Figure 151) and by the UV absorption at 261 nm (Figure 150) (Markham, 1982).

The two sharp singlet proton signals at δ 13.02 (chelated hydroxyl) and δ 8.25 (H-2 of isoflavone) and the carbon signal at δ 154.0 (C-2) in the ^1H and ^{13}C NMR spectra (Table 23 and Figures 153-154) confirmed the existence of the isoflavone nucleus (Dagne and Bekele, 1990). The ^1H NMR spectrum also exhibited a methoxyl signal at δ 3.98 (3H, s) and two doublets at δ 6.39 and 6.58, assignable to the two *meta*-coupled aromatic protons H-6 and H-8 of ring A. The assignment of H-6 was based on the HMBC correlation of C-6 (δ 98.1) with 5-OH. The presence of an AA'BB' spin system at δ 7.49 (2H, d, J = 8.8 Hz, H-2' and H-6') and δ 6.94 (2H, d, J = 8.8 Hz, H-3' and H-5') indicated a simple *para*-substituted B ring. The positions of H-2' and H-6' were assigned on the basis of its HMBC correlation (Figure 158) with C-3. The ^1H-^1H COSY experiment (Figure 155) revealed the interaction of H-2' (H-6') with H-3' (H-5'). In the EI mass spectrum, the fragment ions at *m/z* 166 and 118 resulting from RDA cleavage of ring C indicated the locations of the methoxyl on ring A and the hydroxyl on ring B (Drewes, 1974). The methoxyl should be placed at C-7, as shown by its NOESY interaction with H-6 and H-8. The HMBC correlation of C-4' at δ 158.1 (oxygenated carbon) with H-2' and H-6', confirmed the attachment of the hydroxyl at C-4'.

Compound ME16 was identified as prunetin [**451**] based on the above spectral data. Its ^1H NMR properties are in agreement with previously published values (Lin, Chen and Kuo, 1991).



[**451**]

Table 23 NMR Spectral data of compound ME16 (acetone-d_6) and prunetin (DMSO-d_6)

position	Compound ME16		Prunetin	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
2	8.25 (s)	154.0 (d)	8.04 (s)	-
3	-	123.8 (s)	-	H-2', H-6' and H-2
4	-	181.1 (s)	-	H-2
5	-	163.0 (s)	-	-
6	6.39 (d, 2.3)	98.1 (d)	6.26 (d, 2.1)	H-8 and HO-5
7	-	166.0 (s)	-	H-8* , HO-5, and MeO-7
8	6.58 (d, 2.3)	92.2 (d)	6.40 (d, 2.1)	-
9	-	158.3 (s)	-	H-8*
10	-	106.0 (s)	-	H-6, H-8 and HO-5
1'	-	122.2 (s)	-	H-3' and H-5'
2'	7.49 (d, 8.8)	130.3 (d)	7.25 (d, 8.5)	H-6'
3'	6.94 (d, 8.8)	115.4 (d)	6.78 (d, 8.5)	H-5'
4'	-	158.1 (s)	-	H-2' and H-6'
5'	6.94 (d, 8.8)	115.4 (d)	6.78 (d, 8.5)	H-3'
6'	7.49 (d, 8.8)	130.3 (d)	7.25 (d, 8.5)	H-2'
MeO-7	3.98 (s)	55.8 (q)	3.80 (s)	-
5-OH	13.02 (s)	-	12.82 (s)	-
4'-OH	8.48 (s)	-	9.28 (s)	-

*Two-bond coupling.

1.19 Structure Determination of Compound ME17

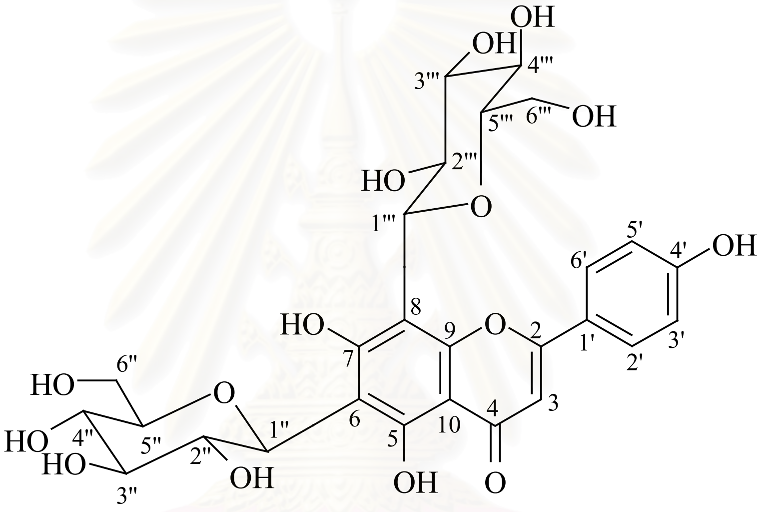
Compound ME17 was obtained as a yellow powder. The molecular weight should be 594 ($C_{27}H_{15}O_{30}$) as shown by an $[M\text{-}H]^-$ ion at *m/z* 592.8 in the negative ESIMS and an $[M+Na]^+$ ion at *m/z* 617.5 in the positive ESIMS (Figures 161-162). The IR spectrum exhibited absorption bands at 3382 (OH group), 1628 (conjugated C=C) and 1577 (aromatic C=C) cm^{-1} (Figure 160). The UV absorptions at 215, 270 and 325 nm (Figure 159) and the presence of two sharp proton singlets in the ^{1}H NMR spectrum at δ 14.3 (chelated hydroxyl) and δ 6.60 (1H, H-3) were indicative of a flavone skeleton with a hydroxyl group at C-5 (Markham, 1982).

The ^{1}H NMR (Table 24 and Figures 163-165) spectrum revealed the presence of an AA'BB' spin system at δ 8.25 (2H, d, J = 8.5 Hz, H-2' and H-6') and δ 7.23 (2H, d, J = 8.5 Hz, H-3' and H-5'), indicating *para*-substitution for ring B. In addition, the presence of two anomeric proton signals at δ 5.85 (d, J = 9.5 Hz, H-1") and δ 5.73 (d, J = 9.5 Hz, H-1"') suggested that compound ME17 should be a diglycoside of apigenin (Harborne, 1994).

The ^{13}C NMR (Table 24 and Figure 166) and HSQC spectra (Figure 172) displayed 12 signals for sugar carbons including two anomeric carbons at δ 76.3 (C-1") and 75.2 (C-1"'), in addition to 15 carbons of apigenin nucleus. The upfield shift of two anomeric carbons indicated that the two sugar moieties were attached to apigenin with C-linkage (δ 70-80 ppm for *C*-glycoside and 90-112 ppm for *O*-glycoside) (Agrawal, 1992).

To identify the two sugar units, a TOCSY experiment (Figures 167-168) was carried out. The TOCSY spectrum showed scalar couplings of the protons belonging to the same sugar unit. The first sugar moiety exhibited signals at δ 5.85, 4.65, 4.50, 4.42, 4.39, 4.37 and 4.06 and the other sugar moiety showed the signals at δ 5.73, 4.92, 4.63, 4.61, 4.54, 4.44 and 4.13. The connectivities of the seven sugar protons and the assignment of ^{13}C NMR signals in each sugar moiety were determined by ^{1}H-^{1}H COSY (Figure 169), HMQC and HSQC-TOCSY (Figure 171) experiments, as shown in Table 24. The observed vicinal coupling constants of J = 9.5 Hz between the *trans* diaxial oxymethine protons H-1" and H-2", and H-1"' and H-2"' suggested that H-1" and H-1"' were β-anomeric protons.

The ^3J couplings of H-1"' with C-9 (δ 163.2) and of H-1" with C-5 (δ 157.2) in HMBC spectrum, along with the ROESY correlations of H-2"' with H-2' and H-6' suggested the attachment of two sugar units at C-6 and C-8 of the flavone aglycone, respectively. The two-bond correlations of C-6 (δ 108.4) with H-1" and C-8 (δ 106.4) with H-1"' were also observed. In addition, the extensive doubling of ^{1}H NMR signals of compound ME17, the two signals were noted for H-3 (δ 6.60, 6.76), H-2' and H-6' (δ 8.12, 8.25) and HO-5 (δ 14.35, 14.50). This observation suggested that, in flavones, interaction occurred between a C-linked monohexose at C-8 and ring B. This phenomenon was observed in almost all compounds containing an 8-*C*-hexosyl substituent (Harborne, 1994). Comparison of the chemical shifts of the sugar carbons with reported data (Mahmoud *et al*., 1989) indicated that compound ME17 was an apigenin with 6,8-β-D-glucopyranosyl substitution (vicenin II [**452**]). This compound was previously found in several plants i.e. *Scolymus hispanicus* (Romussi and Ciarallo, 1978), *Fortunella japonica* (Kumamoto *et al*., 1985) and *Ephedra aphylla* (Hussein *et al*., 1997).



[**452**]

Table 24 NMR Spectral data of compound ME17 (pyridine-d_5) and vicenin II (DMSO-d_6)

position	Compound ME17		Vicenin II		HMBC (correlation with ^{1}H)
	^{1}H (mult., J in Hz)	^{13}C (mult.)	^{1}H (mult., J in Hz)	^{13}C	
2	-	164.7 (s)	-	164.1	H-2', H-6' and H-3*
3	6.60 (s)	103.3 (d)	6.76 (s)	102.6	-
4	-	183.1 (s)	-	182.3	H-3*
5	-	160.2 (s)	-	158.5	H-1''
6	-	108.4 (s)	-	107.5	H-1''*
7	-	162.3 (s)	-	161.5	H-1'' and H-1'''
8	-	106.4 (s)	-	105.3	H-1'''*
9	-	156.3 (s)	-	155.1	H-1'''
10	-	105.2 (s)	-	103.8	H-3
1'	-	122.7 (s)	-	121.5	H-3, H-3' and H-5'
2'	8.25 (d, 8.5)	129.6 (d)	8.00 (d, 8.0)	129.0	H-6'
3'	7.23 (d, 8.5)	116.8 (d)	6.92 (d, 8.0)	115.8	H-5'
4'	-	162.7 (s)	-	160.7	H-3'* and H-5'*
5'	7.23 (d, 8.5)	116.8 (d)	6.92 (d, 8.0)	115.8	H-3'
6'	8.25 (d, 8.5)	129.6 (d)	8.00 (d, 8.0)	129.0	H-2'
1''	5.85 (d, 9.5)	76.3 (d)	4.84 (br s)	74.0	H-2''*
2''	4.65 (t, 9.5)	74.4 (d)	3.08-3.88	71.9	H-1''* and H-3''*
3''	4.37 (t, 9.0)	79.7 (d)	3.08-3.88	78.8	H-1''
4''	4.50 (t, 9.5)	70.5 (d)	3.08-3.88	70.5	H-3''* and H-6''
5''	4.06 (br d, 9.5)	82.8 (d)	3.08-3.88	81.8	H-4''*
6''	4.39 (m)	61.1 (t)	3.08-3.88	60.5	H-4''
	4.42 (m)	-	3.08-3.88	-	-
1'''	5.73 (d, 9.5)	75.2 (d)	4.84 (br s)	73.3	H-2'''*
2'''	4.92 (t, 9.5)	72.9 (d)	3.08-3.88	70.8	H-1'''*
3'''	4.44 (t, 9.0)	80.8 (d)	3.08-3.88	77.8	H-1''' and H-2'''*
4'''	4.63 (m)	72.4 (d)	3.08-3.88	69.1	H-3'''*
5'''	4.13 (m)	83.4 (d)	3.08-3.88	80.8	H-4'''* and H-1'''
6'''	4.54 (m)	63.1 (t)	3.08-3.88	61.3	H-4'''
	4.61 (br d, 9.5)	-	3.08-3.88	-	-
HO-5	14.5 (s)	-	-	-	-

*Two-bond correlation.

1.20 Structure Determination of Compound ME18

Compound ME18 was obtained as gum. The negative ESI mass spectrum (Figures 177-178) exhibited an $[M-H]^-$ ion at *m/z* 443.4 and the positive ESIMS showed an $[M+Na]^+$ ion at *m/z* 467.4, suggested the molecular weight 444 and the molecular formula $C_{21}H_{32}O_{10}$. The IR spectrum (Figure 176) showed absorption bands at 3374 (hydroxyl), 1688 (C=C stretching), 1380 (CH_2 bending) and 1164 and 1076 (C-O stretching) cm^{-1}. Compound ME18 exhibited an UV absorption maximum at 257 nm (Figure 175).

The ^1H NMR spectrum of compound ME18 revealed the presence of three methyl groups: two tertiary methyls (δ 0.94, 1.17, each, s, Me-12 and Me-13) and a methyl attached to an olefinic carbon (δ 2.00, s, Me-10); two methylenes at δ 1.79 (m), 1.98 (dd, *J* = 12.5, 6.5 Hz, H-2) and δ 1.79 (m), 2.20 (dd, *J* = 13.5, 5.6 Hz, H-4); an oxymethylene [δ 3.75 (d, *J* = 7.0 Hz), 3.80 (dd, *J* = 7.0, 2.0 Hz), H-11]; a secondary oxymethine group at δ 4.26 (m, H-3); two doublet signals for *trans*-olefinic protons at δ 6.52 (d, *J* = 16.0 Hz, H-7) and 8.00 (d, *J* = 16.0 Hz, H-8) and a olefinic proton singlet at δ 5.76 (H-14) (Table 25 and Figures 179-180). Furthermore, proton signals for a sugar moiety at δ 4.37 (d, *J* = 7.5 Hz, H-1') and δ 3.16-3.87 (6H, m) were also observed.

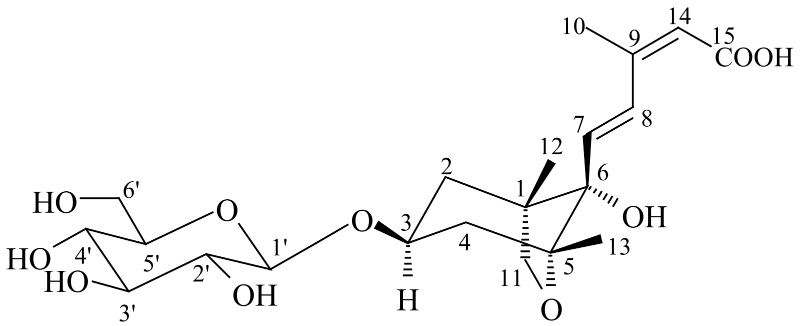
The ^{13}C NMR spectrum (Table 25 and Figure 181) indicated the presence of twenty-one signals, six of which were assigned to a β-D-glucopyranosyl unit and fifteen to the aglycon moiety. From the ^{13}C NMR and HMQC spectra, fifteen signals were identified as three methyls, three methylenes, four methines, four quaternary carbons and a conjugated carboxyl carbon (δ 168.4) (Biemann, 1989). It appeared that the structure of compound ME18 was composed of three units: a cyclohexane ring, an aliphatic side chain and a sugar moiety.

The connectivities of the cyclohexane unit were assigned by analysis of the ^1H-^1H COSY (Figure 183), HSQC (Figure 186) and HMBC (Figures 187-188) spectra. The HMBC spectrum showed three-bond correlation of a hydroxylated quaternary carbon (δ 82.0, C-6) with H-2 and H-4. The ^1H-^1H COSY spectrum exhibited cross peaks for H-2, H-3 and H-4, suggesting the connection of these three protons. The multiplet proton signal with two large coupling constants (*J* = 11.5 Hz) of H-3, indicated its axial orientation. In addition, the two-bond couplings of C-3 (δ 72.7) with H-2 and H-4, C-5 (δ 86.5) with H-4, and C-1 (δ 48.3) with H-2 were displayed. The first methyl at δ 0.94 (3H, s, Me-12) was located at C-1 on the cyclohexane ring as suggested by its ^3J correlation peak with C-6 (δ 82.0) and C-2 (δ 41.6) and its ^2J correlation peak with C-1 in the HMBC spectrum. The three-bond correlations between the methyl signal at δ 1.17 (3H, s, Me-13) and C-4 (δ 41.7) and C-6 (δ 82.0), along with its two-bond coupling with C-5, placed this methoxyl at C-5. The C-1 and C-5 carbons of the cyclohexane ring were linked with an oxygenated methylene bridge as supported by the HMBC correlations (^{3}J coupling) of H-11 [δ 3.75 (d, J =7.0) and δ 3.80 (dd, J = 7.0, 2.0 Hz)] with C-2 and C-5. Moreover, the two-bond coupling between C-1 and H-11 was also observed. The proton signal of H-11 at δ 3.80 appeared as a double doublet with a small coupling constant (J = 2 Hz) due to ω-coupling to the H_{ax}-2. The NOESY interaction (Figures 184-185) between H-3 and H-11 suggested that the orientations of H-3 and the oxygenated methylene bridge were in the same direction.

For the aliphatic side chain assignment, HMBC, ^{1}H-^{1}H COSY and NOESY experiments were carried out, and direct ^{1}H-^{13}C correlation were assigned by the HMQC spectrum. The ^{1}H-^{1}H COSY spectrum showed the correlation between the *trans*-olefinic protons at δ 6.52 (d, J = 16.0 Hz, H-7) and δ 8.00 (d, J = 16.0 Hz, H-8). In the HMBC spectrum, three-bond correlation between H-7 and C-9 (δ 150.4), and H-8 and C-14 (δ 118.1) displayed the connectivities from C-7 to C-8 to C-9 to C-14. The end of the aliphatic side chain was substituted with a carboxylic group as shown by two-bond correlation of the carboxyl carbon (δ 168.4, C-15) with a singlet proton signal of H-14 (δ 5.76). The third methyl (δ 2.00, 3H, s, Me-10) was placed at C-9, as indicated by its HMBC correlation with C-8 and C-14. A NOESY experiment revealed the interactions of Me-10 with H-14 and H-7, suggesting the *trans*-position between Me-10 and the carboxyl group. A NOESY cross peak between H-7 and H_2-2 and H_2-4 was also displayed.

Regarding the sugar unit, the connectivities of seven sugar protons were determined by a ^{1}H-^{1}H COSY experiment and their directly bonded carbons were assigned by an HMQC experiment. The ^{1}H-^{13}C long-range correlation in the HMBC spectrum between anomeric proton H-1' (δ 4.37, d, J = 7.5 Hz) and C-5' (δ 76.8) indicated a pyranose ring with an ether linkage between C-1' and C-5'. The presence of a diaxial-coupling constants (J = 9 Hz) of each sugar proton together with NOESY correlations of H-1' with H-3' and H-5" and of H-2' with H-4' indicated that this sugar was a glucopyranoside. The connection of the three units were determined by HMBC corelations, the aliphatic side chain was attached at C-6 as supported by three-bond couplings of H-8 with C-6, and of H-1 and H-5 with C-7. The HMBC correlation of H-3 with C-1' suggested the placement of the glucose unit at C-3.

Based on the above spectral evidence and by comparison of its ^{1}H and ^{13}C NMR data with reported data (Champavier et al., 1999), compound ME18 was identified as dihydrophaseic acid-4'-*O*-β-D-glucopyranoside [**453**] (Champavier et al., 1999).



[**453**]

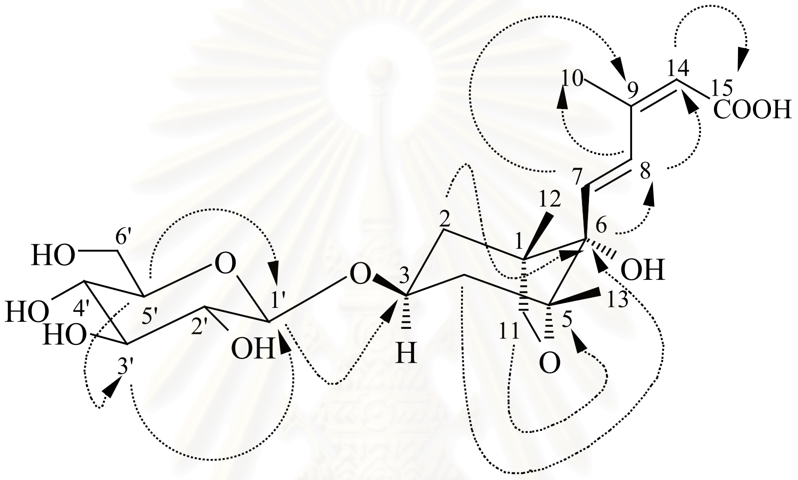


Figure 8 Selected HMBC correlations of compound ME18

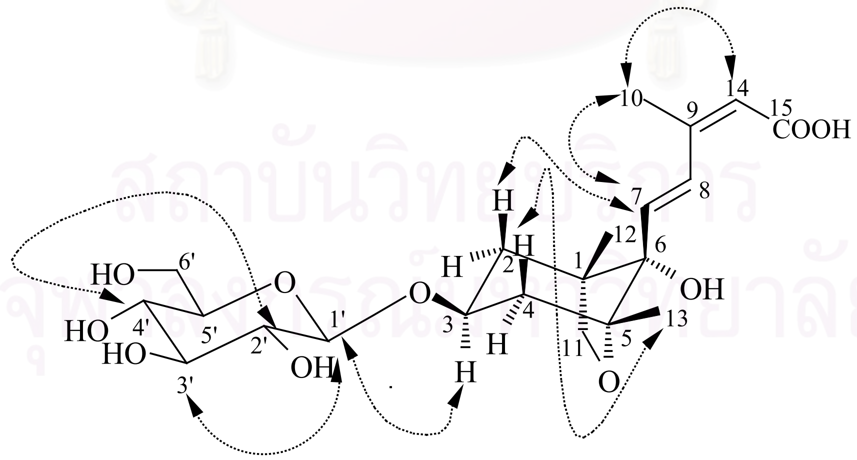


Figure 9 Selected NOESY correlations of compound ME18

Table 25 NMR Spectral data of compound ME 18 and dihydrophaseic acid-4'-*O*-β-D-glucopyranoside (methanol-d_4)

position	Compound ME18		Dihydrophaseic acid-4'-*O*-β-D-glucopyranoside		HMBC (correlation with ^{1}H)
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	^{13}C	
1	-	48.3 (s)	-	49.9	H-7, H-11*, H-2* and Me-12*
2	1.98 (dd, 12.5, 6.5)	41.6 (t)	1.98 (m)	42.8	H-4, H-11, H-3* and Me-12
	1.79 (m)	-	2.19 (m)	-	-
3	4.26 (m)	72.7 (d)	4.25 (m)	73.9	H-1', H-2* and H-4*
4	2.20 (dd, 13.5, 5.6)	41.7 (t)	1.80 (m)	42.9	H-2, H-3* and Me-13
	1.79 (m)	-	1.98 (m)	-	-
5	-	86.5 (s)	-	87.6	H-11, H-7, H-4* and Me-13*
6	-	82.0 (s)	-	83.2	H-2, H-4, H-8, H-11, Me-12, Me-13 and H-7*
7	6.52 (d, 16.0)	133.9 (d)	6.49 (d, 15.9)	134.6	H-8*
8	8.00 (d, 16.0)	130.7 (d)	7.96 (d, 15.9)	132.0	H-7*, H-14 and Me-10
9	-	150.4 (s)	-	150.4	H-7, H-8* and Me-10
10	2.00 (s)	20.1 (q)	2.07 (s)	21.2	H-8 and H-14
11	3.75 (d, 7.0)	76.0 (t)	3.75 (d, 7.4)	77.2	Me-12
	3.80 (dd, 7.0, 2.0)	-	3.80 (dd, 7.4, 2.1)	-	-
12	0.94 (s)	15.2 (q)	0.94 (s)	16.3	-
13	1.17 (s)	18.5 (q)	1.17 (s)	19.7	-
14	5.76 (s)	118.1 (d)	5.78 (s)	120.5	H-8 and Me-10
15	-	168.4 (s)	-	Not detected	H-14*
1'	4.37 (d, 7.5)	101.9 (d)	4.36 (d, 7.8)	103.1	H-3 and H-2'*
2'	3.16 (dd, 9.0, 8.0)	73.9 (d)	3.14 (dd, 9.1, 7.8)	75.1	H-3'*
3'	3.35 (m)	76.9 (d)	3.30 (m)	78.1	H-2'*
4'	3.28 (m)	70.5 (d)	3.30 (m)	71.7	-
5'	3.28 (m)	76.8 (d)	3.28 (m)	78.0	H-1', H-4'* and H-6'*
6'	3.67 (dd, 11.5, 5.0)	61.6 (t)	3.67 (dd, 11.9, 5.5)	62.8	H-4'
	3.87 (11.5, 1.5)	-	3.87 (dd, 11.9, 1.5)	-	-

*Two-bond coupling.

1.21 Structure Determination of Compound ME19

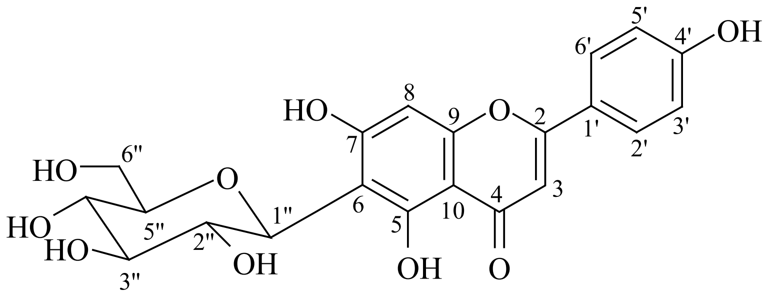
Compound ME19 was obtained as a yellow powder. It showed an $[M-H]^-$ ion at *m/z* 431.4 in the negative ESIMS (Figure 191) and an $[M+H]^+$ ion at *m/z* 433.2 in the positive ESIMS (Figure 192), corresponding to the molecular weight 432 and the molecular formula $C_{21}H_{20}O_{10}$. The IR bands of hydroxyl (3369 cm^{-1}), conjugated carbonyl (1654 cm^{-1}) (Figure 190) and the UV absorptions at 211, 269 and 329 nm (Figure 189) were similar to those of compound ME17, suggestive of a flavone skeleton with a chelated hydroxyl at C-5.

The ^1H NMR spectrum (Table 26 and Figure 193) confirmed the existence of the flavone nucleus with signals for H-3 at δ 6.71 and a chelated hydroxyl at δ 13.98. It also exhibited a pair of doublets, 2H each at δ 8.29 and δ 7.25 (*J* = 8.5 Hz), suggesting that ring B is oxygenated at C-4. Furthermore, an aromatic singlet signal at δ 6.78 (H-8) and aliphatic proton signals for a sugar moiety [δ 5.94 (1H, d, *J* = 10.0 Hz), 5.04 (1H, m), 4.67 (1H, t, *J* = 9.5 Hz), 4.63 (1H, dd, *J* = 13.5, 8.5 Hz), 4.54 (1H, br d, *J* = 12.5 Hz), 4.48 (1H, t, *J* = 8.5 Hz) and 4.23 (1H, m)] were also observed.

The connectivities of the seven sugar protons were determined by a ^1H-^1H COSY experiment (Figure 194), and their directly bonded carbons were assigned by an HSQC experiment (Figure 196). The coupling constant of *J* = 10 Hz for H-1" suggested that H-1" was an β-anomeric proton (Agrawal, 1992). The sugar moiety was attached to the flavone nucleus with C-linkage as supported by the chemical shift of C-1" at δ 74.8 (Agrawal, 1992).

Comparison of its ^1H and ^{13}C NMR data with those of compound ME17 indicated that compound ME19 differed from compound ME17 by only one glucose unit, thereby suggesting two possible structures: apigenin-6-β-D-glucopyranoside or apigenin-8-β-D-glucopyranoside. In a ROESY experiment (Figure 195), the cross peaks were not observed for H-2" and H-2' and H-6', implying that the glucose unit should not be located at C-8. Compound ME19 was identified as apigenin-6-β-D-glucopyranoside by TLC analysis of its R_f value in comparison with reported data (R_f 0.39, silica gel, 10% acetic acid; R_f 0.20 for apigenin-8-β-D-glucopyranoside) (Gentili and Horowitz, 1968). Furthermore, the doubling of B ring proton signals was not observed for this compound, confirming the absence of the glucose moiety at C-8 (Harborne, 1994).

By analysis of the above spectroscopic data and comparison of its ^1H and ^{13}C NMR values with previously reported data (Ramarathnam *et al*., 1989), compound ME19 was thus identified as apigenin-6-*O*-β-D-glycoside or isovitexin [**454**]. This compound has been reported to be present widely in plants such as *Citrullus colocynthis* (Maatooq *et al*., 1997) and tea (Engelhardt, Finger and Kuhr, 1993).



[**454**]

Table 26 NMR Spectral data of compound ME19 (pyridine-d_5) and isovitexin (methanol-d_4)

position	Compound ME19		Isovitexin		HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	^{13}C	(correlation with ^{1}H)
2	-	165.0 (s)	-	165.9	H-3*
3	6.71 (s)	103.4 (d)	6.49 (s)	103.8	-
4	-	Not detected	-	183.9	-
5	-	157.2 (s)	-	161.5	H-1"
6	-	106.0 (s)	-	109.1	H-1"*
7	-	164.5 (s)	-	164.6	H-1" and H-8*
8	6.78 (s)	99.8 (d)	6.56 (s)	95.3	-
9	-	163.2 (s)	-	158.4	H-8*
10	-	104.1 (s)	-	105.1	H-3 and H-8
1'	-	121.3 (s)	-	123.0	H-3, H-3' and H-5'
2'	8.29 (d, 8.5)	128.8 (d)	7.78 (d, 9.0)	129.3	Not detected
3'	7.25 (d, 8.5)	116.8 (d)	6.95 (d, 9.0)	116.9	Not detected
4'	-	164.5 (s)	-	162.5	Not detected
5'	7.25 (d, 8.5)	116.8 (d)	6.95 (d, 9.0)	116.9	Not detected
6'	8.29 (d, 8.5)	128.8 (d)	7.78 (d, 9.0)	129.3	Not detected
1"	5.94 (d, 10.0)	74.8 (d)	4.21 (d)	75.3	Not detected
2"	5.04 (m)	72.4 (d)	3.78-3.93 (m)	72.7	Not detected
3"	4.48 (t, 8.5)	80.0 (d)	3.78-3.93 (m)	80.1	Not detected
4"	4.67 (t, 9.5)	71.0 (d)	3.78-3.93 (m)	71.8	Not detected
5"	4.23 (m)	82.0 (d)	3.78-3.93 (m)	82.5	Not detected
6"	4.63 (dd, 13.5, 8.5)	62.0 (t)	3.78-3.93 (m)	62.9	Not detected
	4.54 (br d, 12.5)	-	3.78-3.93 (m)	-	-
HO-5	13.98 (s)	-	-	-	-

*Two-bond coupling.

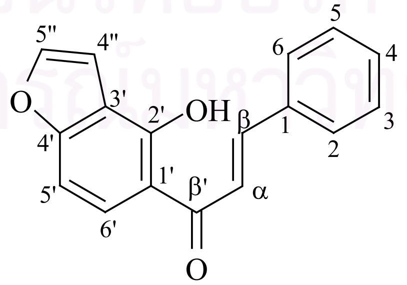
1.22 Structure Determination of Compound ME20

Compound ME20, showed a [M^+] ion peak at *m/z* 264, analyzed for $C_{18}H_{14}O_4$. The IR spectrum (Figure 199) exhibited absorption bands for chelated hydroxyl (2956 cm^{-1}), conjugated carbonyl (1641 cm^{-1}) and olefinic (1599 cm^{-1}) functionalities. The UV spectrum (Figure 198) showed absorptions at 214, 249, 263 and 307 nm.

The *trans*-olefinic proton signals at δ 7.76 (d, J = 15.3 Hz, H-α) and δ 7.99 (d, J = 15.3 Hz, H-β), a chelated hydroxyl proton at δ 14.00 (s) and the ^{13}C NMR (Table 27 and Figure 202) signal at δ 193.4 (C-β') suggested that compound ME22 was a chalcone with hydroxyl at C-2'. The ^{1}H NMR spectrum also revealed the presence of two doublet proton signals for a furan ring at δ 7.08 (d, J = 2.1 Hz, H-4") and δ 7.64 (d, J = 2.1 Hz, H-5") and signals for unsubstituted B ring at δ 7.73 (2H, m, H-2 and H-4) and δ 7.49 (3H, m, H-3, H-4 and H-5) (Table 27 and Figure 201).

In the EI mass spectrum, the fragment ions at *m/z* 161 and 103 resulting from α-cleavage suggested the location of the furan ring and the hydroxyl on ring A and confirmed the existence of an unsubstituted B ring (Drewes, 1974). The appearance of the two proton doublet signals with *ortho*-aromatic coupling at δ 7.14 (d, J = 9.0 Hz, H-5') and δ 7.91 (d, J = 9.0 Hz, H-6'), together with the HMBC correlation (Figures 205-206) between C-β' (δ 193.4) and H-6' clearly indicated that the furan ring was fused in an angular form on ring A at C-3' and C-4'. This was confirmed by the three-bond correlation of C-4' (δ 159.7) with H-4", H-5" and H-6'. The coupling between H-5' and H-6' was also observed in the ^{1}H-^{1}H COSY spectrum (Figure 203).

From all of the above spectral data, it was concluded that compound ME20 was 1-(4-hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one [**289**]. It was first isolated from the roots of *Millettia ovalifolia* (Saxena *et al.*, 1987).



[**289**]

Table 27 NMR Spectral data of compound ME20 and 1-(4-hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one (CDCl$_3$)

position	Compound ME20		1-(4-Hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
1	-	134.7	-	H-α, H-3 and H-5
2	7.73 (m)	128.6	7.34-8.00	H-β, H-4 and H-6
3	7.49 (m)	129.0	7.34-8.00	H-5
4	7.49 (m)	130.8	7.34-8.00	H-2 and H-6
5	7.49 (m)	129.0	7.34-8.00	H-3
6	7.73 (m)	128.6	7.34-8.00	H-β, H-2 and H-4
1'	-	114.4	-	H-5' and HO-2'
2'	-	160.3	-	H-6' and HO-2'*
3'	-	117.8	-	H-4''*, H-5'' and HO-2'
4'	-	159.7	-	H-6', H-4'' and H-5''
5'	7.14 (d, 9.0)	103.8	7.18 (d, 9.0)	-
6'	7.91 (d, 9.0)	126.0	7.78 (d, 9.0)	-
α	7.76 (d, 15.3)	120.7	7.34-8.00	H-β*
β	7.99 (d, 15.3)	144.8	7.34-8.00	H-2 and H-6
β'	-	193.4	-	H-α*, H-β and H-6'
4''	7.08 (d, 2.1)	105.1	7.10 (d, 2.0)	-
5''	7.64 (d, 2.1)	144.5	7.68 (d, 2.0)	-
HO-2'	14.00 (s)	-	13.95	-

*Two-bond coupling.

1.23 Structure Determination of Compound ME21

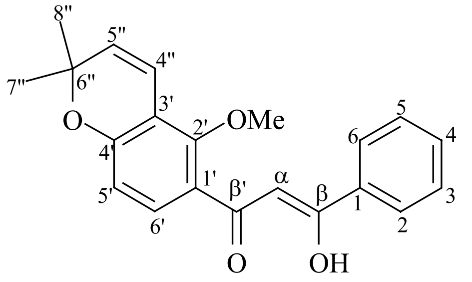
Compound ME21 was obtained as yellowish needles. It showed an [M$^+$] ion peak at *m/z* 336 in the EI mass spectrum (Figure 209), corresponding to the molecular formula $C_{21}H_{20}O_4$. The UV spectrum showed a maximal absorption at 359 nm (Figure 207) and the IR spectrum exhibited bands at 3061 (H-bonded OH), 1636 (conjugated C=O), 1592 (conjugated C=C) and 1219 and 1166 (C-O stretching) cm^{-1} (Figure 208).

The ^{1}H NMR spectrum (Table 28 and Figure 210) showed a chelated hydroxyl group (δ 15.9,s) and an olefinic proton signal (δ 7.20, s, H-α) which correlated with a carbon at δ 96.7 in the HMQC spectrum, suggesting that compound ME22 was a hydroxy chalcone derivative in a *Z*-configuration. This was also confirmed by the appearance of ^{13}C NMR signals at δ 184.4 (C-β) and 185.2 (C-β'), due to tautomerism effect (Table 28 and Figure 211). The ^{13}C NMR and HSQC spectra (Figure 214) showed 21 carbon signals, analyzed for two methyls, one methoxyl, ten methines and eight quaternary carbons. Comparison of the ^{13}C NMR spectrum with that of compound ME5 indicated that this compound differed from compound ME5 by one methoxyl on ring A.

The ^{1}H NMR spectrum showed similar signals to those of compound ME5, with the presence of a dimethylpyran ring [δ 1.60 (6H, Me x 2), δ 5.74 (d, *J* = 9.6 Hz, H-5") and δ 6.70 (d, *J* = 9.6 Hz, H-6")] and an unsubstituted B ring [δ 8.02 (2H, m, H-2 and H-6) and δ 7.54 (3H, m, H-3, H-4 and H-5)]. The appearance of two *ortho*-coupled aromatic proton at δ 6.72 (d, *J* = 8.7 Hz, H-5') and δ 7.78 (d, *J* = 8.7 Hz, H-6'), which showed ^{1}H-^{1}H COSY correlation (Figure 212), suggested the lack of 5'-OMe. The assignment of H-6' was based on its long-range (^3J) coupling to the carbonyl carbon (C-β', δ 185.2) in the HMBC spectrum (Figure 215).

In the EIMS, the fragment ion at *m/z* 217 confirmed the location of a dimethylpyran ring and a methoxyl on ring A (Drewes, 1974). The NOESY correlations of the methoxyl with H-α and H-4" supported the attachment of the methoxyl at C-2' and a dimethylpyran ring at C-3' and C-4' (Figure 213). A three-bond correlation was also found between C-4 (δ 157.6) and H-2.

Based on the above spectral evidence and by comparison of its ^{1}H NMR data with previously published data (Rao and Raju, 1984), compound ME21 was identified as purpurenone [**455**]. This compound was first reported from the roots of *Tephrosia purpurea* (Rao and Raju, 1984).



[**455**]

Table 28 NMR Spectral data of compound ME21 and purpurenone ($CDCl_3$)

position	Compound ME21		Purpurenone	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
1	-	135.7 (s)	-	H-3 and H-5
2	8.02 (m)	127.1 (d)	7.90 (m)	H-4 and H-6
3	7.54 (m)	128.5 (d)	7.42 (m)	H-5
4	7.54 (m)	132.1 (d)	7.42 (m)	H-2 and H-6
5	7.54 (m)	128.5 (d)	7.42 (m)	H-3
6	8.02 (m)	127.1 (d)	7.90 (m)	H-2 and H-4
1'	-	121.8 (s)	-	H-5'
2'	-	156.2 (s)	-	H-6' and MeO-2'
3'	-	115.1 (s)	-	H-5' and H-5''
4'	-	157.6 (s)	-	H-6' and H-4''
5'	6.72 (d, 8.7)	113.0 (d)	6.60 (d, 9.0)	-
6'	7.78 (d, 8.7)	130.7 (d)	7.66 (d, 9.0)	-
α	7.20 (s)	96.7 (d)	7.09 (s)	-
β	-	184.4 (s)	-	H-2 and H-6
β'	-	185.2 (s)	-	H-6' and H-α*
4''	6.70 (d, 9.6)	116.4 (d)	6.58 (d, 10.0)	-
5''	5.74 (d, 9.6)	130.7 (d)	5.62 (d, 10.0)	-
6''	-	77.0 (s)	-	H-4'', H-5''*, H-7'' and H-8''
7''	1.60 (s)	28.1 (q)	1.42 (s)	H-8''
8''	1.60 (s)	28.1 (q)	1.42 (s)	H-7''
MeO-2'	3.84 (s)	62.6 (q)	3.77 (s)	-
HO-β	15.9 (s)	-	13.50 (s)	-

*Two-bond coupling.

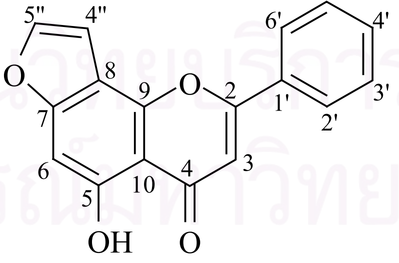
1.24 Structure Determination of Compound ME22

Compound ME22 was isolated as a yellowish powder. The EIMS (Figure 218) showed the $[M^+]$ ion peak at *m/z* 278, analyzed for $C_{17}H_{10}H_4$. The IR spectrum showed absorption bands at 2924 (chelated hydroxyl), 1666 (conjugated carbonyl) and 1249 and 1141 (ether linkage) cm^{-1} (Figure 217). The presence in the UV spectrum of intense bands at 217 and 280 nm (Figure 216) and the two singlet proton signals at δ 12.7 (chelated hydroxyl) and δ 6.83 (1H, s, H-3) in the ^1H NMR spectrum were indicative of a flavone derivative with a hydroxyl group at C-5 (Markham, 1982).

The ^1H NMR spectrum (Table 29 and Figure 219) also displayed a two-proton multiplet centred at δ 7.97 (2H, H-2' and H-6') and a three-proton multiplet centred at δ 7.61 (3H, H-3', H-4' and H-5'), suggesting an unsubstituted B ring. Typical two one-proton doublets at δ 7.08 and 7.64 which correlated to the ^{13}C NMR signals at δ 103.5 and δ 144.2 (oxygenated carbon) in the HSQC spectrum (Figure 222) could be assigned to the H-4" and H-5" of a furan ring, respectively.

The fragment ions at *m/z* 176 and 102 due to RDA cleavage of ring C in the EIMS confirmed an unsubstituted B ring and suggested the placement of a methylenedioxy group and a hydroxyl group on ring A (Drewes, 1974). The presence of a sharp singlet proton signal at δ 6.98, assignable to H-6, together with the HMBC correlations between C-8 (δ 108.6) and H-6 and H-5" suggested the fusion of the furan ring at C-7 and C-8 in an angular form on ring A (Figure 223).

By analysis of above spectroscopic studies and comparison with reported ^1H NMR data (Talapatra, Mallik, and Talapatra, 1980), this compound was identified as pongaglabol [**337**]. It was first found in the flowers of *Pongamia glabra* (Talapatra, Mallik, and Talapatra, 1980).



[**337**]

Table 29 NMR Spectral data of compound ME22 and pongaglabol ($CDCl_3$)

position	Compound ME22		Pongaglabol	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
2	-	163.6 (s)	-	H-3*, H-2' and H-6'
3	6.83 (s)	106.6 (d)	6.80 (s)	-
4	-	183.6 (s)	-	H-3*
5	-	158.5 (s)	-	H-6* and HO-5*
6	6.98 (s)	95.9 (d)	6.95 (s)	HO-5
7	-	159.4 (s)	-	H-6*, H-4'' and H-5''
8	-	108.6 (s)	-	H-6, H-4''* and H-5''
9	-	150.0 (s)	-	-
10	-	107.6 (s)	-	H-3, H-6 and HO-5
1'	-	131.1 (s)	-	H-3, H-3' and H-5'
2'	7.97 (m)	126.2 (d)	7.90-8.01 (m)	H-4' and H-6'
3'	7.61 (m)	129.2 (d)	7.90-8.01 (m)	H-5'
4'	7.61 (m)	132.0 (d)	7.90-8.01 (m)	H-2' and H-6'
5'	7.61 (m)	129.2 (d)	7.90-8.01 (m)	H-3'
6'	7.97 (m)	126.2 (d)	7.90-8.01 (m)	H-4' and H-6'
4''	7.08 (d, 2.0)	103.5 (d)	7.04 (dd, 2.1, 0.9)	-
5''	7.64 (d, 2.0)	144.2 (d)	7.53-7.61 (m)	-
HO-5	12.70 (s)	-	12.73 (s)	-

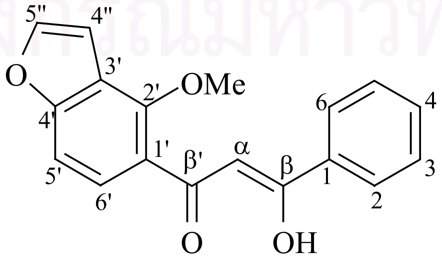
*Two-bond coupling.

1.25 Structure Determination of Compound ME23

Compound ME23 has a molecular formula of $C_{18}H_{14}O_4$, as indicated by the molecular ion peak at *m/z* 294 in the EI mass spectrum (Figure 226). The UV absorptions at 237 and 347 nm (Figure 224) and the IR bands displaying the presence of H-bonded hydroxyl (3459 cm^{-1}), conjugated carbonyl (1599 cm^{-1}) and aromatic (1565 cm^{-1}) functional groups (Figure 225) were indicative of a chalcone having a chelated hydroxyl group (Markham, 1982).

In the ^1H NMR spectrum (Table 30 and Figure 227), a sharp singlet proton at δ 7.10 (s, H-α) which correlated with a carbon at δ 97.2 in HSQC spectra (Figure 230) and the ^{13}C NMR signals at δ 184.9 (C-β) and δ 183.9 (C-β') supported a β-hydroxychalcone skeleton of compound ME23. The ^1H NMR spectrum also exhibited signals for a furan ring at δ 7.04 (1H, d, *J* = 2.1 Hz, H-4") and δ 7.67 (1H, d, *J* = 2.1 Hz, H-5"), a methoxyl group at δ 4.17 (s) and for an unsubstituted B ring at δ 7.99 (2H, m, H-2 and H-6), 7.50 (2H, m, H-3 and H-5) and 7.56 (1H, m, H-4).

The ^{13}C NMR (Table 30 and Figure 228) and HMQC spectra displayed 18 carbon signals, corresponding to one methoxyl, ten methines and seven quaternary carbons. In the EIMS, the α-cleavage ion at *m/z* 175 indicated the location of a furan ring and a methoxyl group on ring A (Drewes, 1974). The appearance of AB splitting system at δ 7.33 (1H, d, *J* = 8.7 Hz, H-5') and δ 7.89 (1H, d, *J* = 8.7 Hz, H-6'), along with the HMBC correlations (Figure 231) of C-2' (δ 154.8) with H-6', and C-4' (δ 159.8) with H-6', H-4" and H-5" suggested the position of a furan ring at C-3' and C-4' and a methoxyl group at C-2'. The assignment of H-6' was done on the basis of its three-bond correlation with C-β' in the HMBC spectrum. A ^1H-^1H COSY experiment (Figure 229) was used for identifying each pair of *ortho*-coupled aromatic protons, as follows: H-5'/H-6', H-4"/H-5" and H-2 (H-6)/H-3 (H-5). Compound ME23 was identified as pongamol [**315**] based on the above spectral evidence and by comparison of the NMR data with previously reported data (Parmar *et al*., 1989). Pongamol was first isolated from the whole plant of *Tephrosia purpurea* (Parmar *et al*., 1989).



[**315**]

Table 30 NMR Spectral data of compound ME23 and pongamol ($CDCl_3$)

position	Compound ME23		Pongamol		HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	^{13}C	(correlation with ^{1}H)
1	-	136.7 (s)	-	135.70	H-α, H-3 and H-5
2	7.99 (m)	128.1 (d)	7.54 (m)	128.62	H-4 and H-6
3	7.50 (m)	129.6 (d)	7.48 (m)	127.16	H-5
4	7.56 (m)	133.1 (d)	7.48 (m)	132.08	H-2 and H-6
5	7.50 (m)	129.6 (d)	7.48 (m)	127.16	H-3
6	7.99 (m)	128.1 (d)	7.94 (m)	128.62	H-2 and H-4
1'	-	123.2 (s)	-	119.60	H-α and H-3'
2'	-	154.8 (s)	-	158.78	H-6', H-4" and MeO-2'
3'	-	120.5 (s)	-	122.18	H-3', H-4"* and H-5"
4'	-	159.8 (s)	-	152.78	H-5'*, H-6', H-4" and H-5"
5'	7.33 (d, 8.7)	107.9 (d)	7.28 (d, 9.0)	105.25	-
6'	7.89 (d, 8.7)	127.4 (d)	7.85 (d, 9.0)	122.59	-
α	7.18 (s)	98.7 (d)	7.16 (s)	97.97	-
β	-	185.5 (s)	-	184.32	H-α*, H-2 and H-6
β'	-	187.4 (s)	-	186.14	H-α* and H-6'
4"	7.02 (d, 2.3)	106.1 (d)	6.96 (d, 2.0)	107.09	H-5"*
5"	7.64 (d, 2.3)	145.9 (d)	7.60 (d, 2.0)	144.85	H-4"*
MeO-2'	4.19 (s)	61.8 (q)	4.12 (s)	61.76	-

*Two-bond coupling.

1.26 Structure Determination of Compound ME24

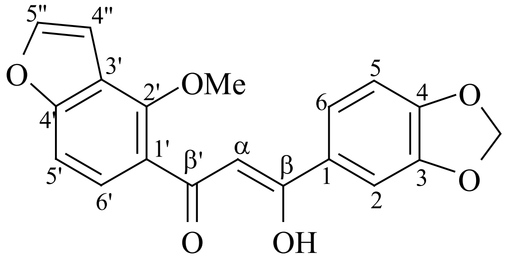
Compound ME24 was obtained as yellow needles. The molecular formula was determined as $C_{19}H_{14}O_6$ by HREIMS, with its [M^+] ion at *m/z* 338.07878 (calcd 338.07904). The UV absorptions at 237 and 361 nm (Figure 232) and the broad shallow band between 3440 and 3078 cm^{-1} (OH group) and the band at 1600 (conjugated carbonyl) in the IR spectrum (Figure 233) were recognized as a chalcone having a chelated hydroxyl group (Markham, 1982).

This was supported by a ^{1}H NMR singlet proton signal at δ 7.10 (H-α) (Table 31 and Figure 235) and three ^{13}C NMR signals at δ 97.2 (C-α), δ184.9 (C-β) and δ183.9 (C-β') (Table 31 and Figure 236), due to keto-enol tautomerism effect. The ^{1}H NMR spectrum, in addition, revealed the presence of a methylenedioxy group (δ 6.10, s, 2H), a methoxyl group (δ 4.17, s, 3H) and a furan ring (δ 7.64 and 7.04, each d, J = 2.1 Hz). Moreover, a pair of aromatic protons with *ortho*-coupling (δ 7.90 and 7.35, each d, 1H, J = 8.7 Hz, H-6' and H-5'), and an ABM (3H) splitting system consisting of a doublet at δ 6.93 (J = 8.1 Hz, H-5), a broad singlet at δ 7.50 (H-2) and a broad doublet at δ 7.63 (J = 8.1 Hz, H-6) were observed.

In the EIMS, the fragment ion at *m/z* 175 formed by α-β' fission suggested the placement of the methoxyl group and the furan moiety on ring A. (Drewes, 1974). The methylenedioxy unit should then be placed on ring B, as shown by the fragment ion at *m/z* 149, which was generated by α-β fission. This was supported by the HMBC correlation (Figures 240-241) of H-2 and H-6 with C-β (δ 184.9).

The NOESY interaction (Figure 238) of the methoxyl protons with H-4'' and H-α placed the methoxyl group at C-2'. The appearance of H-6' and H-5' as a pair of *ortho*-coupled doublets and the HMBC correlation of H-6' with C-β' (δ 183.9) and C-4' (δ 158.5) indicated that the furan ring should be fused in an angular position at C-4' (oxygenated) and C-3'. Interactions through ^3J coupling of C-4' with H-5'' and H-4'', and of C-3' (δ 119.6) with H-5'' were also observed.

Compound ME24 identified as ovalitenone [**312**] by analysis of the above NMR spectral data and comparison of its ^{1}H NMR with previously published data (Gupta and Krishnamurti, 1997b). Ovalitenone [**312**] was first isolated from seeds of *Millettia ovalifolia* (Gupta and Krishnamurti, 1997b).



[**312**]

Table 31 NMR Spectral data of compound ME24 and ovalitenone ($CDCl_3$)

position	Compound ME24		Ovalitenone	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
1	-	130.3 (s)	-	H-5
2	7.50 (br s)	107.2 (d)	7.05-7.37 (m)	H-6
3	-	148.1 (s)	-	H-5, H-2* and -OCH$_2$O-
4	-	151.2 (s)	-	H-2, H-6 and -OCH$_2$O-
5	6.93 (d, 8.1)	108.2 (d)	6.86 (d, 8.0)	-
6	7.63 (d, 8.1)	122.8 (d)	7.05-7.37 (m)	H-2
1'	-	121.9 (s)	-	H-5'
2'	-	153.6 (s)	-	H-6' and MeO-2'
3'	-	119.6 (s)	-	H-5', H-4''* and H-5''
4'	-	158.5 (s)	-	H-6', H-4'' and H-5''
5'	7.35 (d, 8.7)	107.0 (d)	6.73 (d, 9.0)	-
6'	7.90 (d, 8.7)	126.3 (d)	7.70 (d, 9.0)	-
α	7.10 (s)	97.2 (d)	7.05-7.37 (m)	-
β	-	184.9 (s)	-	H-α, H-2 and H-6
β'	-	183.9 (s)	-	H-α and H-6'
4''	7.04 (d, 2.1)	105.2 (d)	6.83 (d, 2.0)	H-5''*
5''	7.67 (d, 2.1)	144.8 (d)	7.45 (d, 2.0)	-
MeO-2'	4.17 (s)	61.1 (q)	4.02 (s)	-
-OCH$_2$O-	6.10 (s)	101.8 (t)	6.0 (s)	-

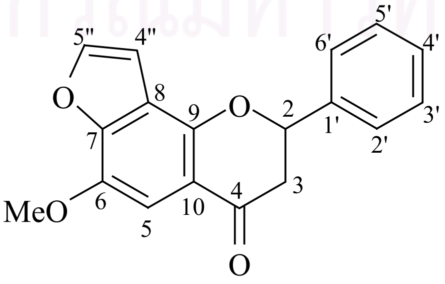
*Two-bond coupling.

1.27 Structure Determination of Compound ME25

Compound ME25, colorless needles, showed its molecular ion [M^+] at *m/z* 294.08423 in the HREIMS, indicating a molecular of $C_{18}H_{14}O_4$ (calcd 294.08920). The IR spectrum (Figure 243) showed absorption bands for carbonyl (1679 cm^{-1}) and ether (1246, 1212 cm^{-1}) functionalities. The UV absorptions at 234, 247 and 341 nm (Figure 242) were indicative of a flavanone skeleton. (Markham, 1982)

In the ^{1}H NMR spectrum (Table 32 and Figure 246), the aliphatic proton signals at δ 2.90 (*dd*, *J* = 16.8 and 3.0 Hz), δ 3.12 (dd, *J* = 13.2 and 16.8 Hz) and δ 5.57 (dd, *J* = 3.0 and 13.2 Hz) are typical for H-3eq, H-3ax and H-2, respectively. This was confirmed by the HSQC spectrum (Figure 250) in which the two former protons correlated with a carbon at δ 44.3 ppm and the latter (δ 5.57) exhibited a cross peak with a carbon at δ 80.4 ppm. The ^{1}H NMR spectrum of compound ME25 also revealed the presence of a methoxyl group (δ 3.99, s, 3H), and a furan ring, as evidenced by two one-proton doublets (*J* = 2.1 Hz) at δ 6.91 (H-4") and δ 7.61 (H-5"). A two-proton multiplet centred at δ 7.51 and a three-proton multiplet centred at δ 7.41 suggested that ring B was unsubstituted.

In the EIMS, the fragment ions at *m/z* 190 and 104 resulting from *retro*-Diels-Alder cleavage of ring C suggested the placement of the furan ring and the methoxyl on ring A (Drewes, 1974). The methoxyl should be situated at C-6, as shown by its NOESY interaction with the proton at δ 7.27 (1H, *s*, H-5) and the HMBC correlation of H-5 with C-4 (δ 191.2). The position of the furan ring on ring A was determined by the HMBC connection (Figure 251) between H-5 and C-7. The CD spectrum showed a positive Cotton effect at 350 nm and a negative one at 281 nm, consistent with the 2*S*-configuration (Gaffield, 1970; Yenesew *et al*., 1998). Based on above spectral evidence, compound 25 was identified as a new flavonoid, (-)-(2*S*)-6-methoxy-[2",3":7,8]-furanoflavanone [**456**] (Sritularak *et al*., 2002b).



[**454**]

Table 32 NMR Spectral data of compound ME25 ($CDCl_3$)

position	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	HMBC (correlation with ^{1}H)
2	5.57 (dd, 13.2, 3.0)	80.4 (d)	H-2' and H-6'
3	3.12 (dd, 16.8, 13.2)	44.3 (t)	-
	2.90 (dd, 16.8, 3.0)	-	-
4	-	191.2 (s)	H-2, H-3* and H-5
5	7.27 (s)	102.1 (d)	-
6	-	141.5 (s)	MeO-6
7	-	149.8 (s)	H-5, H-4" and H-5"
8	-	119.2 (s)	H-5"
9	-	151.6 (s)	H-5 and H-4"
10	-	115.3 (s)	H-3
1'	-	138.8 (s)	H-3, H-3' and H-5'
2'	7.51 (m)	126.2 (d)	H-4' and H-6'
3'	7.41 (m)	128.8 (d)	H-5'
4'	7.41 (m)	128.8 (d)	H-2' and H-6'
5'	7.41 (m)	128.8 (d)	H-3'
6'	7.51 (m)	126.2 (d)	H-2' and H-6'
4"	6.91 (d, 2.1)	105.3 (d)	H-5"*
5"	7.61 (d, 2.1)	145.3 (d)	H-4"*
MeO-6	3.99 (s)	53.3 (q)	-

*Two-bond coupling.

1.28 Structure Determination of Compound ME26

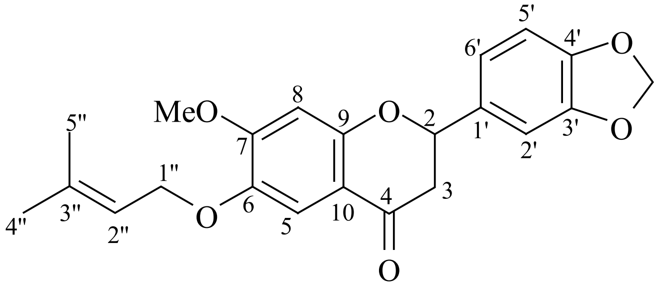
Compound ME26 was obtained as colorless needles. It showed a molecular [M^+] ion at *m/z* 382 in the EIMS (Figure 254), corresponding to $C_{22}H_{22}O_6$. The IR spectrum (Figure 253) showed absorption bands for conjugated carbonyl (1677 cm^{-1}), olefin (1611 cm^{-1}) and ether (1179, 1036 cm^{-1}) functionalities. The UV spectrum exhibited absorptions at 240, 277 and 338 nm (Figure 252), suggestive of a flavanone moiety (Markham, 1982).

The ^1H NMR (Table 33 and Figure 255) spectrum showed characteristic signals for a flavanone nucleus at δ 2.81 (dd, J = 16.8, 2.7 Hz), δ 3.16 (dd, J = 16.8, 13.2 Hz) and δ 5.40 (dd, J = 13.2, 2.7 Hz), assignable to H-3eq, H-3ax and H-2, respectively. This was confirmed by the HSQC spectrum (Figure 259) in which the two protons correlated with a carbon at δ 44.2 and the H-2 proton showed a cross peak with a carbon at δ 80.1.

The ^{13}C NMR (Table 33 and Figure 256) and HMQC spectra showed 22 carbons, analyzed for two methyls, one methoxyl, three methylenes, seven methines and nine quaternary carbons. Comparison of its ^1H and ^{13}C NMR spectrum with compound ME10 showed that it differed from compound ME10 only by lacking unsaturation at C-2 and C-3. The proton signals similar to those of compound ME10 were as follows: a methylenedioxy group at δ 6.40 (s), a methoxyl group at δ 3.93 (3H, s), a γ,γ-dimethylallyloxy group at δ 1.80 (3H, s, Me-5''), δ 1.83 (3H, s, Me-4''), δ 5.56 (1H, t, J = 6.6 Hz, H-2'') and δ 4.61 (2H, d, J = 6.6 Hz, H-1'') and two sharp proton singlets at δ 7.38 and δ 6.54 (H-5 and H-8). The assignment of H-5 was based on its HMBC correlation (Figure 260) with C-4 (δ 190.6).

In the ^1H NMR spectrum, the presence of an ABM spin system at δ 7.04 (br s, H-2'), δ 6.92 (br d, J = 7.8 Hz, H-6') and δ 6.88 (d, J = 7.8 Hz, H-5') and the fragment ion at *m/z* 148 suggested the placement of the methylenedioxy group at C-3' and C-4' of ring B (Drewes, 1974). In the EIMS, the [M^+] through the loss of the prenyl group with H transfer gave a fragment ion at *m/z* 314, and this ion then underwent RDA cleavage of ring C to give an ion at *m/z* 166, thereby confirming the presence of the prenyloxy group on ring A (Drewes, 1974). To determine the positions of the methoxyl group and the prenyloxy group on ring A, a NOESY experiment was carried out (Figure 258). The NOE interactions of a methoxyl at δ 3.93 with H-8 placed this methoxyl at C-7. The prenyloxy group should be located at C-6 according to the NOE effects of H-1'' of prenyloxyl with H-5.

Based on the above spectral data, compound ME26 was identified as ponganone V [**457**]. This compound was first separated from the root bark of *Pongamia pinnata* (Tanaka *et al*., 1992).



[**457**]

Table 33 NMR Spectral data of compound ME26 and ponganone V (CDCl$_3$)

position	Compound ME26		Ponganone V	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
2	5.40 (dd, 13.2, 2.7)	80.1 (d)	5.35 (dd, 14.0, 3.0)	H-3*, H-2' and H-6'
3	3.05 (dd, 16.8, 13.2)	44.2 (t)	3.01 (dd, 16.0, 14.0)	-
	2.81 (dd, 16.8, 2.7)	-	2.79 (dd, 16.0, 3.0)	-
4	-	190.6 (s)	-	H-3* and H-5
5	7.38 (s)	108.4 (d)[a]	7.33 (s)	-
6	-	143.9 (s)	-	H-5*, H-8 and H-1"
7	-	156.7 (s)	-	H-5, H-8* and MeO-7
8	6.54 (s)	100.2 (d)	6.50 (s)	-
9	-	157.9 (s)	-	H-5 and H-8*
10	-	113.1 (s)	-	H-5* and H-8
1'	-	132.7 (s)	-	H-3 and H-5'
2'	7.04 (br s)	106.7 (d)	6.99 (d, 2.0)	H-6'
3'	-	147.9 (s)[b]	-	H-2'*, H-5' and -OCH$_2$O-
4'	-	148.1 (s)[b]	-	H-2', H-6' and -OCH$_2$O-
5'	6.88 (d, 7.8)	108.3 (d)[a]	6.85 (d, 8.0)	-
6'	6.92 (br d, 7.8)	120.0 (d)	6.93 (dd, 8.0, 2.0)	H-2'
1"	4.61 (d, 6.6)	66.0 (t)	4.58 (br d, 7.0)	-
2"	5.56 (t, 6.6)	119.3 (d)	5.55 (br t, 7.0)	H-1"*
3"	-	138.3 (s)	-	H-1", H-4"* and H-5"*
4"	1.83 (s)	25.8 (q)	1.79 (s)	H-5"*
5"	1.80 (s)	18.2 (q)	1.76 (s)	H-4"*
MeO-7	3.93 (s)	56.2 (q)	3.89 (s)	-
-OCH$_2$O-	6.04 (s)	101.3 (t)	6.00 (s)	-

[a,b] Interchangable within the same column.

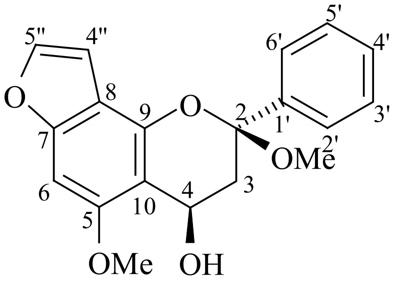
*Two-bond coupling.

1.29 Structure Determination of Compound ME27

Compound ME27 showed a molecular ion $[M^+]$ at 326.11934 in the HREIMS, corresponding to the molecular formula $C_{19}H_{18}O_5$ (calcd 326.11542). The IR spectrum (Figure 262) demonstrated the presence of a hydroxyl (3447 cm^{-1}) but not a carbonyl group. The UV maximal absorptions at 214 and 250 nm (Figure 261) were suggestive of a flavan skeleton (Gómez *et al*., 1985).

The presence of a one-proton multiplet at δ 5.00 (H-4) and two one-proton doublets of doublets at δ 2.18 (*J* = 4.5 and 14.8 Hz, H-3ax) and δ 2.79 (*J* = 1.3 and 14.8 Hz, H-3eq) in the ^1H NMR spectrum (Table 34 and Figure 265), together with the appearance of the quaternary carbon signal at δ 101.9 (C-2) in the HSQC spectrum (Figure 269) indicated that compound ME27 should be a flavan with oxygenation at C-2 and C-4.

Four substituents were attached to the flavan skeleton, as indicated by signals for two methoxyls at δ 3.97 (3H, *s*) and δ 3.13 (3H, *s*), for a furan ring at δ 7.52 (d, *J* = 2.0 Hz, H-5") and δ 6.91 (d, *J* = 2.0 Hz, H-4"), and for a hydroxyl group at δ 4.07 (br d, *J* = 9.8 Hz, exchangeable with D_2O) in the ^1H NMR spectrum. The presence of an AA'BB'C spin system at δ 7.72 (2H, m, H-2' and H-6'), δ 7.48 (2H, m, H-3' and H-5') and δ 7.42 (1H, m, H-4') indicated an unsubstituted B ring. The first methoxyl and the furan ring should be on ring A and the second methoxyl should be at C-2, as evident from the fragment ions at *m/z* 192 and 134 caused by RDA cleavage of ring C in the mass spectrum (Drewes, 1974). On ring A, a NOESY cross peak between the methoxyl at δ 3.97 and H-4, suggested the location of this methoxyl at C-5. This was confirmed by the 3-bond HMBC (Figure 270) correlation of C-5 (δ 157.4) with this methoxyl protons and with H-4. The location of the second methoxyl (δ 3.13) at C-2 was confirmed by its NOESY correlation peak with H-2'/H-6', together with the HMBC correlations of C-2 (δ 101.9) with the methoxyl protons, H-2'/H-6' and H-4. The furan ring was fused in an angular position at C-7 and C-8, as established by the NOESY interaction (Figure 268) of H-4" with H-2'/H-6' and MeO-2, and two-bond coupling of H-6 with C-5 and C-7 in the HMBC spectrum. The relative configuration of compound ME27 was established by the NOESY interaction between the OH and OMe groups, indicating their *cis*-orientation. Thus, structure of compound ME27 was established as a new flavan, 2,5-dimethoxy-4-hydroxy-[2",3':7,8]-furanoflavan [**458**], the first representative of flavan-4-ols with a methoxyl group at C-2 (Sritularak *et al*., 2002b).



[**458**]

Table 34 NMR Spectral data of compound ME27 ($CDCl_3$)

position	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	HMBC (correlation with ^{1}H)
2	-	101.9 (s)	H-4, H-2', H-6' and MeO-2
3	2.18 (dd, 14.8, 4.5)	42.4 (t)	-
	2.79 (dd, 14.8, 1.3)	-	-
4	5.00 (m)	59.7 (d)	H-3*
5	-	157.4 (d)	H-4, H-6* and MeO-5
6	6.81 (s)	88.8 (d)	-
7	-	156.7 (s)	H-6*, H-4" and H-5"
8	-	110.9 (s)	H-6 and H-5"
9	-	144.4 (s)	H-4 and H-4"
10	-	190.0 (s)	H-3 and H-6
1'	-	140.1 (s)	H-3' and H-5'
2'	7.72 (m)	126.4 (d)	H-4' and H-6'
3'	7.48 (m)	128.8 (d)	H-5'
4'	7.42 (m)	128.8 (d)	H-2' and H-6'
5'	7.48 (m)	128.8 (d)	H-3'
6'	7.72 (m)	126.4 (d)	H-2' and H-4'
4"	6.91 (d, 2.0)	103.8 (d)	H-5"*
5"	7.52 (d, 2.0)	143.1 (d)	H-4"*
MeO-2	3.13 (s)	50.7 (q)	-
MeO-5	3.97 (s)	56.4 (q)	-
HO-4	4.07 (s)	-	-

*Two-bond coupling.

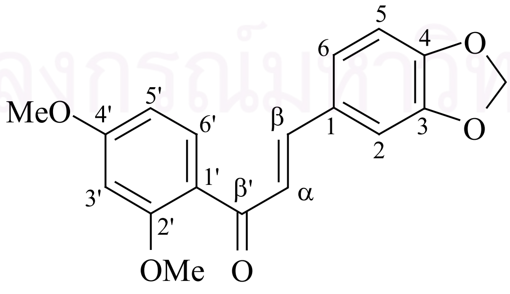
1.30 Structure Determination of Compound ME28

Compound ME28, showing [M$^+$] at *m/z* 312 in the EI mass spectrum (Figure 273), was obtained as a yellow powder. The IR bands at 1653 (C=O) and 1602 (C=C) cm^{-1} (Figure 272) and the UV absorption at 351 nm (Figure 271) were suggestive of a chalcone skeleton (Markham, 1982).

The ^{1}H NMR signals for a set of *trans*-olefinic protons at δ 7.36 and δ 7.24 (each d, *J* = 15.9 Hz) confirmed the existence of the chalcone nucleus (Table 35 and Figure 274). The ^{13}C NMR (Table 35 and Figure 275) and HSQC spectra (Figure 278) showed 19 signals, corresponding to two methoxyls, one methylene, eight methines and seven quaternary carbons. Three substituents were attached to the chalcone nucleus, as indicated by signals for two methoxyls at δ 3.91 and δ 3.94 (each 3H, *s*) and for a methylenedioxy at δ 6.05 (2H, *s*) in the ^{1}H NMR spectrum.

The two methoxyls should be located on ring A and the methylenedioxyl on ring B, as shown by the fragment ions at *m/z* 147 and 165 in the EIMS (Drewes, 1974). On ring A, an ABM splitting system consisting of two doublets at δ 6.53 (*J* = 1.8 Hz, H-3') and δ 7.78 (*J* = 8.7 Hz, H-6') and a broad doublet at δ 6.59 (*J* = 8.7, H-5'), together with the HMBC correlation (Figure 279) of H-6' with C-β' (δ 141.9) suggested the location of the two methoxyls at C-2' and C-4' positions. This was confirmed by NOESY interactions (Figure 277) of MeO-2' (δ 3.94, *s*) with H-3' and MeO-4' (δ 3.91, *s*) with H-3' and H-5', respectively. On ring B, the ^{1}H NMR ABM spin system at δ 7.16 (1H, br *s*, H-2), 7.10 (1H, br d, *J* = 8.1 Hz, H-6) and 6.85 (1H, *d*, *J* = 8.1 Hz, H-5), together with the HMBC correlation of C-β (δ 141.9) with H-2 and H-6, indicated the placement of the methylenedioxyl at C-3 and C-4.

Thus, compound ME28 was identified as 3,4-methylenedioxy-2',4'-dimethoxychalcone [**459**]. Although this compound has been earlier synthesized (Salem *et al.*, 2000), this is the first time it has been found as a naturally occurring compound. Prior to this study, the ^{13}C NMR data of this compound have not been reported (Sritularak *et al.*, 2002b).



[**459**]

Table 35 NMR Spectral data of compound ME28 and 3,4-methylenedioxy-2',4'-dimethoxy-chalcone ($CDCl_3$)

position	Compound ME28		3,4-Methylenedioxy-2',4'-dimethoxy-chalcone	HMBC
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	(correlation with ^{1}H)
1	-	129.9 (s)	-	H-α and H-5
2	7.16 (br s)	106.6 (d)	6.60 (d, 3.1)	H-β and H-6
3	-	148.2 (s)		H-5 and -OCH_2O-
4	-	149.4 (s)		H-2, H-6 and -OCH_2O-
5	6.85 (d, 8.1)	108.6 (d)	7.30-7.70 (m)	-
6	7.10 (br d, 8.1)	124.8 (d)	7.30-7.70 (m)	H-β and H-2
1'	-	122.4 (s)	-	H-α, H-3' and H-5'
2'	-	160.3 (s)	-	H-6' and MeO-2'
3'	6.53 (d, 1.8)	98.7 (d)	6.50 (d, 3.1)	H-5'
4'	-	164.1 (s)	-	H-3'*, H-6' and MeO-4'
5'	6.59 (br d, 8.7)	105.1 (d)	7.30-7.70 (m)	H-3'
6'	7.78 (d, 8.7)	132.8 (d)	7.80 (d, 9.1)	-
α	7.39 (d, 15.9)	125.4 (d)	7.30-7.70 (m)	-
β	7.64 (d, 15.9)	141.9 (d)	7.30-7.70 (m)	H-α*, H-2 and H-6
β'	-	190.4 (s)	-	H-α* and H-β
MeO-2'	3.94 (s)	55.7 (q)	3.90 (s)	-
MeO-4'	3.92 (s)	55.5 (q)	3.80 (s)	-
-OCH_2O-	6.05 (s)	101.5 (t)	5.90 (s)	-

*Two-bond coupling.

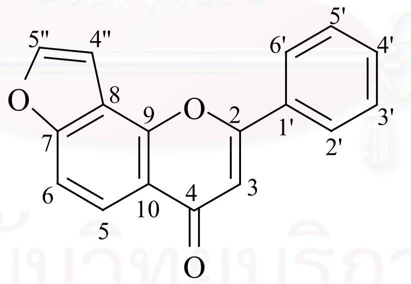
1.31 Structure Determination of Compound ME29

Compound ME29, colorless needles, showed a molecular [M^+] ion at *m/z* 262, analyzed for $C_{17}H_{10}O_3$. The UV absorptions at 215, 263 and 297 nm (Figure 280) and the IR bands at 1645 (conjugated carbonyl), 1605 (conjugated C=C) and 1215 and 1116 (ether) cm^{-1} (Figure 281) were characteristics of a flavone skeleton (Markham, 1982).

This was confirmed by the presence of a sharp singlet proton signal at δ 6.94 of H-3 in the ^1H NMR spectrum (Table 36 and Figure 283). The ^1H NMR spectrum showed, in addition to the signals for a furan ring at δ 7.28 and δ 7.83 (each d, *J* = 2.1 Hz, H-4" and H-5"), two doublet signals with *ortho*-aromatic coupling at δ 8.23 (d, *J* = 9.0 Hz, H-5) and δ 7.62 (d, *J* = 9.0 Hz, H-6) and signals for an unsubstituted B ring at δ 8.02 (2H, m, H-2' and H-6') and δ 7.62 (3H, m, H-3', H-4' and H-5').

The ^{13}C NMR spectrum (Table 36 and Figure 284) showed 17 carbon signals, representing ten methines and seven quaternary carbons. In the EIMS, the fragment ion at *m/z* 160 resulting from RDA cleavage confirmed the location of a furan ring on ring A (Drewes, 1974).

By analysis of ^1H and ^{13}C NMR spectral data and comparison with previously reported data (Tanaka *et al.*, 1992; Mbafor *et al.*, 1995), compound ME29 was identified as lanceolatin B [**278**], a flavone previously found in *Lonchocarpus latifolius* (Magalhães *et al.*, 2000) and *Pongamia glabra* (Malik, Sharma and Seshadri, 1977).



[**278**]

Table 36 NMR Spectral data of compound ME29 and lanceolatin B ($CDCl_3$)

position	Compound ME29		Lanceolatin B	
	^{1}H (mult., *J* in Hz)	^{13}C (mult.)	^{1}H (mult., *J* in Hz)	^{13}C
2	-	162.6 (s)	-	162.7
3	6.94 (s)	108.1 (d)	6.90 (s)	108.1
4	-	178.2 (s)	-	178.2
5	8.23 (d, 9.0)	121.8 (d)	8.18 (d, 9.0)	121.8
6	7.62 (d, 9.0)	110.2 (d)	7.58 (d, 9.0)	110.2
7	-	158.3 (s)	-	158.4
8	-	117.1 (s)	-	117.2
9	-	150.8 (s)	-	150.9
10	-	119.4 (s)	-	119.4
1'	-	131.8 (s)	-	131.8
2'	8.02 (m)	126.2 (d)	7.98 (m)	126.2
3'	7.62 (m)	129.1 (d)	7.58 (m)	129.1
4'	7.62 (m)	131.5 (d)	7.58 (m)	131.5
5'	7.62 (m)	129.1 (d)	7.58 (m)	129.1
6'	8.02 (m)	126.2 (d)	7.98 (m)	126.2
4"	7.28 (d, 2.1)	104.2 (d)	7.26 (d, 2.0)	104.2
5"	7.83 (d, 2.1)	145.7 (d)	7.79 (d, 2.0)	145.8

2. Tyrosinase Inhibitory Activity

Tyrosinase is a copper monooxygenase enzyme widely distributed in nature. It has been found in plants, fungi, insects and animals. A number of physiological functions of this enzyme have been studied (Gelder *et al*., 1997). Tyrosinase is one of the important key enzymes involved in the molting process of insects (Kubo *et al*., 1995). A search for its inhibitors may therefore lead to the discovery of insect control agents. In plants, tyrosinase has been found to be responsible for browning in plants, especially in fruits and vegetables. (Gelder *et al*., 1997). In mammals and humans, the function of tyrosinase in the biosynthesis of the skin pigment melanin is well-established (Gelder *et al*., 1997). The biosynthesis of melanin has been studied intensively by Raper (Britton, 1983) and subsequently by Mason (Britton, 1983) which led to the proposal of Raper-Mason scheme of melanogenesis. Thus, the study of tyrosinase inhibitors should be useful for the treatment of localized hyperpigmentation in human such as nevus, lentigo, post-inflammatory state, ephelis and melanoma of pregnancy. Moreover, tyrosinase inhibitors are becoming more important for the development of cosmetic products. (Kubo *et al*., 1995).

In this study, the tyrosinase inhibitory activity of each pure compound and each crude extract was determined by the dopachrome method. It was modified from the procedures describe by Masamoto (Masamoto *et al*., 1980), Iida (Iida *et al*., 1995) and Morita (Morata *et al*., 1994). The MeOH extract of *A. gomezianus* showed 82.5% of tyrosinase inhibition whereas the extracts of *M. erythrocalyx* were devoid of activity (crude extract 30 mg in EtOH 10 mL). The activities of pure compounds were expressed as IC_{50} values (concentration of 50% inhibition) in comparison with kojic acid, a well-known inhibitor of tyrosinase.

Artogomezianol [**441**] and andalasin A [**442**] showed moderate tyrosinase inhibition with IC_{50} values 68 and 39 μM, respectively (kojic acid, IC_{50} 27 μM). As expected, andalasin A [**442**], having two 4-substituted resorcinol moieties (ring A and C), was nearly two times as inhibitory as artogomezianol [**441**], which possesses only one 4-substituted resorcinol structure (ring C). The relationship of 4-substituted resorcinol skeleton and tyrosinase inhibitory activity have been extensively discussed (Shimizu, Kondo and Sakai, 2000).

3. Anti-Herpes Simplex Activity

Herpes simplex viruses (HSV) are extremely common human pathogens, which cause a broad spectrum of illness, ranging from asymptomatic infections to fulminant, disseminate disease resulting in death. There are two types of HSV, type 1 and type 2. The two types vary in biochemical composition, have different biologic properties, and can be readily distinguished from one another by a variety of immunologic techniques. In general, HSV type 1 (HSV-1) is responsible for orofacial infections, visceral infections in immunocompromised hosts, and herpes simplex encephalitis in adults. HSV type 2 (HSV-2) is more commonly associated with infections of the genital tract, and it causes the majority of neonatal disease. Despite these generalizations, however, there exists considerable overlap in the spectrum of clinical disease by these two closely related agents. (Belshe, 1991).

In this study, evaluations of anti-herpes simplex activity of pure compounds and crude extracts were performed using the plaque reduction assay (inactivation) (Lipipun *et al*., 2000; Abou-karam and Shier, 1990). The MeOH extract from the roots of *A. gomezianus* at 20 μg/mL showed 90% and 92% inhibition for HSV-1 and HSV-2, respectively. At the same concentration, the EtOAc extract from the stem bark of *M. erythrocalyx* showed 75% (HSV-1) and 50% (HSV-2) virus inhibition and the hexane extract from the roots showed 70% (HSV-1) and 40% (HSV-2) virus inhibition.

Pure compounds from *M. erythrocalyx* and *A. gomezianus* were tested for anti-HSV activity at ≤ 50 μg/mL. Compounds exhibiting more than 50 % inhibition without cytotoxicity at 50 μg/mL were further evaluated for ED_{50}. Acyclovir was used as positive control and the cytotoxicity of normal cell was also evaluated. The results are summarized in Table 37.

Table 37 Percentage of virus inhibition by pure compounds isolated from *A. gomezianus* and *M. erythrocalyx*

Compounds	Conc. (μg/mL)	% Inhibition		ED_{50} [a] (μM)		Cytotoxicity [b]	CD_{50} [c] (μM)	Selectivity [d] Index	
		HSV-1	HSV-2	HSV-1	HSV-2			HSV-1	HSV-2
Artocarpin [**4**]	10	0	0			-			
	50					+ + +			
Cycloartocarpin [**30**]	5	0	30			-			
	50					+ + +			
Isocyclomorusin [**25**]	6.25	0	0	22.5	22.5	-	41.6	1.8	1.8
	12.5	100	100			-			
	50					+ + +			
Norartocarpetin [**45**]	20	40	50			-			
	50					+			
Cudraflavone C [**69**]	5	20	0			-			
	50					+ + +			

Table 37 (continued)

Compounds	Conc. (μg/mL)	% Inhibition		ED$_{50}$[a] (μM)		Cytotoxicity[b]	CD$_{50}$[c] (μM)	Selectivity[d] Index	
		HSV-1	HSV-2	HSV-1	HSV-2			HSV-1	HSV-2
Artogomezianol [**441**]	50	0	0			-			
Andalasin A [**442**]	12.5	20	75	35.2	3.3	-	102.4	2.9	31.0
	25	100	100			-			
	50					+ +			
Derricidin [**443**]	50	0	0			-			
7-γ, γ -Dimethylallylloxy flavanone [**444**]	50	0	0			-			
2'-Hydroxy-3,4-methylenedioxy-4'-γ, γ -dimethylallyloxy chalcone [**445**]	50	40	0			-			
Lupeol [**169**]	50	20	0			-			
Ponganone I [**446**]	20	0	0			-			
	50					+ +			
Milletenone [**280**]	50	20	0			-			
Ovalifolin [**308**]	25	35	16.7	97.7	108.7	-	289.0	2.9	2.6
	50	77.5	83.3			-			
	100					+ +			
Pongol methyl ether [**447**]	5	0	0	70.5	132.8	-	256.8	3.6	1.9
	10	25	0			-			
	25	56.9	7.1			-			
	50	75.8	88.1			-			
	100					+ + +			
Millettocalyxin B [**448**]	50	25	20			-			
Milletenin C [**298**]	50	25	45			-			
Millettocalyxin C [**449**]	50	45	25			-			
Millettocalyxin A [**450**]	6.25	0	0	53.6	57.3	-	230.1	4.2	4.0
	12.5	19.4	13.0			-			
	25	100	91.7			-			
	50					-			
	100					+ + +			
3',4'-Methylenedioxy- 7-methoxyflavone [**279**]	50	0	0			-			
Pongaglabrone [**314**]	50	60	0			-			

Table 37 (continued)

Compounds	Conc. (μg/mL)	% Inhibition		ED_{50}[a] (μM)		Cytotoxicity[b]	CD_{50}[c] (μM)	Selectivity[d] Index	
		HSV-1	HSV-2	HSV-1	HSV-2			HSV-1	HSV-2
Prunetin[**451**]	6.25	0	0	33.8	33.6	-	176.0	5.2	5.2
	12.5	85	75			-			
	25					-			
	50					+ +			
Vicenin II [**452**]	50	0	0			-			
1-(4-Hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one [**289**]	50	50	0			-			
Pongaglabol [**337**]	50	45	50			-			
Pongamol [**315**]	20	40	30			-			
	50					+ + +			
Ovalitenone [**312**]	50	0	20			-			
(-)-(2*S*)-6-Methoxy-[2",3":7,8]-furano flavanone [**456**]	50	40	0			-			
Ponganone V [**457**]	50	40	25			-			
2,5-Dimethoxy-4-hydroxy-[2",3":7,8]-furanoflavan [**458**]	10	20	NI			-			
	50					+ + +			
3,4-Methylenedioxy-2',4'-dimethoxy-chalcone [**459**]	20	30	16			-			
	50					+			
Lanceolatin B [**278**]	50	40	35			-			
Acyclovir				0.25	2.24	-			

[a] Cytotoxicity (- Non toxic, + < 30%, + + 30-80%, + + + > 80%)

[b] ED_{50} = 50% Effective Dose (weak: ED_{50} > 300 folds greater than acyclovir, moderate: ED_{50} 10-300 folds greater than acyclovir, strong: < 10 folds greater than acyclovir)

[c] CD_{50} = 50% Cytotoxic Dose (Examined by microscopic observation)

[d] Selectivity Index = CD_{50}/ ED_{50}

Seven compounds from *A. gomezianus* were evaluated for anti-herpes simplex activity. Artocarpin [**4**], cycloartocarpin [**30**], norartocarpetin [**45**], cudraflavone C [**69**] and artogomezianol [**441**] showed weak activity against both types of virus whereas isocyclomorusin [**25**] showed moderate activity. Andalasin A [**442**] showed strong activity against HSV-2 but moderate activity against HSV-1. For compounds from *M. erythrocalyx*, 25 compounds were evaluated for anti-HSV activity. Almost all of them showed weak anti-HSV except for 4 flavonoids, including ovalifolin [**308**], pongol methyl ether [**447**], millettocalyxin A [**450**] and prunetin [**451**], which showed moderate activity against both types of herpes simplex virus as compared with acyclovir.

4. Free Radical Scavenging Activity

Oxygen is present in the atmosphere as a stable triplet biradical ($^3O_2$) in the ground state and a vital component for the survival of the human. Once inhaled, it undergoes a gradual reduction process and ultimately gets metabolized into water. In this process, a small amount of reactive intermediates, such as superoxide anion radicals ($O_2^{\bullet\,-}$), hydroxyl radicals ($OH^{\bullet}$), nonfree radical species (such as H_2O_2), and the single oxygen ($^1O_2$) are formed. Those reactive intermediates are collectively termed as reactive oxygen species (ROS). These primary derivatives of oxygen play an important role in mediating ROS-related effects. ROS can easily initiate the peroxidation of membrane lipids, leading to the accumulation of lipid peroxides. The peroxidation products by themselves and their secondary oxidation products, such as malondialdehyde (MDA) and 4-hidroxinonenal (4-HNE) are highly reactive; they react with biological substrates, such as protein, amines, and deoxyribonucleic acid (DNA) (Gülçin *et al*., 2002).

In living organisms various ROS can be formed by different ways. In normal aerobic respiration, stimulated polymorphonuclear leucocytes and macrophages, and peroxisomes appear to be the main endogenous sources of most of the oxidants produced by cells. Exogenous sources of free radicals include tobacco smoke, ionizing radiation, certain pollutants, organic solvents and pesticides. Most living species have efficient defense systems to protect themselves against the oxidative stress induced by ROS. Recent investigations have shown that the antioxidant properties of plants could be correlated with oxidative stress defense and different human diseases including cancer, atherosclerosis, and the aging processes (Gülçin *et al*., 2002).

Antioxidants can interfere with the oxidation process by reacting with free radicals, chelating free catalytic metals and also by acting as oxygen scavengers. Phenolic antioxidants functions are free radical terminators and sometimes also metal chelators. Thus, antioxidant defense systems have coevolved with aerobic metabolism to counteract oxidative damage from ROS. The antioxidants may be used to preserve food quality from oxidative deterioration of lipid. Therefore, antioxidants play a very important role in the food industry. Synthetic antioxidants, such as butylated hydroxyanisole (BHA), butylated hydroxytotoluene (BHT), and *tert*-butylhydroquinone (TBHQ) are widely used in the food industry, but BHA and BHT have been suspected of being responsible for liver damage and carcinogenesis. Therefore, the development and utilization of more effective antioxidants of natural origin are desired (Gülçin *et al*., 2002).

By TLC screening assay, the MeOH extract from the roots of *A. gomezianus*, the EtOAc extract from the stem bark of *M. erythrocalyx*, and the hexane extract from the roots of *M. erythrocalyx* showed free radical scavenging activity.

Pure compounds from *M. erythrocalyx* and *A. gomezianus* were first tested at 3.3 x 10^{-4} μg/mL. Compounds exhibiting more than 50% inhibition were further analyzed for their IC_{50} values. Quercetin was used as positive control. The results are summarized in Table 38.

Table 38 Percentage of free radical scavenging activity by pure compounds isolated from *A. gomezianus* and *M. erythrocalyx*

Compounds	% Scavenging activity at 3.3 x 10^{-4} μg/mL^{a}	IC_{50} (μM)
Artocarpin [**4**]	26.3	-
Cycloartocarpin [**30**]	18.5	-
Isocyclomorusin [**25**]	15.0	-
Norartocarpetin [**45**]	21.0	-
Cudraflavone C [**69**]	21.3	-
Artogomezianol [**441**]	75.8	127.8
Andalasin A [**442**]	76.5	25.3
Derricidin [**443**]	16.3	-
Lupeol [**169**]	8.9	-
Ponganone I [**446**]	19.1	-
Milletenone [**280**]	9.2	-

Table 38 (continued)

Compounds	% Scavenging activity at 3.3×10^{-4} μg/mL[a]	IC_{50} (μM)
Ovalifolin [**308**]	15.8	-
Pongol methyl ether [**447**]	10.7	-
Millettocalyxin B [**448**]	7.3	-
3',4'-Methylenedioxy-6,7-dimethoxyflavone [**298**]	17.3	-
Millettocalyxin C [**449**]	16.1	-
3',4'-Methylenedioxy-7-dimethoxyflavone [**279**]	20.5	-
Prunetin [**451**]	12.9	-
Vicenin II [**452**]	18.2	-
Pongaglabol [**337**]	11.1	-
Pongamol [**315**]	10.6	-
Ovalitenone [**312**]	7.9	-
(-)-(2*S*)-6-Methoxy-[2",3":7,8]-furanoflavanone [**456**]	15.2	-
Ponganone V [**457**]	17.1	-
2,5-Dimethoxy-4-hydroxy-[2",3":7,8]-furanoflavan [**458**]	5.0	-
Lanceolatin B [**278**]	9.2	-
Quercetin[b] [**363**]	85.9	1.7

[a] Compound with > 50% inhibition were further analyzed for IC_{50} values.

[b] Concentration 2.0×10^{-4} μM/mL

From Table 38, only two compounds (artogomezianol [**441**] and andalasin A [**442**]) showed free radical scavenging activity. The structures of these compounds were composed of free hydroxyl substituents. This functional group should therefore be important for the activity. The connecting positions of the stilbene monomers might be significant for the activity, as supported by andalasin A [**442**] (IC_{50} 25.3 μM) showed potency 5 folds greater than artogomezianol [**441**] (IC_{50} 127.8 μM). However, these two compounds were only moderate free radical scavengers as compared with quercetin [**363**] (IC_{50} 1.7 μM). It should be noted that no flavonoids with free radical scavenging activity lack free hydroxyl groups.

CHAPTER V

CONCLUSION

In this investigation, from the roots of *Artocarpus gomezianus* Wall. Ex Tréc. a new dimeric stilbene, namely, artogomezianol [**441**] was isolated along with the known stilbene dimer andalasin A [**442**]. Chemical examination of the stem bark of *Millettia erythrocalyx* Gagnep. led to the isolation of three new compounds, namely, millettocalyxins A-C [**450**, **448** and **449**], and two new natural products pongol methyl ether [**447**] and 2'-hydroxy-3,4-methylenedioxy-4'-γ, γ-dimethylallyloxychalcone [**445**], along with 14 other known compounds. These known compounds are derricidin [**443**], 7-γ, γ-dimethylallyloxyflavanone [**444**], ponganone I [**446**], karanjin [**295**], milletenone [**280**], ovalifolin [**308**], milletenin C [**298**], 3',4'-methylenedioxy-7-methoxyflavone [**279**], pongaglabrone [**314**], prunetin [**451**], vicenin II [**452**], isovitexin [**454**], lupeol [**169**], and dihydrophaseic acid-4'-*O*-β-D-glucopyranoside [**453**]. From the roots of *M. erythrocalyx* Gagnep., two new compounds, 6-methoxy-[2",3":7,8]-furanoflavanone [**456**] and 2,5-dimethoxy-4-hydroxy-[2",3":7,8]-furanoflavan [**458**], and the new natural product 3,4-methylenedioxy-2',4'-dimethoxychalcone [**459**] were isolated, together with 10 other known flavonoids, i.e. 1-(4-hydroxy-5-benzofuranyl)-3-phenyl-2-propen-1-one [**289**], derricidin [**443**], purpurenone [**455**], pongaglabol [**337**], ponganone I [**446**], pongamol [**315**], ovalitenone [**316**], milletenone [**279**], ponganone V [**457**] and lanceolatin B [**278**]. Artogomezianol [**441**] and andalasin A [**442**] from *A. gomezianus* showed moderate tyrosinase inhibitory effects and appreciable free radical scavenging activities. In addition, andalasin A [**422**] showed strong activity against HSV-2 but moderate activity against HSV-1. All of the compounds from *M. erythrocalyx* showed no tyrosinase inhibitory activity. Almost all of them showed weak free radical scavenging activity and weak activity against herpes simplex viruses except for 4 flavonoids, including ovalifolin [**308**], pongol methyl ether [**447**], millettocalyxin A [**450**] and prunetin [**451**], which showed moderate activity against both types of virus.

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APPENDIX

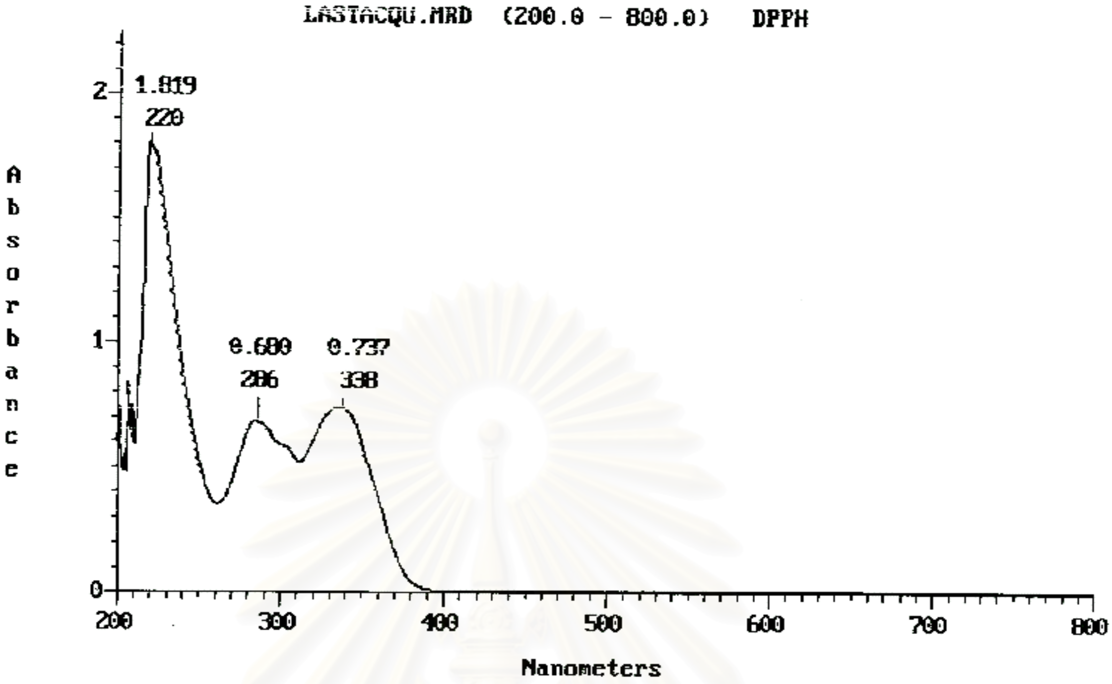

Figure 10 UV Spectrum of compound AG11 (methanol)

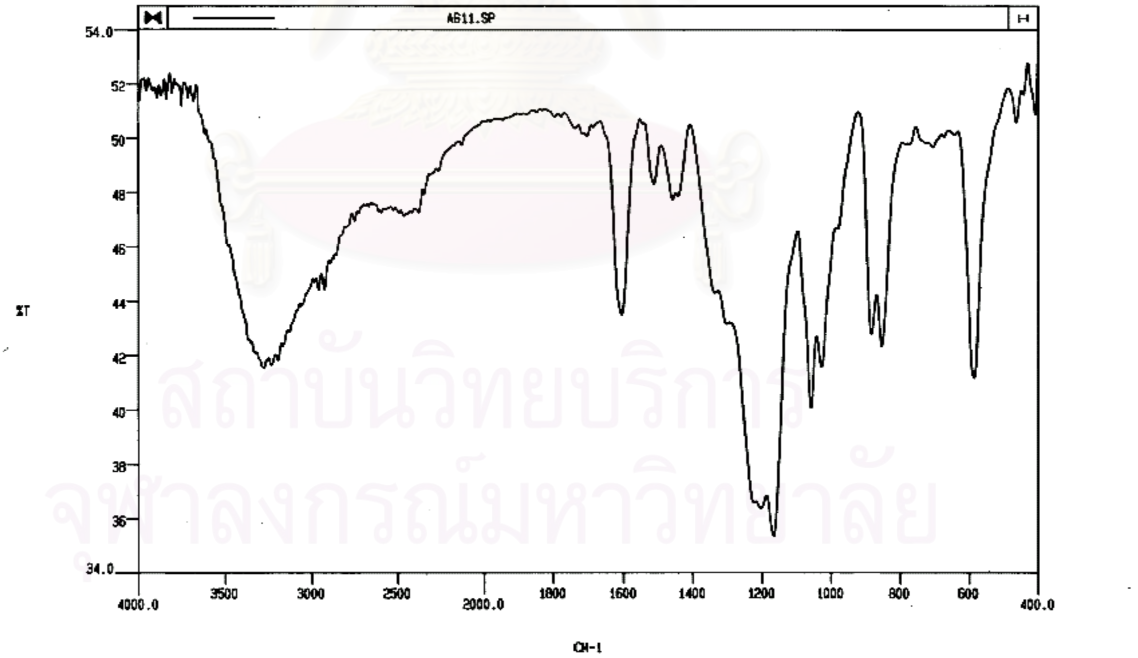

Figure 11 IR Spectrum of compound AG11 (KBr disc)

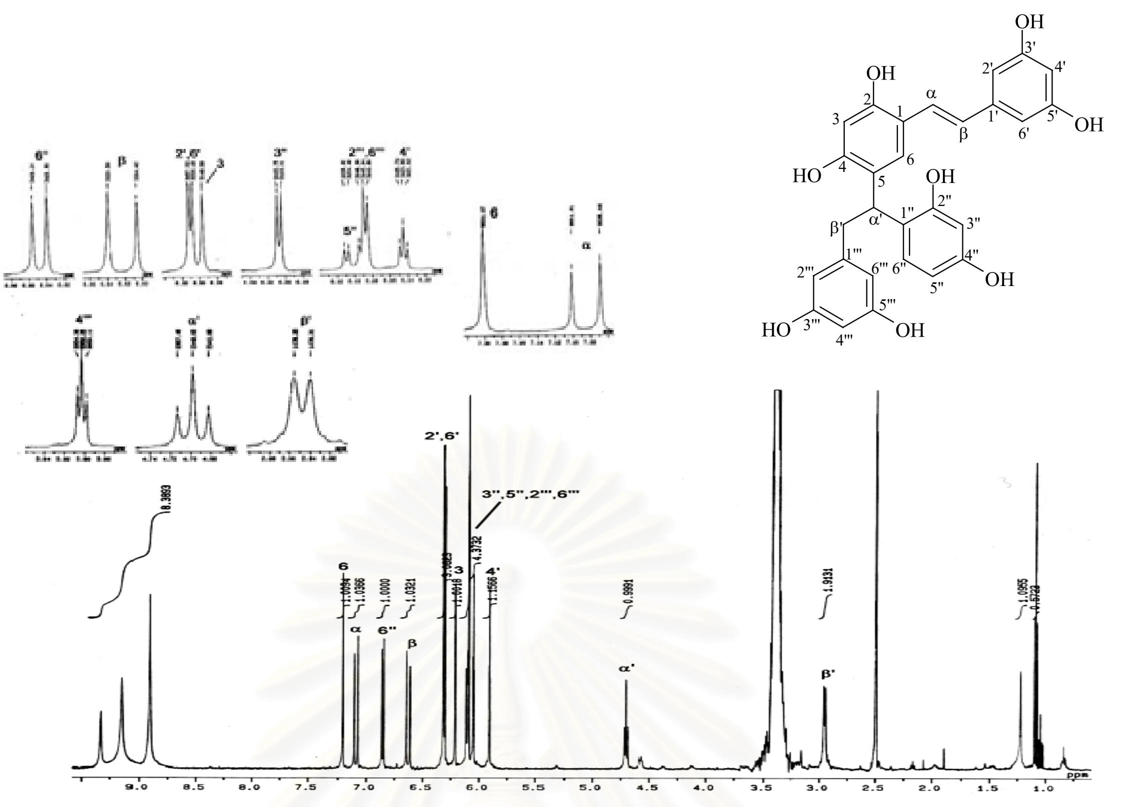

Figure 12 ^{1}H NMR (500 MHz) Spectrum of compound AG11 (DMSO-d_6)

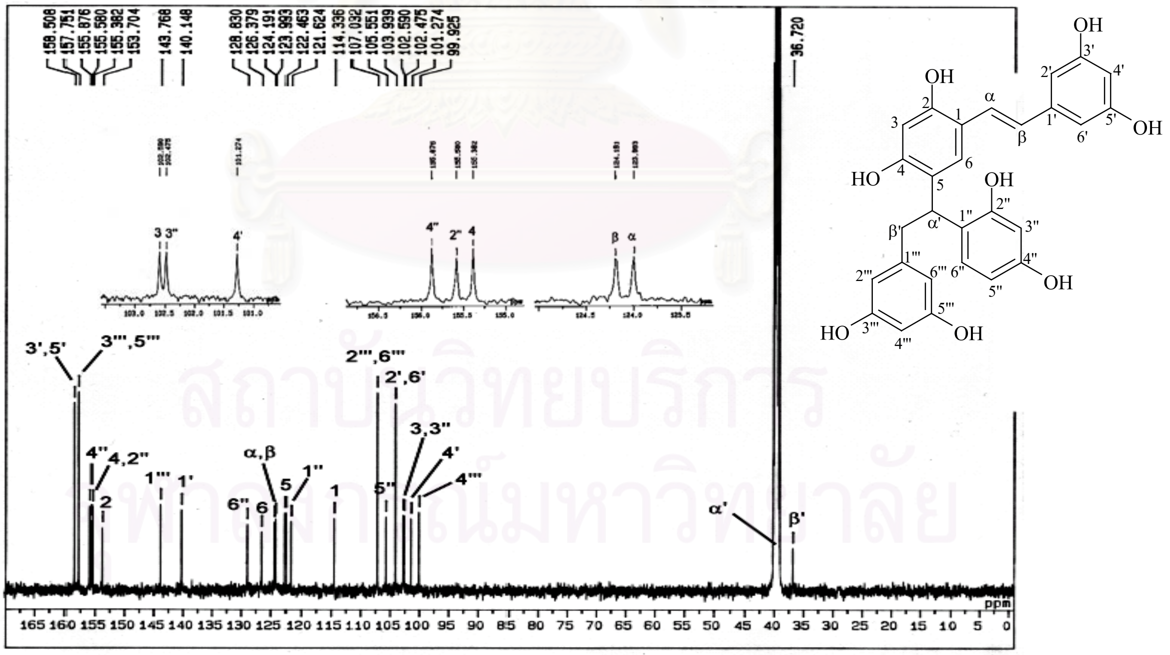

Figure 13 ^{13}C NMR (125 MHz) Spectrum of compound AG11 (DMSO-d_6)

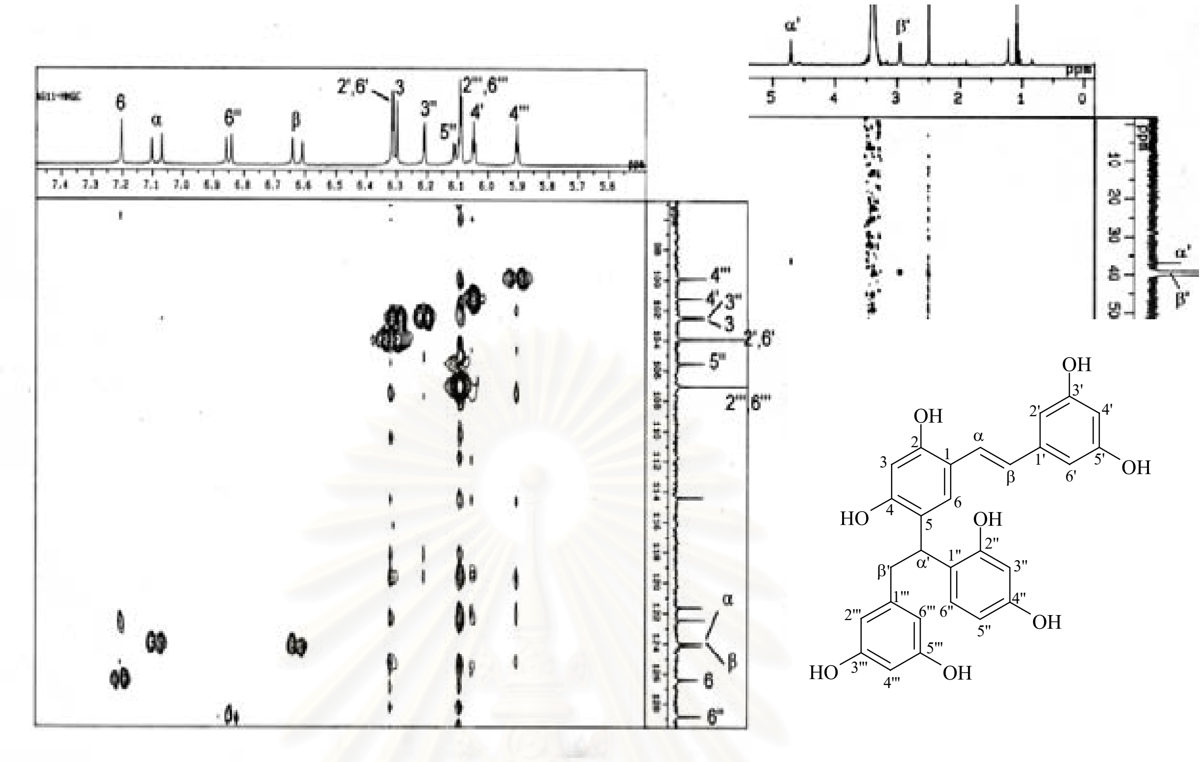

Figure 14 HMQC Spectrum of compound AG11 (DMSO-d_6)

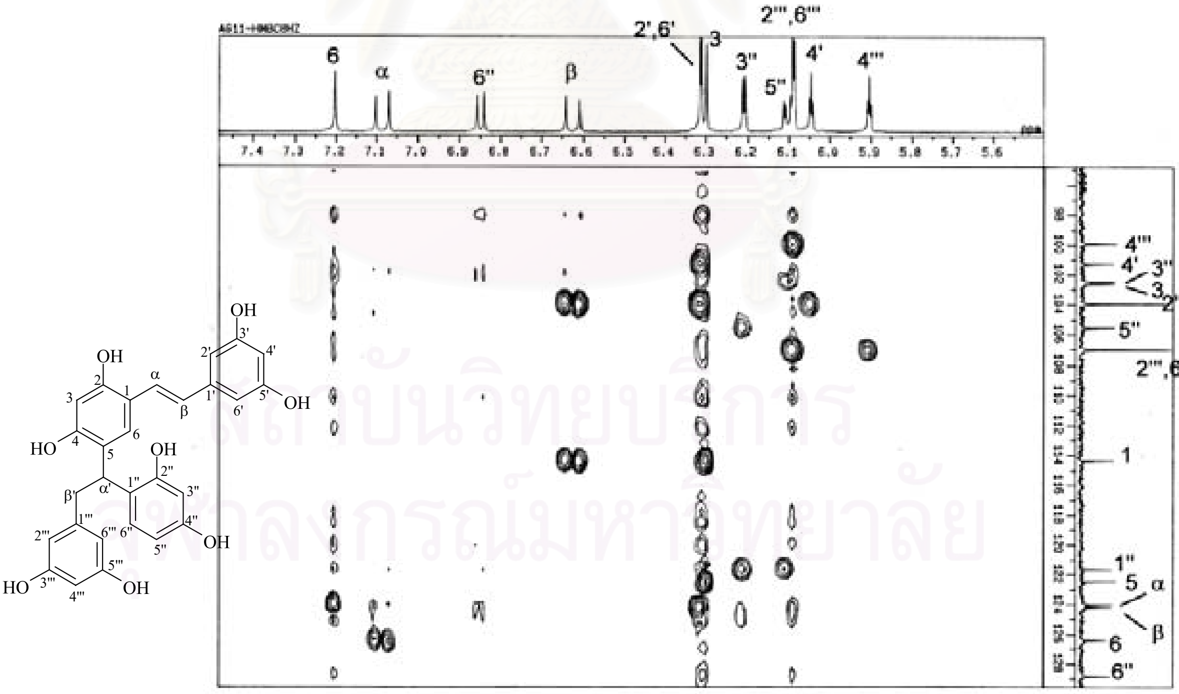

Figure 15 HMBC Spectrum of compound AG11 (DMSO-d_6) [δ_H 5.5-7.5 ppm, δ_C 96-129 ppm]

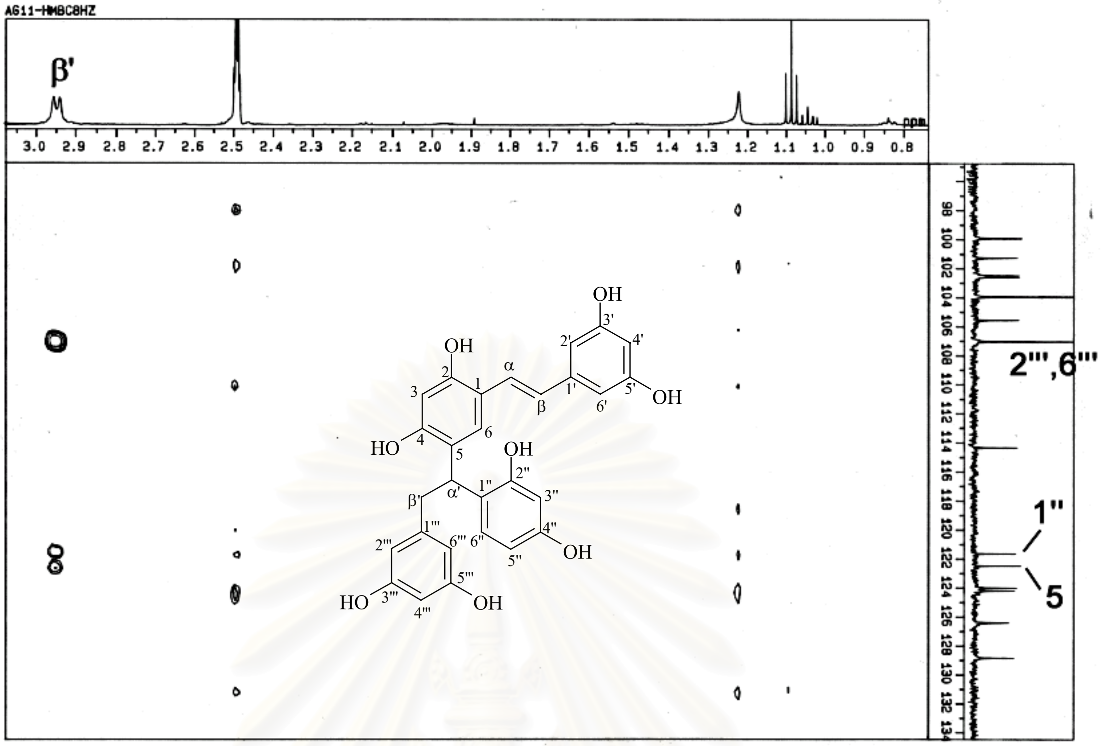

Figure 16 HMBC Spectrum of compound AG11 (DMSO-d_6) [δ_H 5.5-7.5 ppm, δ_C 136-164 ppm]

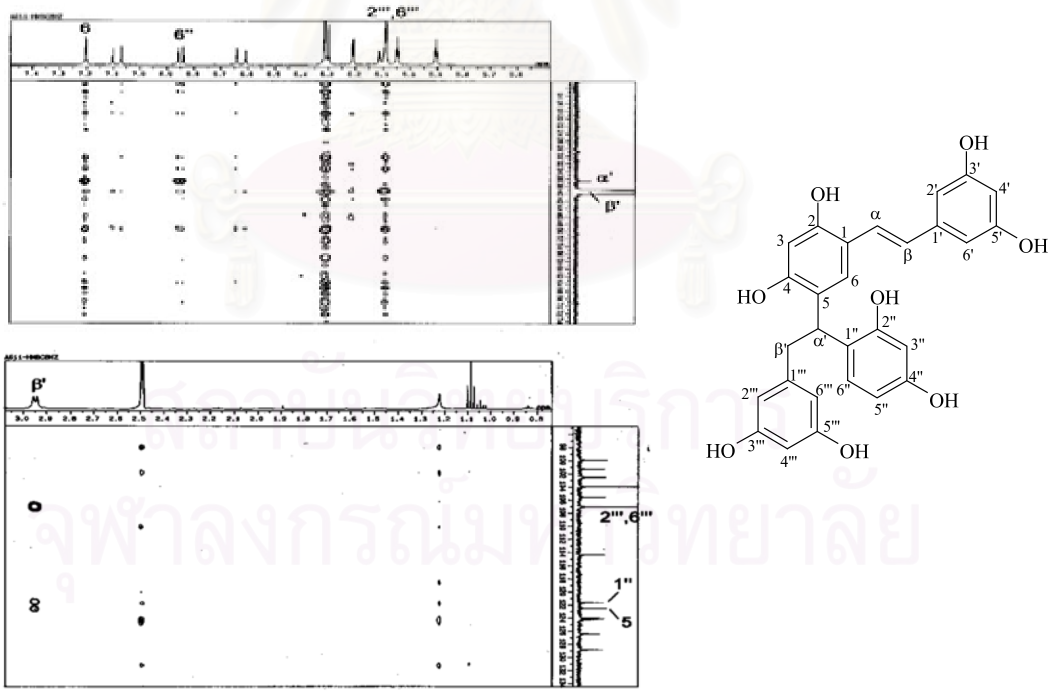

Figure 17 HMBC Spectrum of compound AG11(DMSO-d_6) [δ_H 5.5-7.5 ppm, δ_C 12-74 ppm; δ_H 0.8-3.1 ppm, δ_C 98-134 ppm]

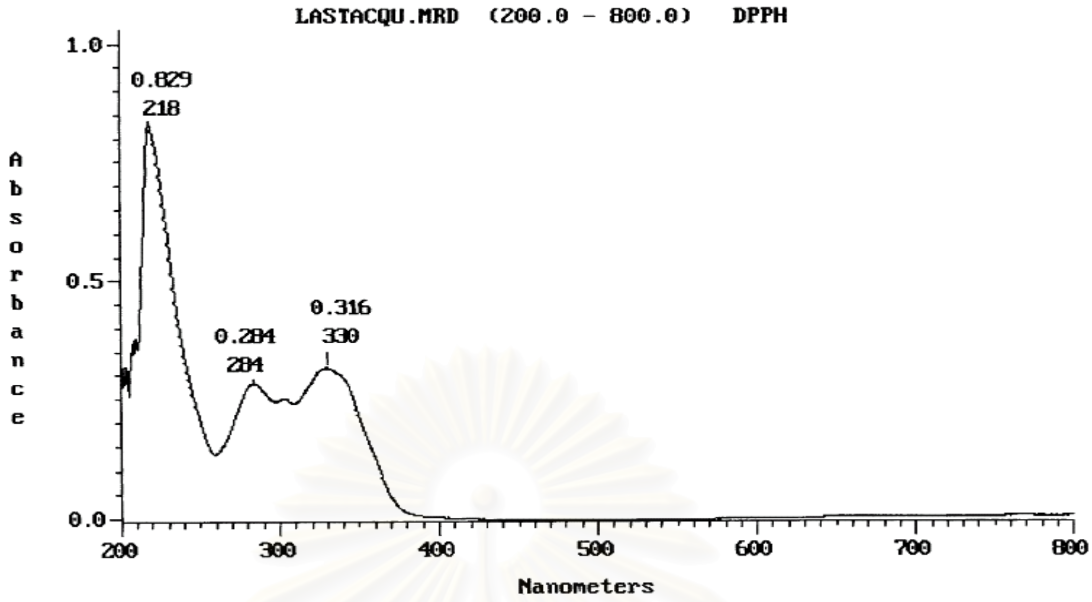

Figure 18 UV Spectrum of compound AG12 (methanol)

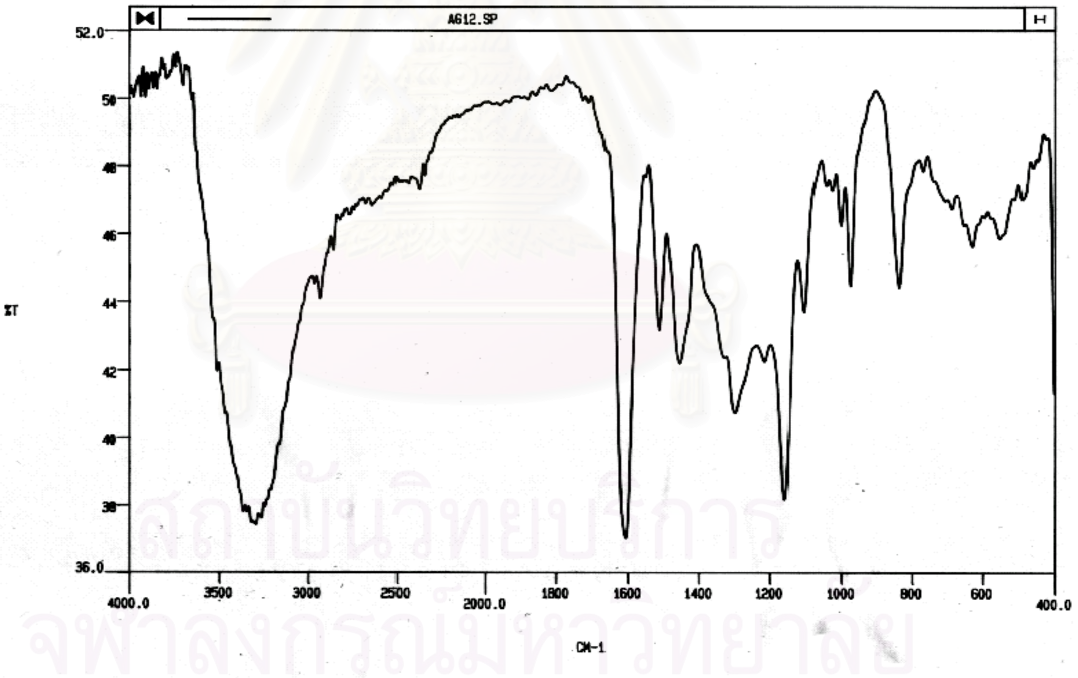

Figure 19 IR Spectrum of compound AG12 (KBr disc)

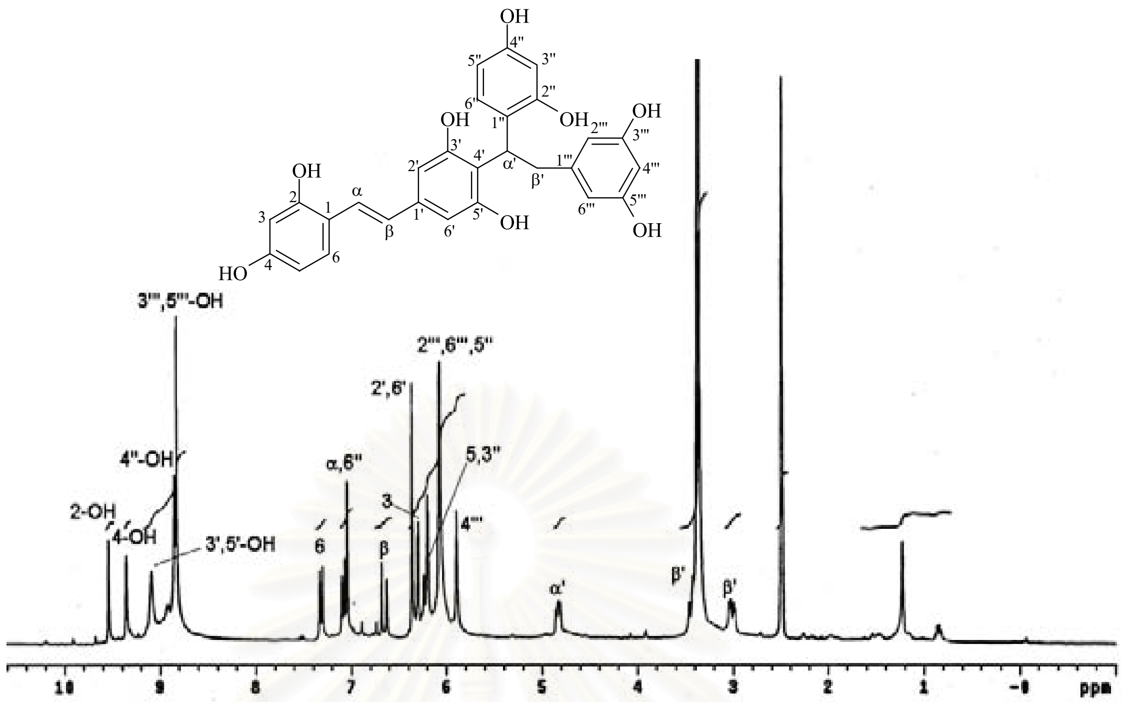

Figure 20 ^{1}H NMR (500 MHz) Spectrum of compound AG12 (DMSO-d_6)

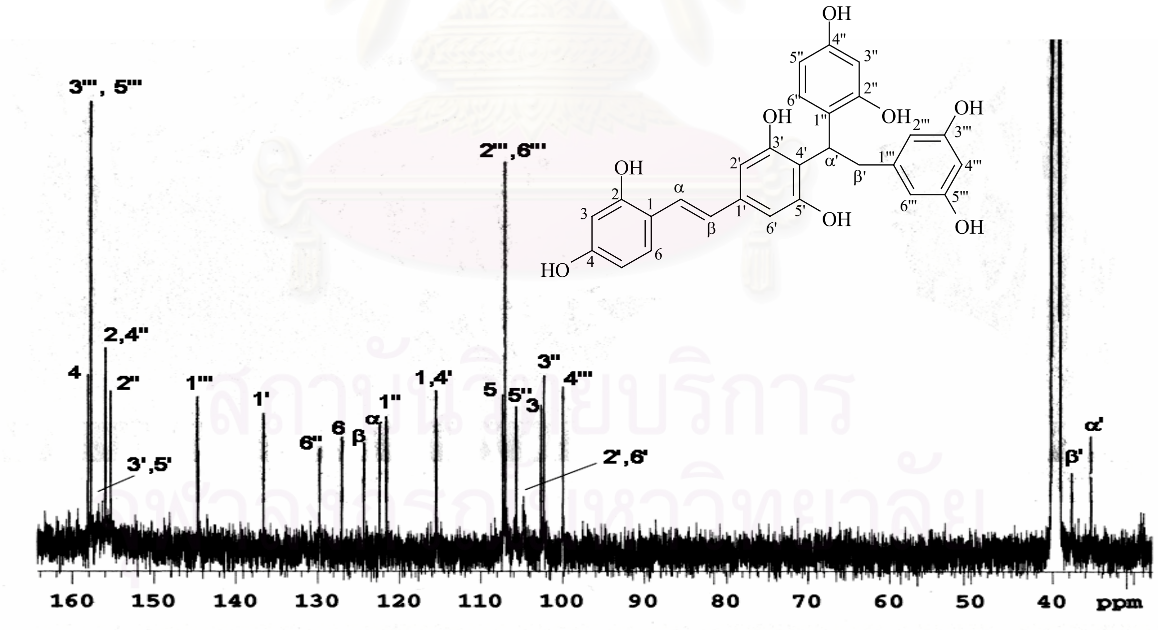

Figure 21 ^{13}C NMR (125 MHz) Spectrum of compound AG12 (DMSO-d_6)

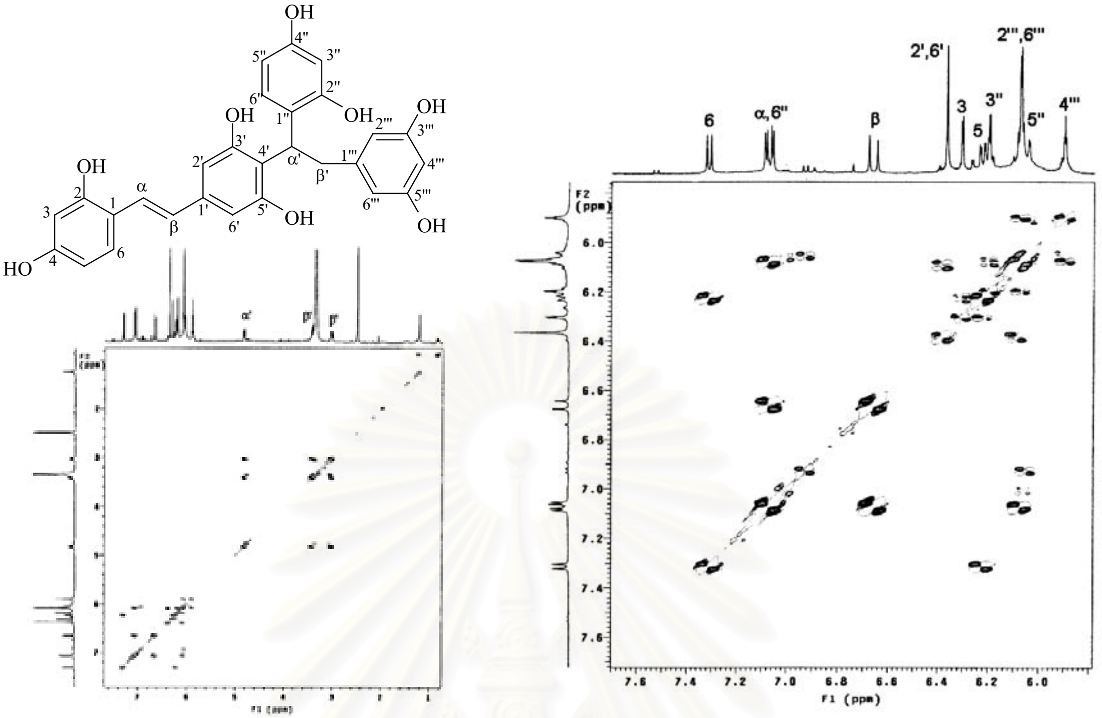

Figure 22 ^{1}H-^{1}H COSY Spectrum of compound AG12 (DMSO-d_6)

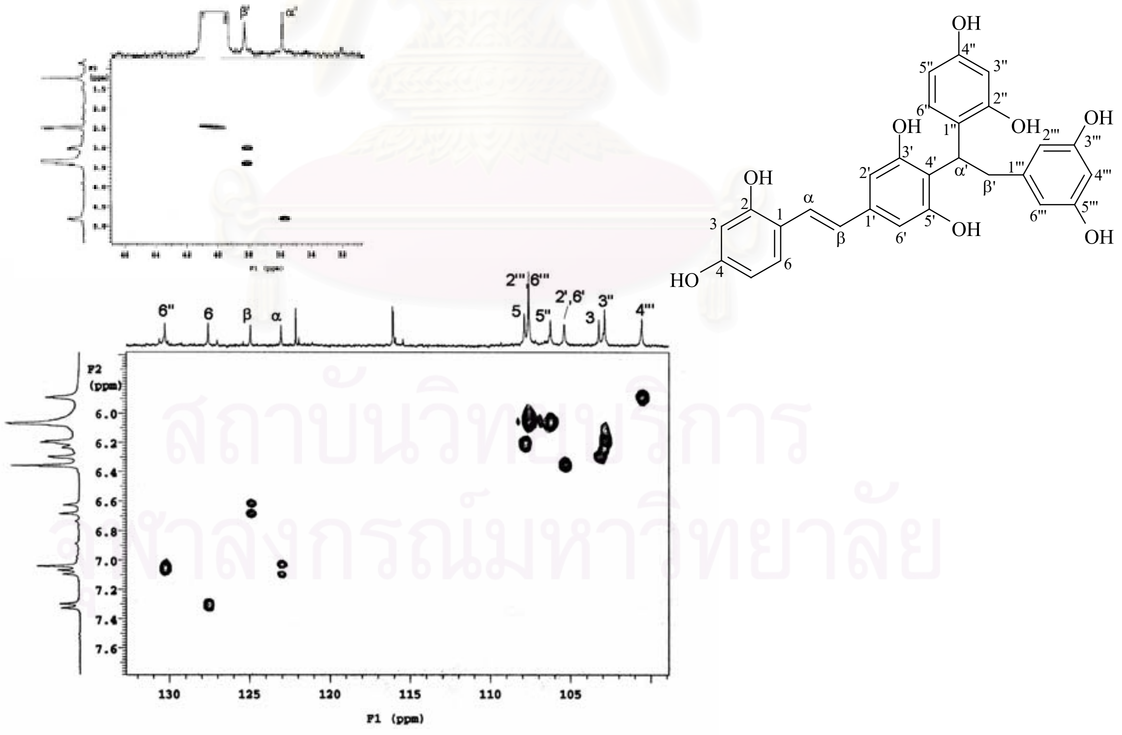

Figure 23 HSQC Spectrum of compound AG12 (DMSO-d_6)

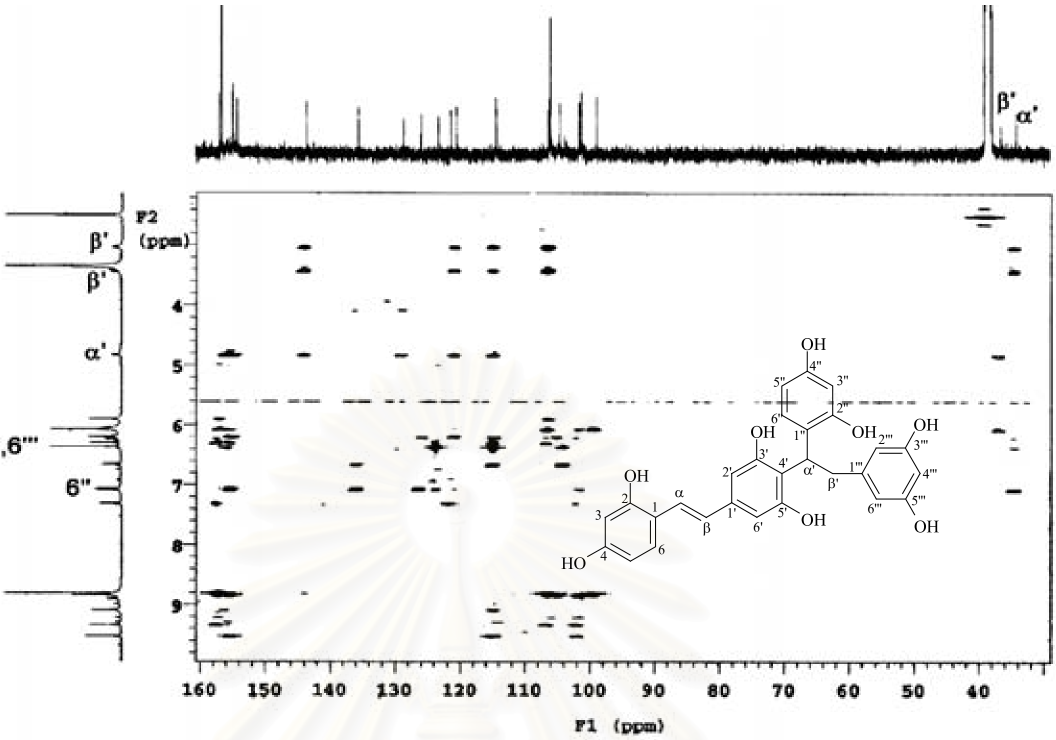

Figure 24 HMBC Spectrum of compound AG12 (DMSO-d_6) [δ_H 2.0-10.0 ppm, δ_C 30-160 ppm]

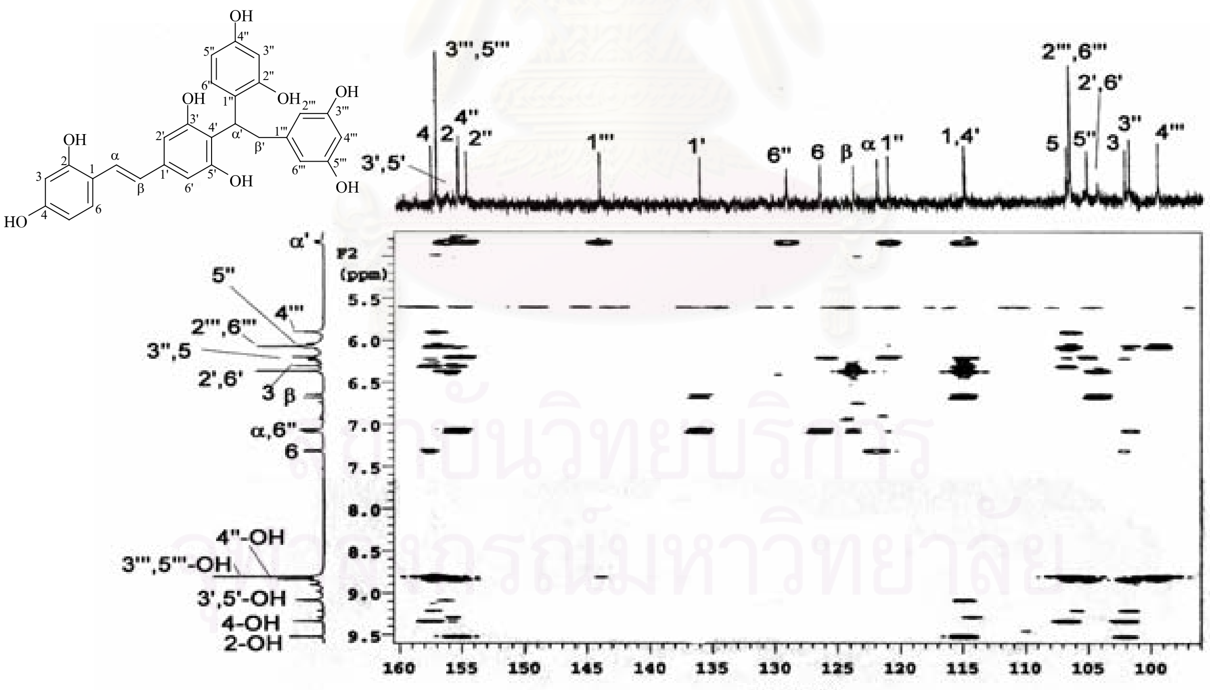

Figure 25 HMBC Spectrum of compound AG12 (DMSO-d_6) [δ_H 4.7-9.5 ppm, δ_C 95-160 ppm]

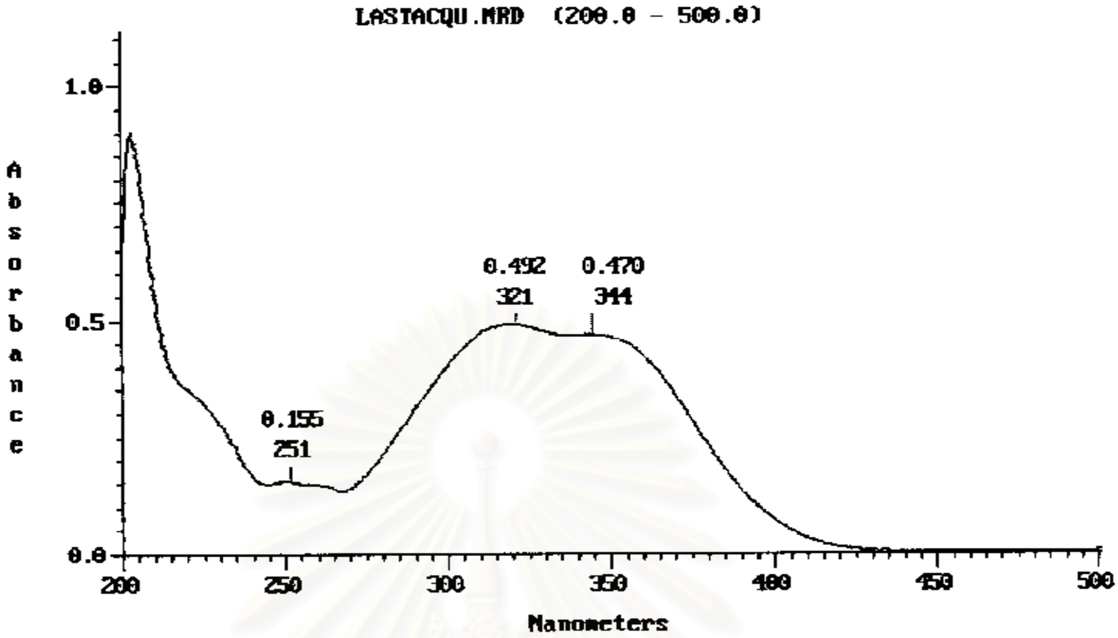

Figure 26 UV Spectrum of compound ME1 (methanol)

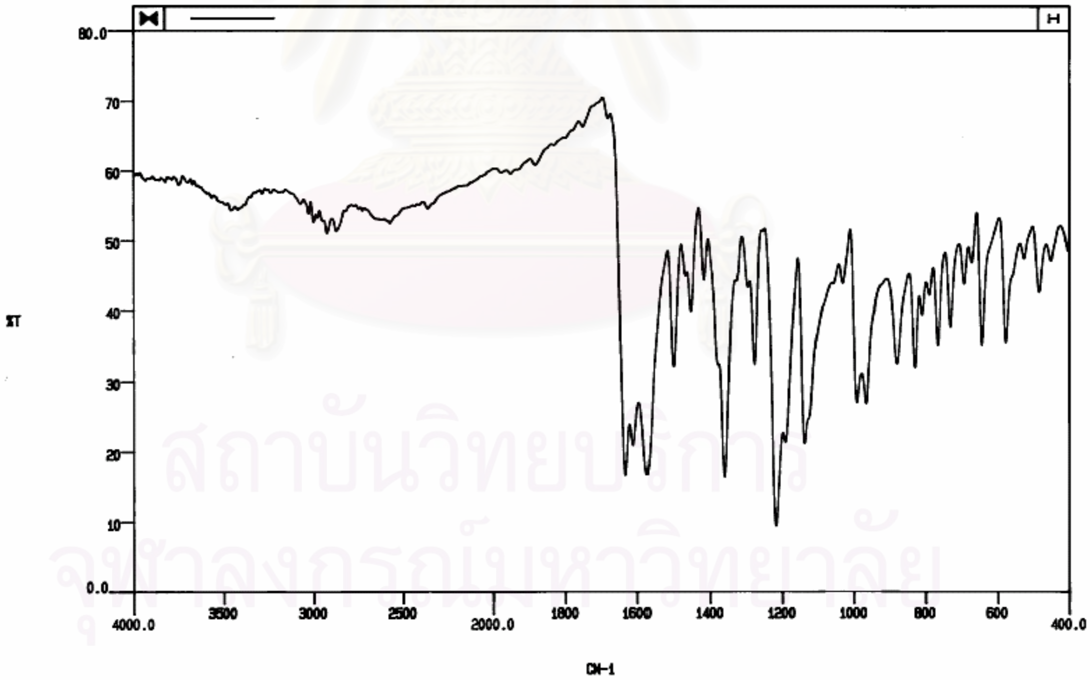

Figure 27 IR Spectrum of compound ME1 (KBr disc)

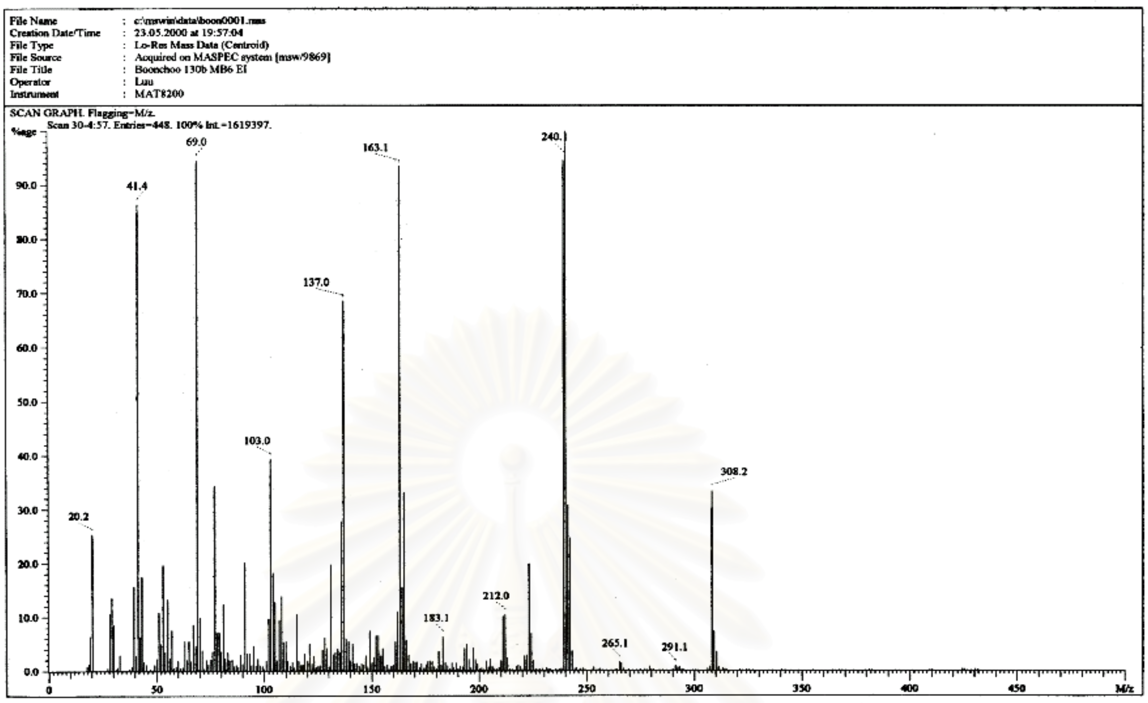

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Figure 28 EI Mass spectrum of compound ME1

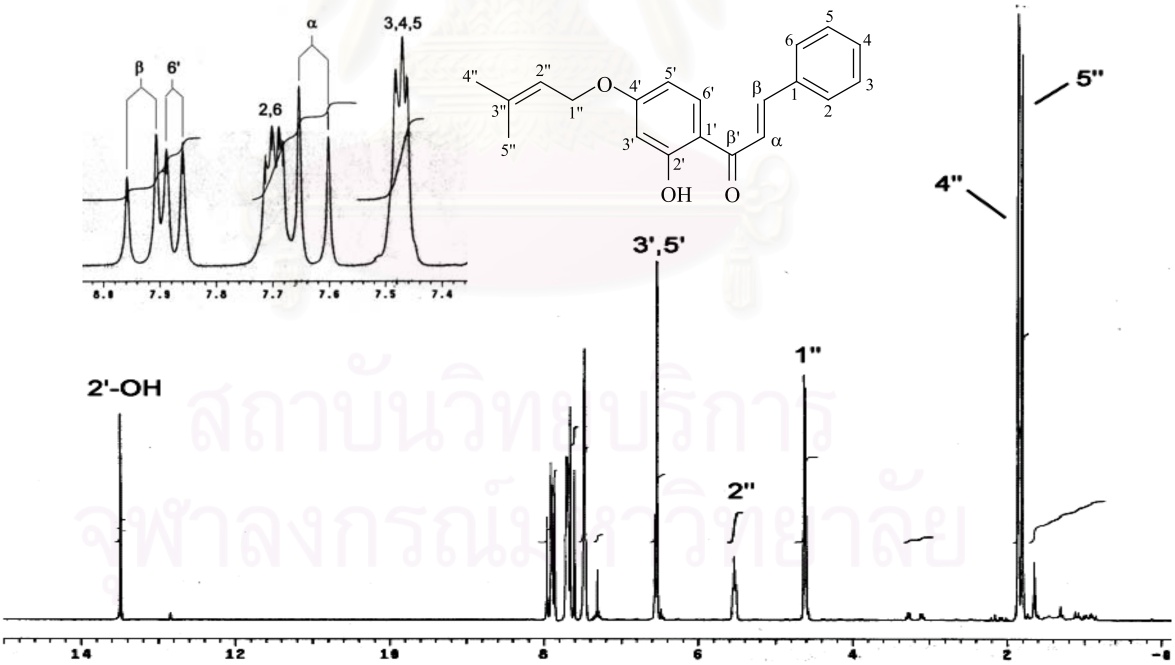

Figure 29 ^{1}H NMR (500 MHz) Spectrum of compound ME1 (CDCl$_3$)

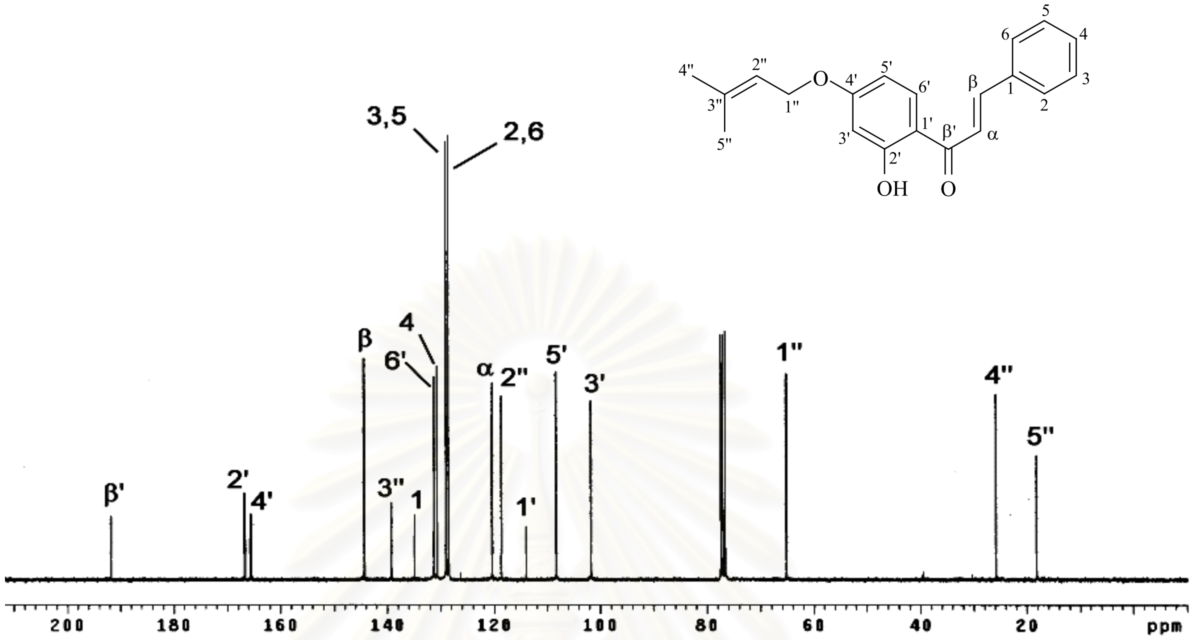

Figure 30 ^{13}C NMR (125 MHz) Spectrum of compound ME1 (CDCl$_3$)

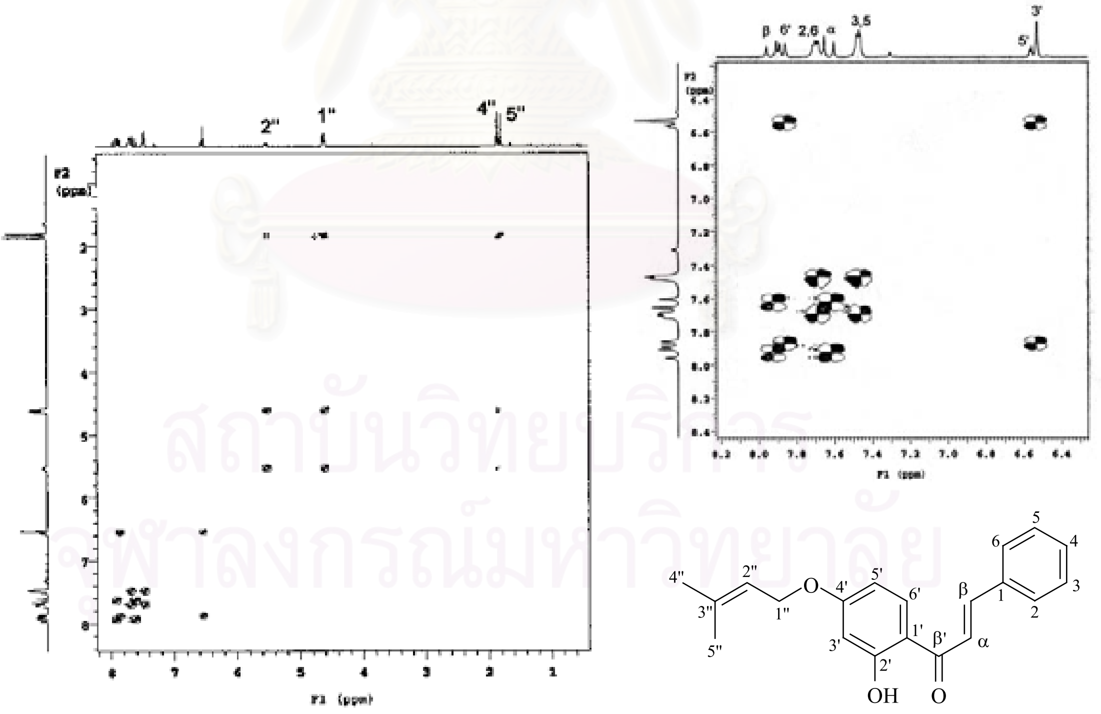

Figure 31 ^{1}H-^{1}H COSY Spectrum of compound ME1 (CDCl$_3$)

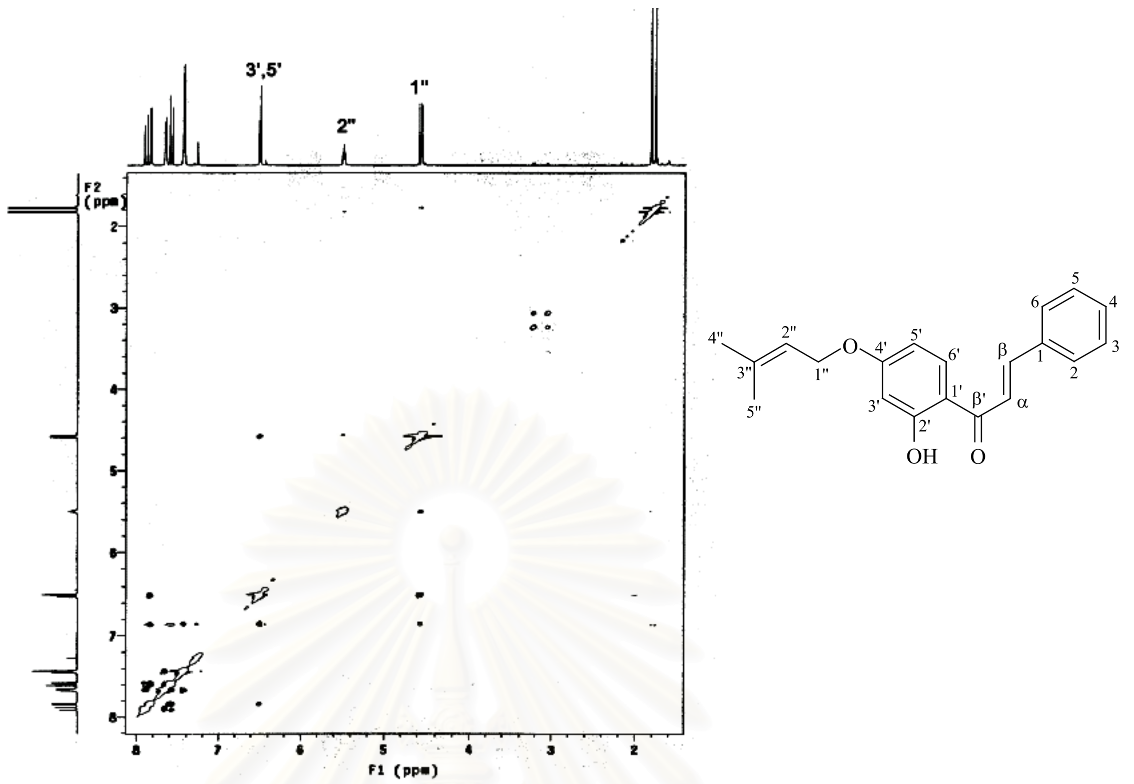

Figure 32 ROESY Spectrum of compound ME1 ($CDCl_3$)

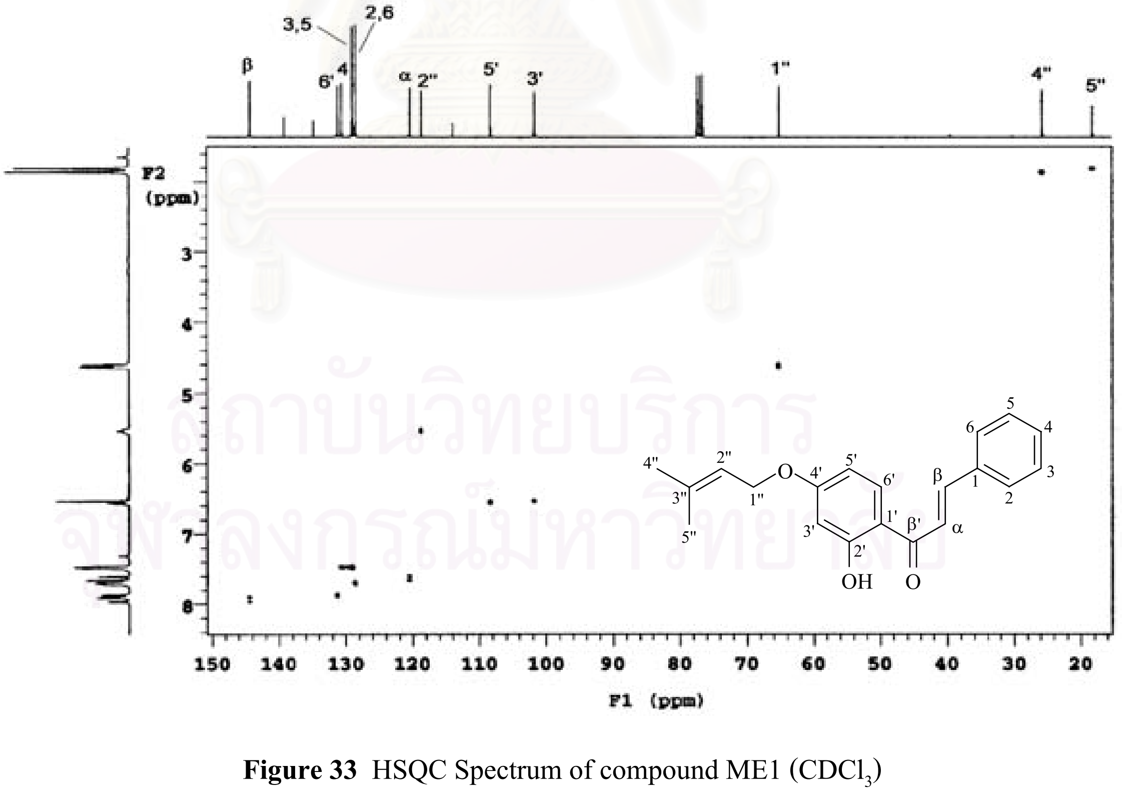

Figure 33 HSQC Spectrum of compound ME1 ($CDCl_3$)

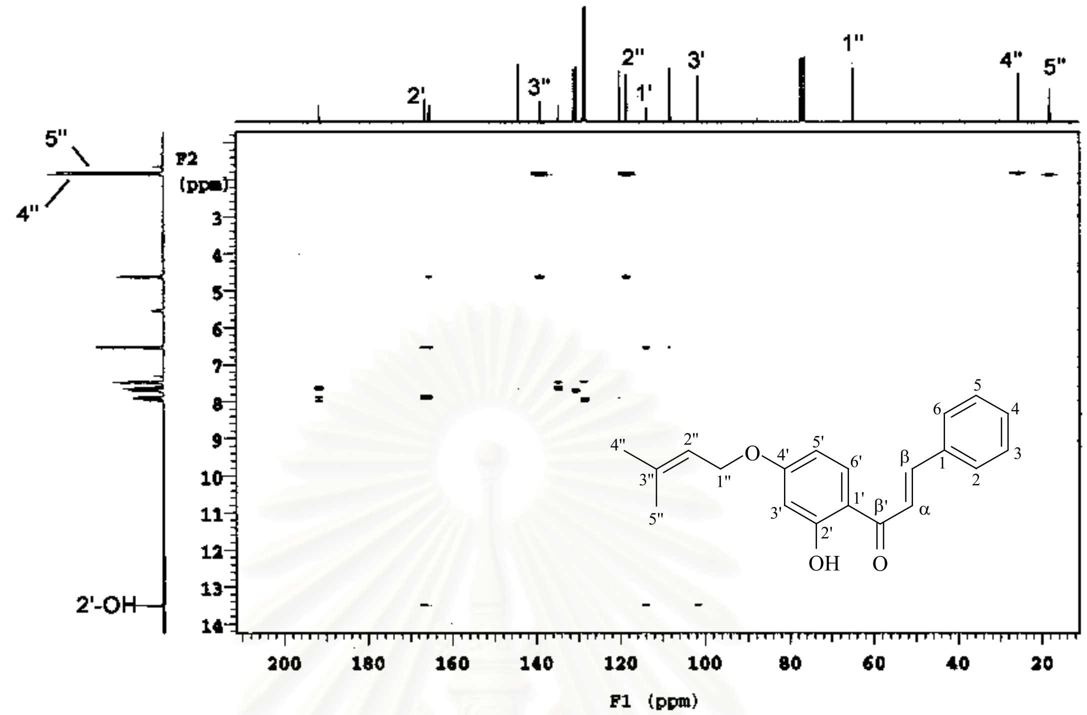

Figure 34 HMBC Spectrum of compound ME1 ($CDCl_3$) [δ_H 0.9-14.1 ppm, δ_C 10-210 ppm]

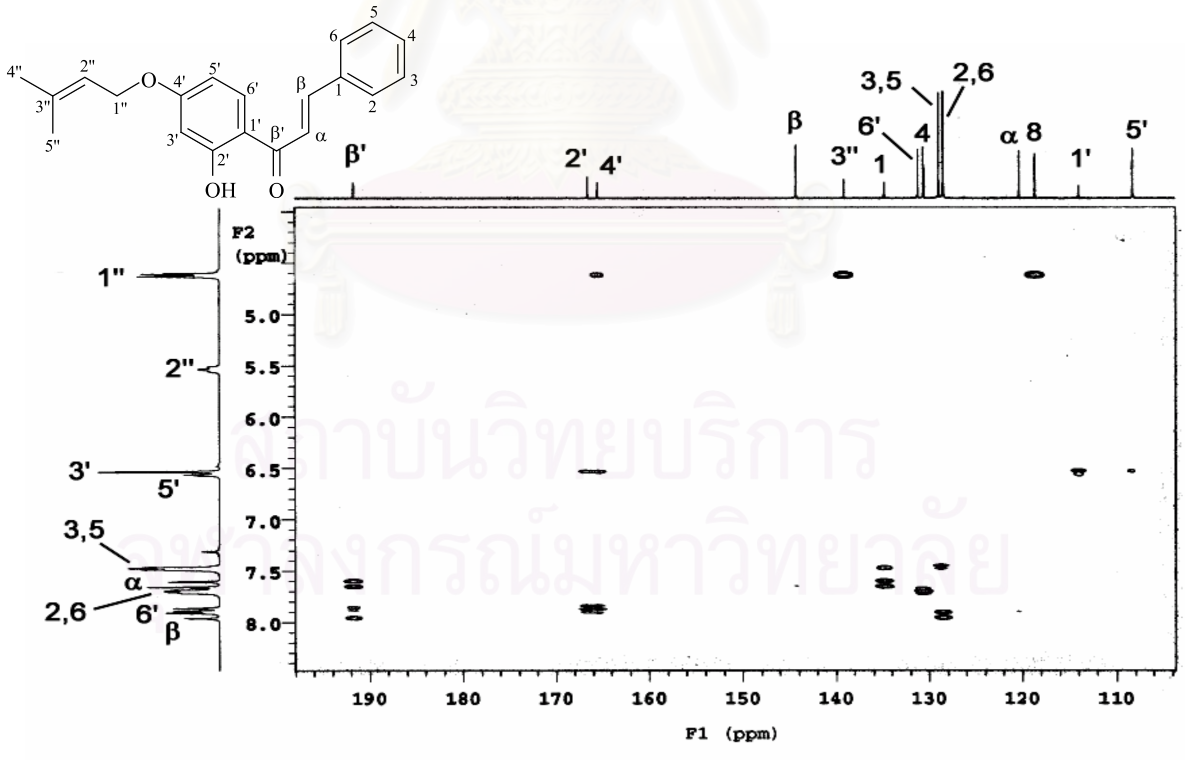

Figure 35 HMBC Spectrum of compound ME1 ($CDCl_3$) [δ_H 4.0-8.5 ppm, δ_C 100-200 ppm]

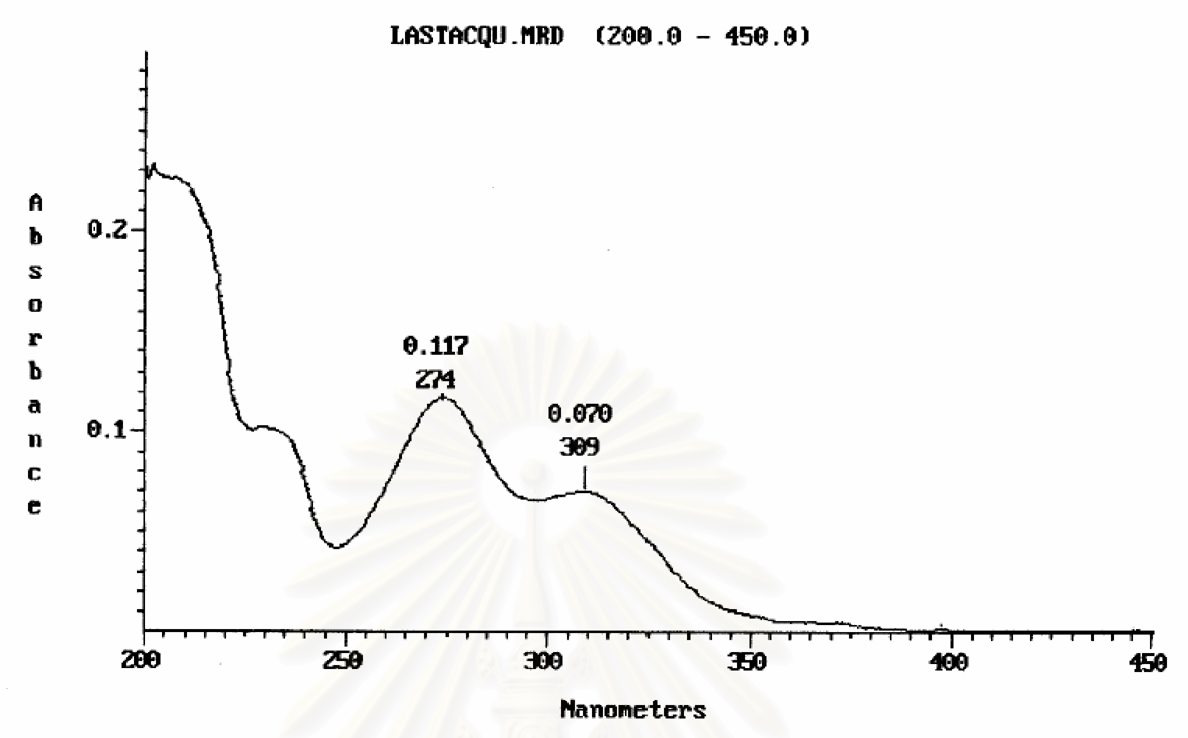

Figure 36 UV Spectrum of compound ME2 (methanol)

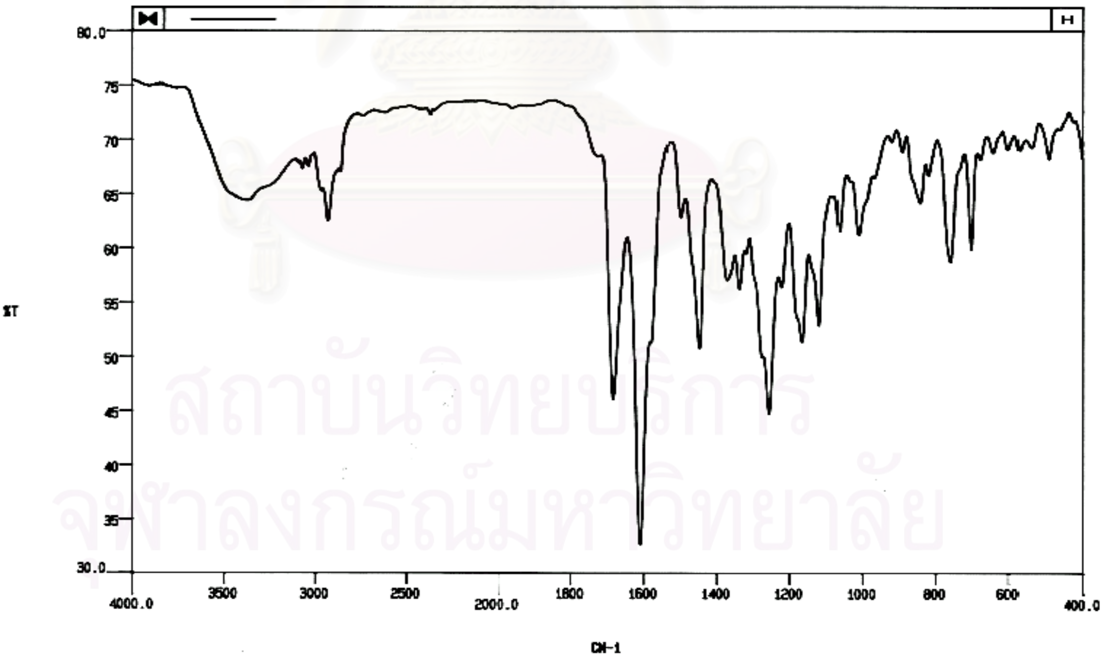

Figure 37 IR Spectrum of compound ME2 (KBr disc)

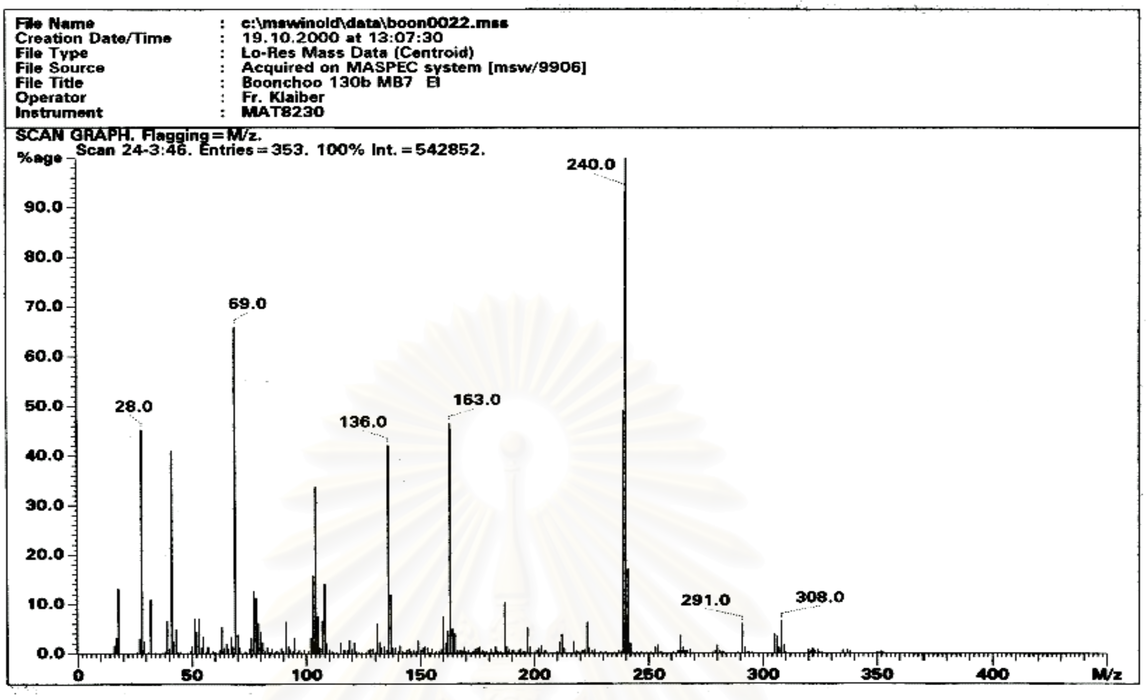

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Figure 38 EI Mass spectrum of compound ME2

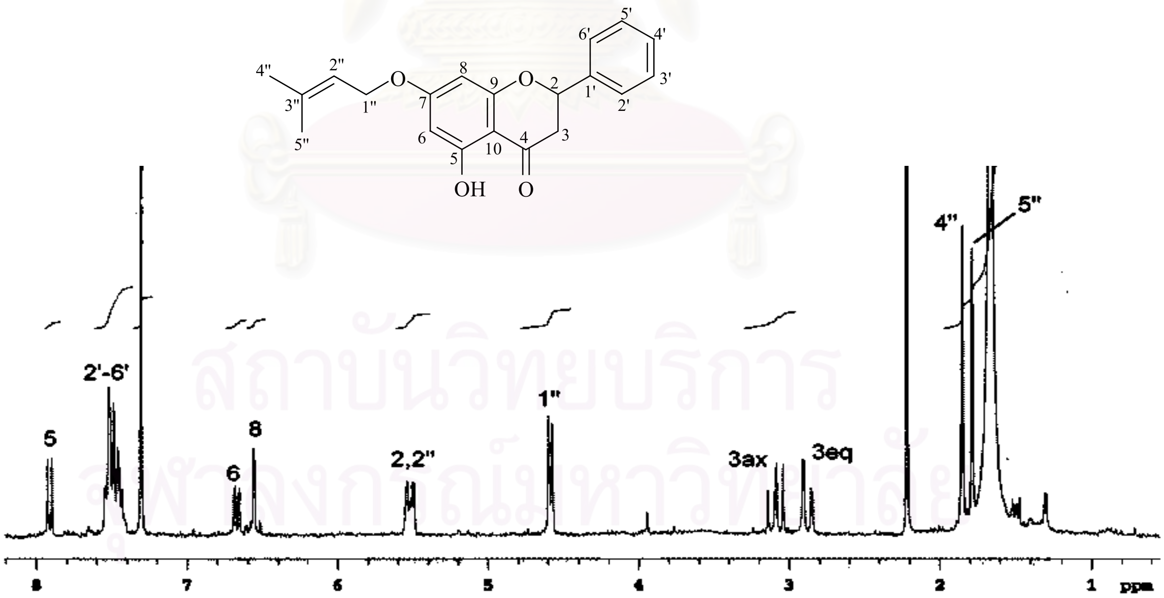

Figure 39 ^{1}H NMR (300 MHz) Spectrum of compound ME2 (CDCl$_3$)

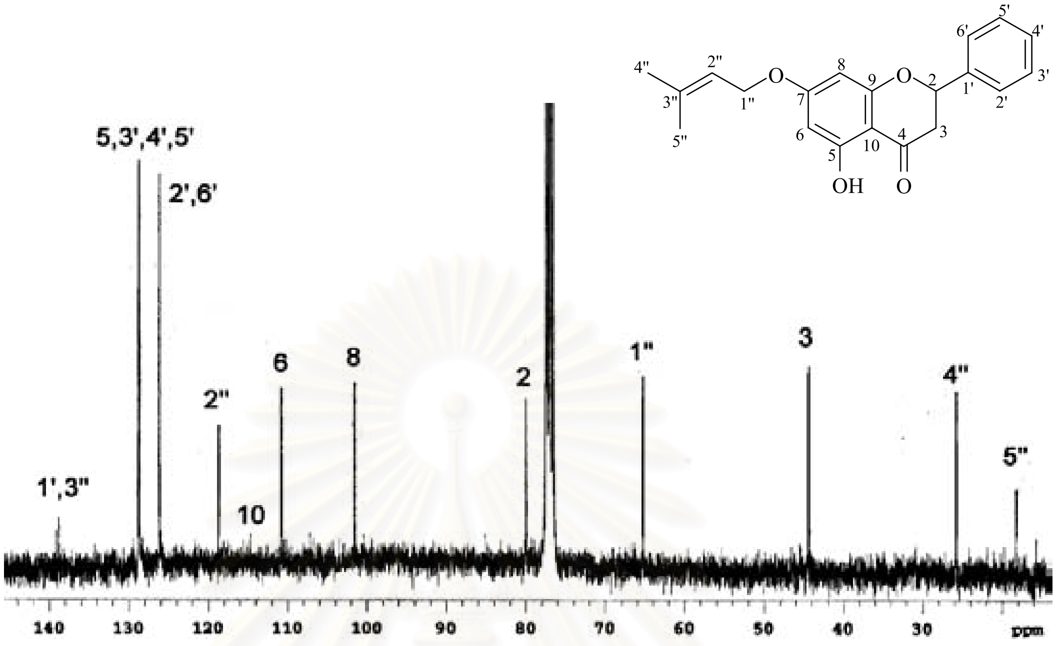

Figure 40 ^{13}C NMR (75 MHz) Spectrum of compound ME2 (CDCl$_3$)

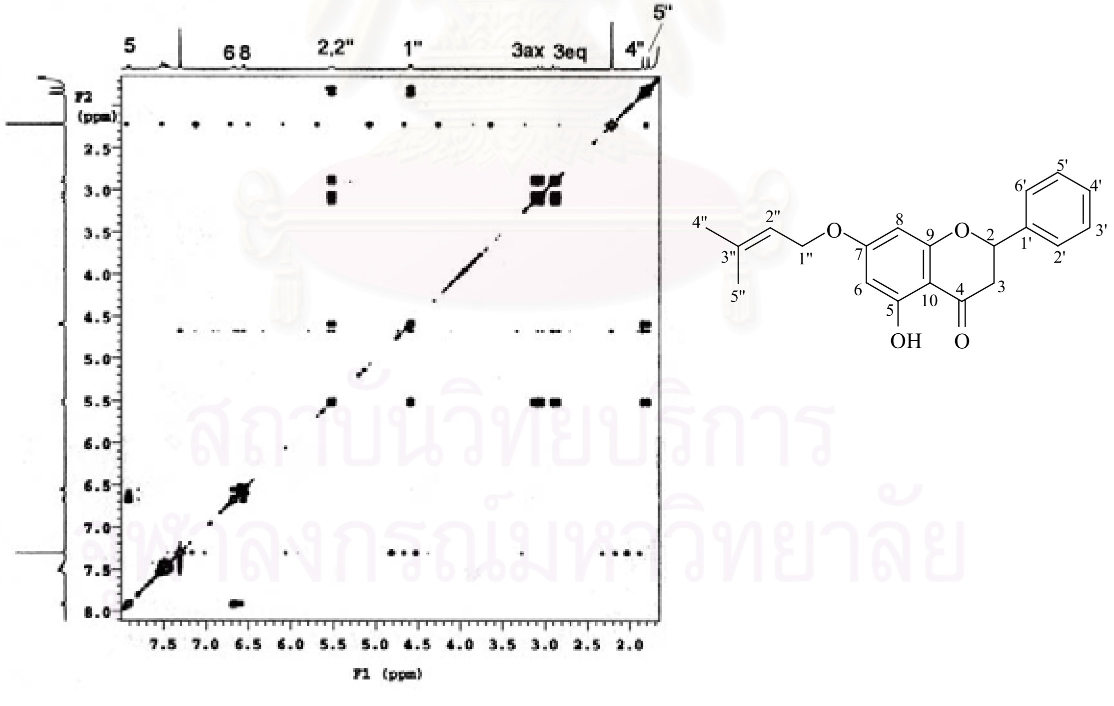

Figure 41 ^{1}H-^{1}H COSY Spectrum of compound ME2 (CDCl$_3$)

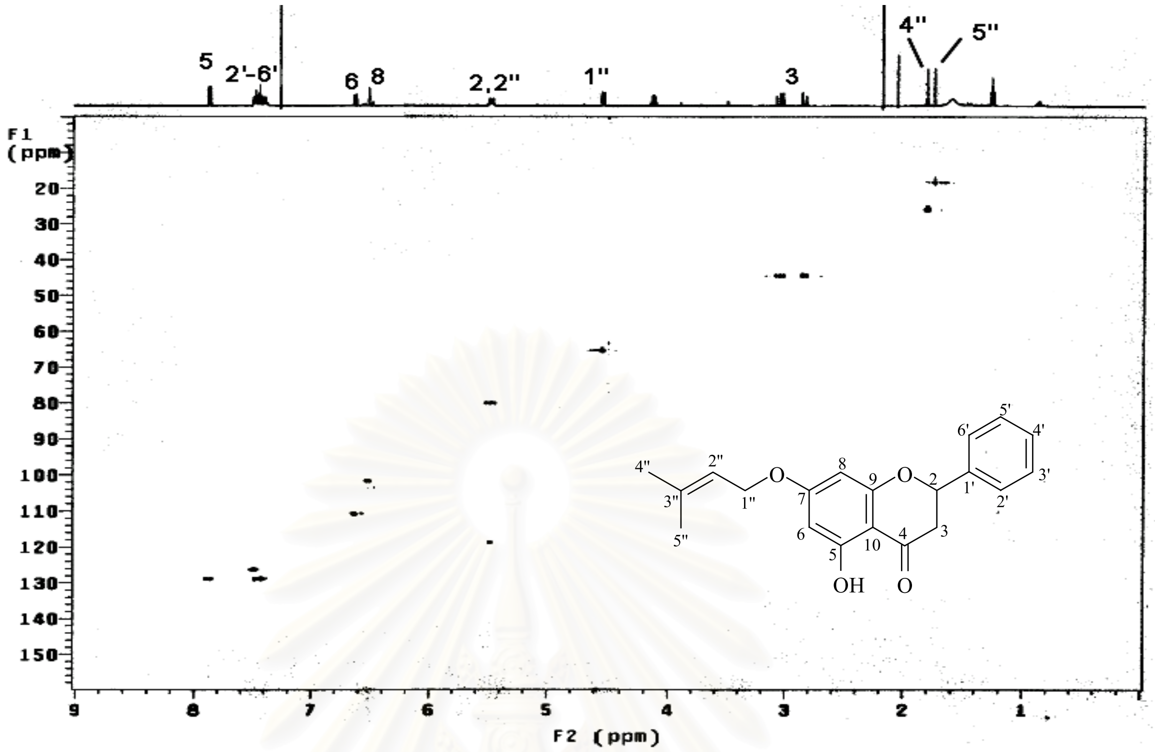

Figure 42 HSQC Spectrum of compound ME2 ($CDCl_3$)

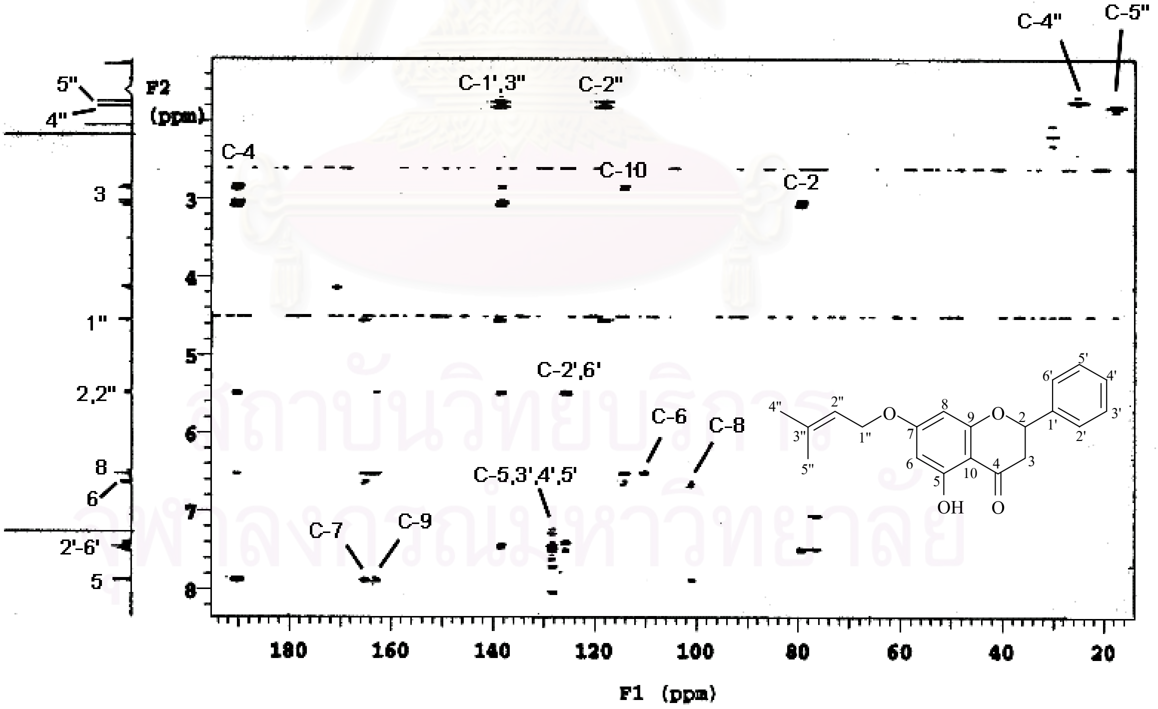

Figure 43 HMBC Spectrum of compound ME2 ($CDCl_3$) [δ_H 1.0-8.4 ppm, δ_C 100-200 ppm]

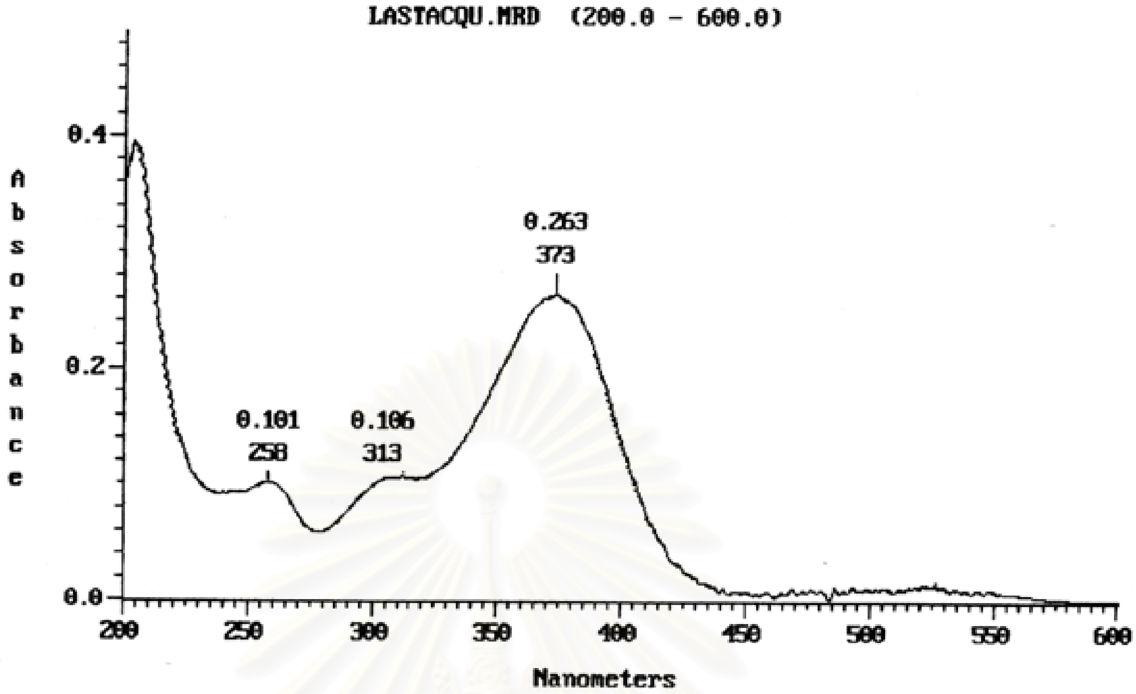

Figure 44 UV Spectrum of compound ME3 (methanol)

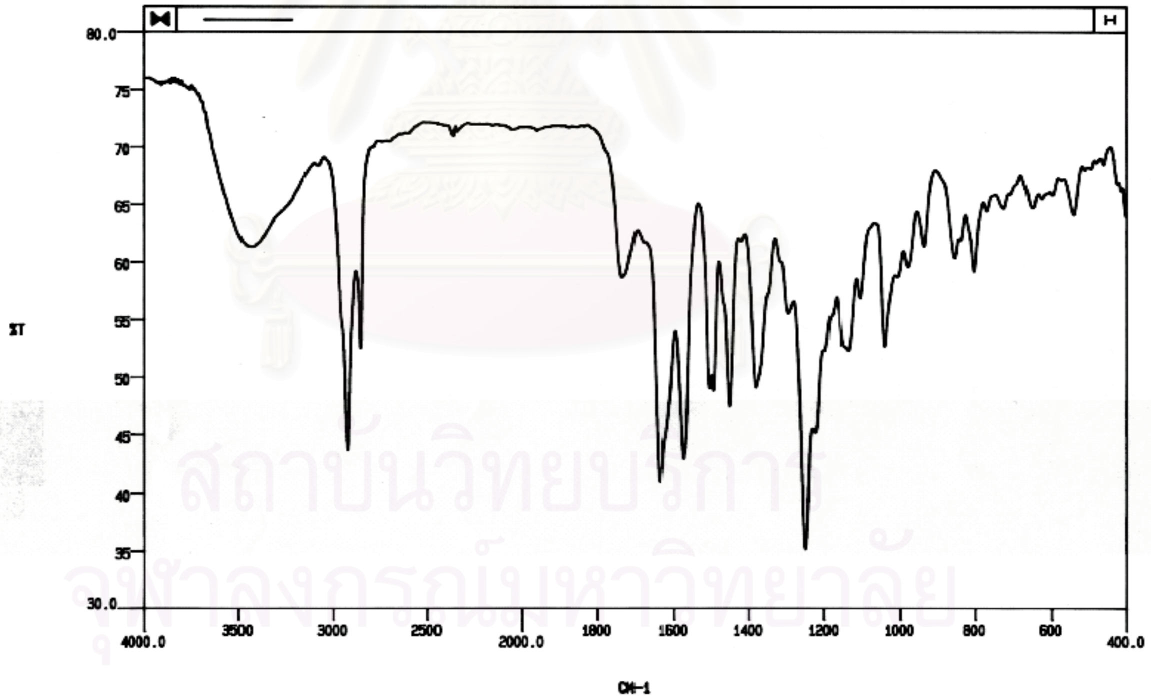

Figure 45 IR Spectrum of compound ME3 (KBr disc)

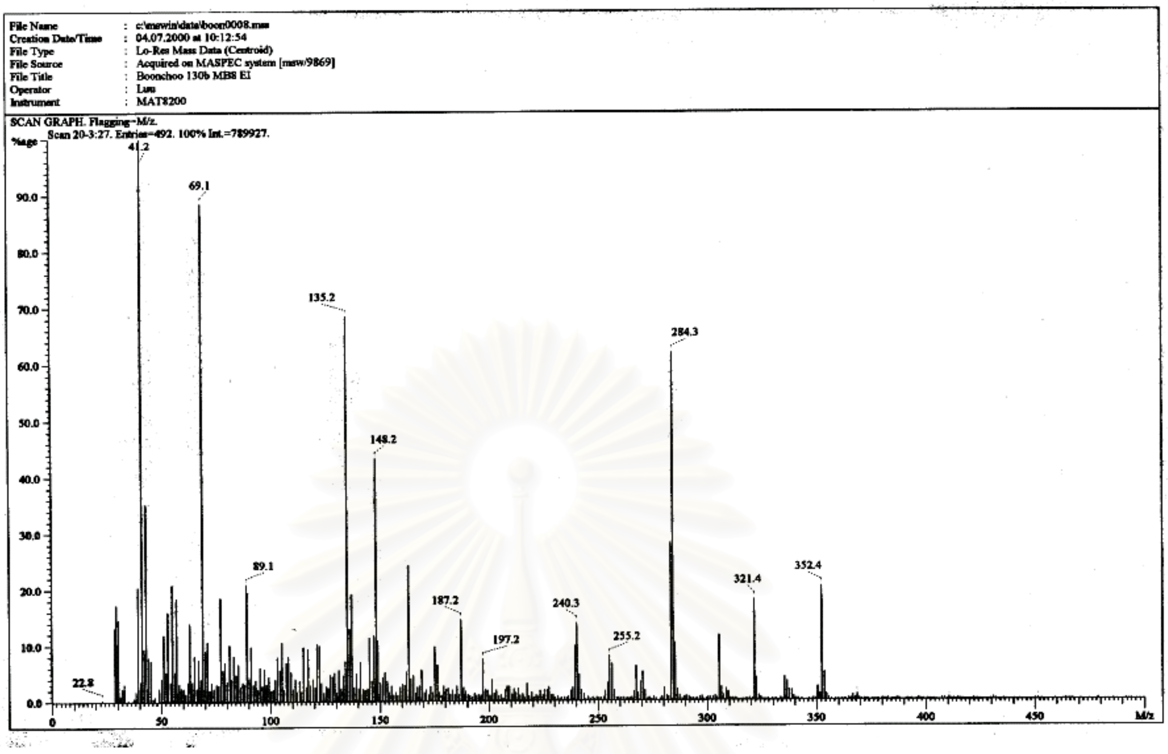

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Figure 46 EI Mass spectrum of compound ME3

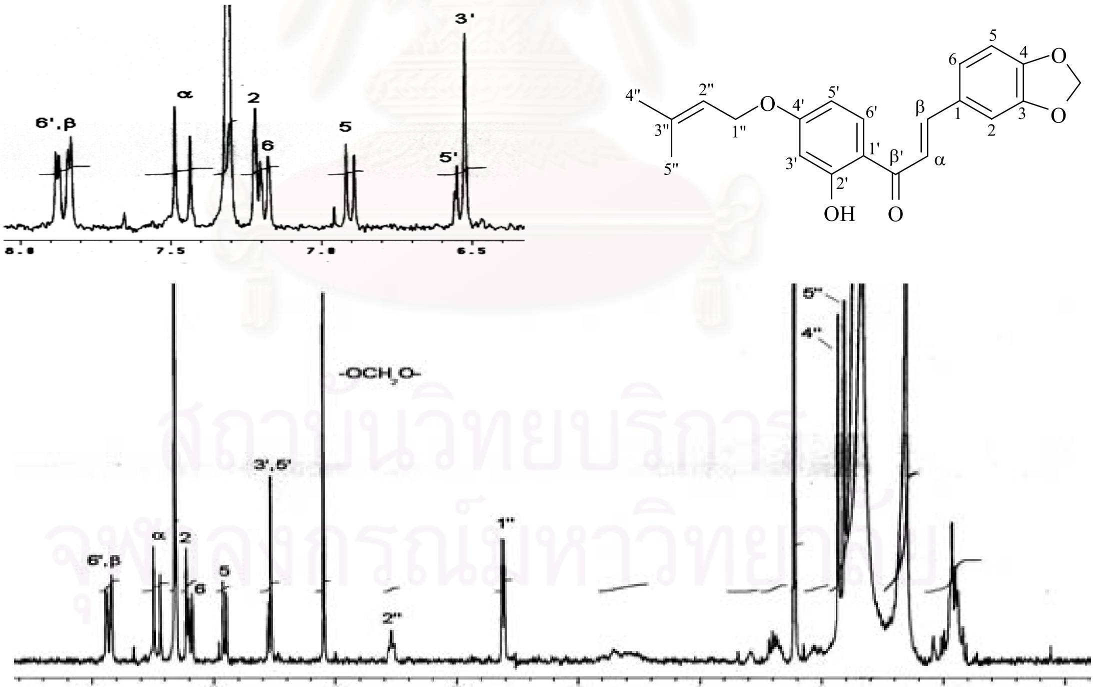

Figure 47 ^{1}H NMR (500 MHz) Spectrum of compound ME3 (CDCl$_3$)

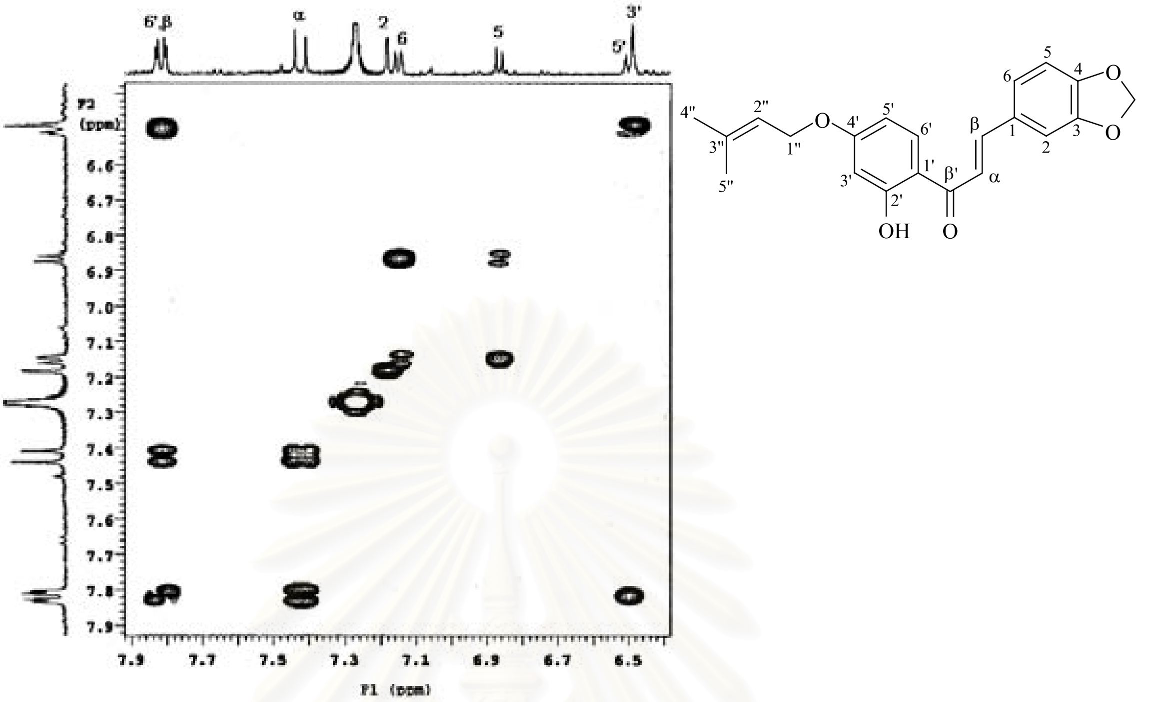

Figure 48 ^{1}H-^{1}H COSY Spectrum of compound ME3 (CDCl$_3$)

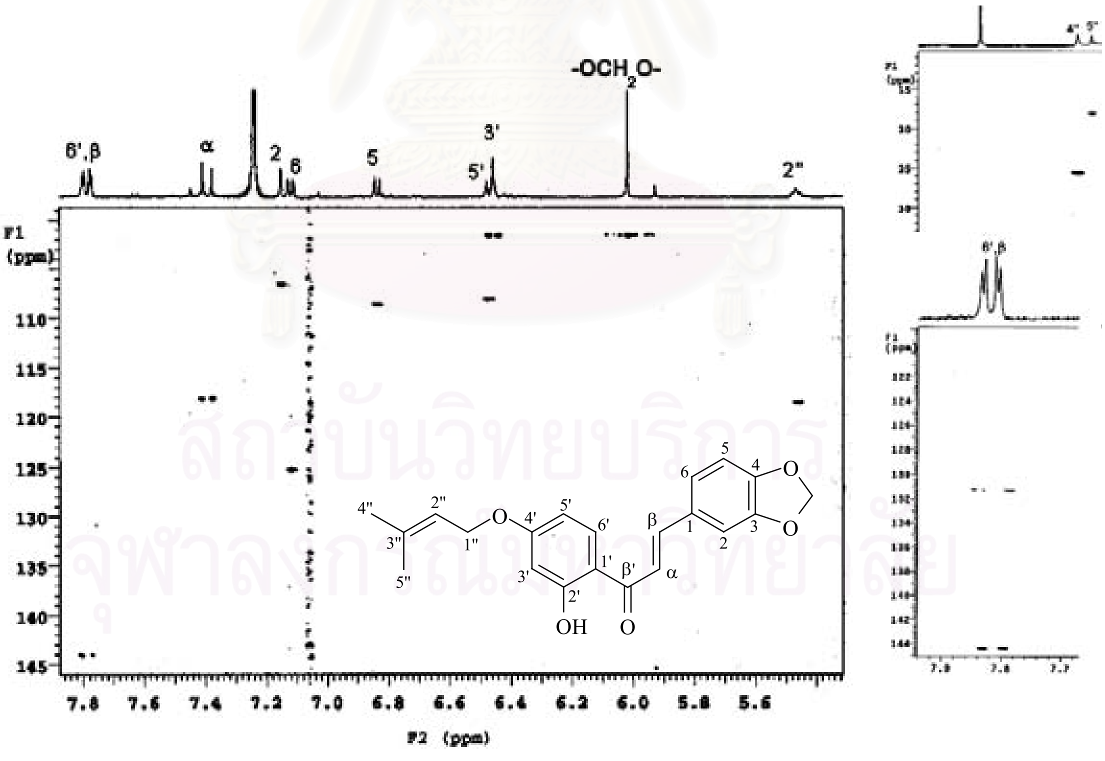

Figure 49 HSQC Spectrum of compound ME3 (CDCl$_3$)

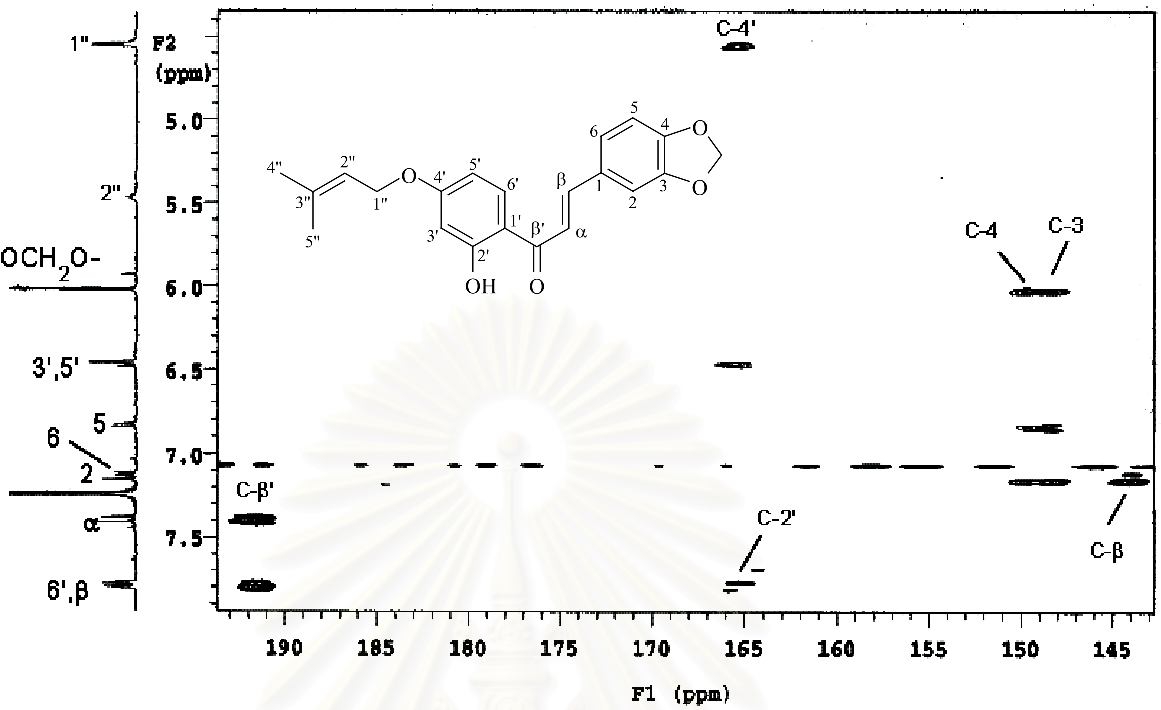

Figure 50 HMBC Spectrum of compound ME3 (CDCl$_3$) [δ_H 4.8-8.0 ppm, δ_C 140-195 ppm]

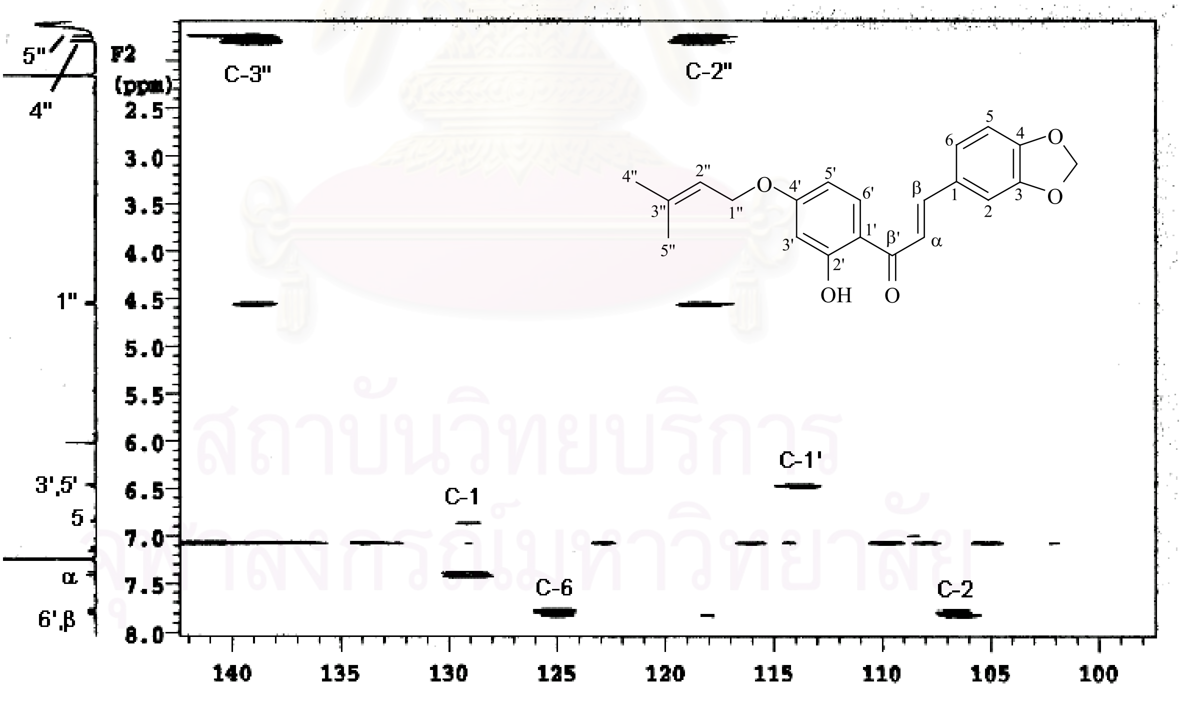

Figure 51 HMBC Spectrum of compound ME3 (CDCl$_3$) [δ_H 1.5-8.0 ppm, δ_C 97-142 ppm]

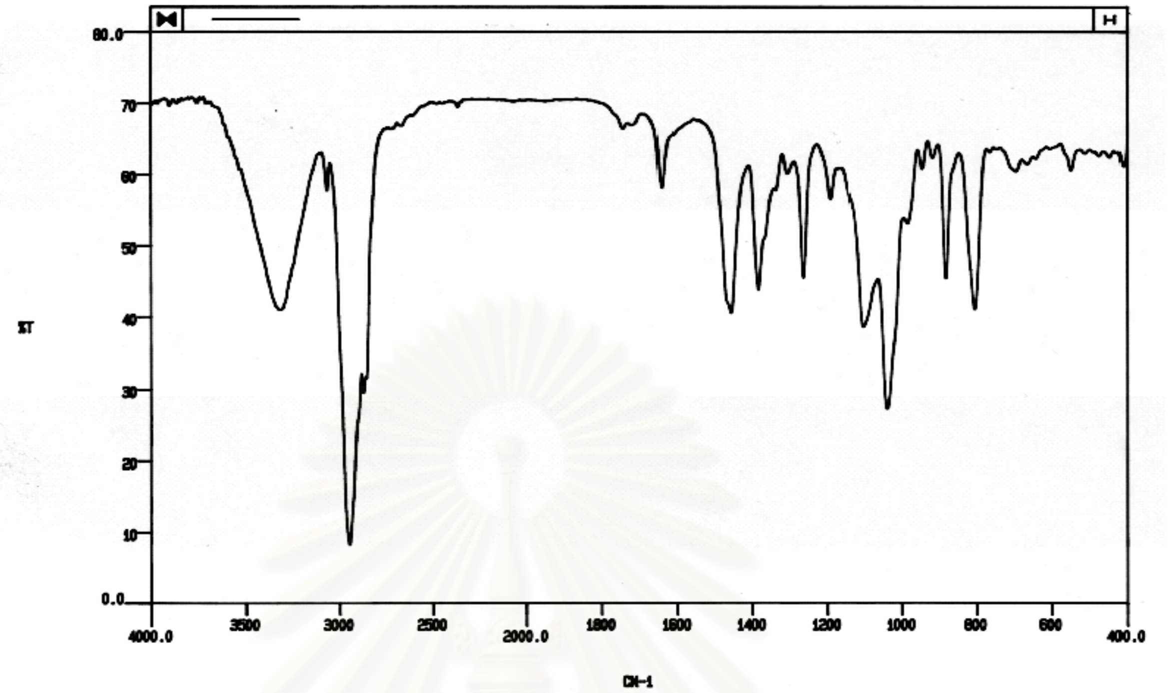

Figure 52 IR Spectrum of compound ME4 (KBr disc)

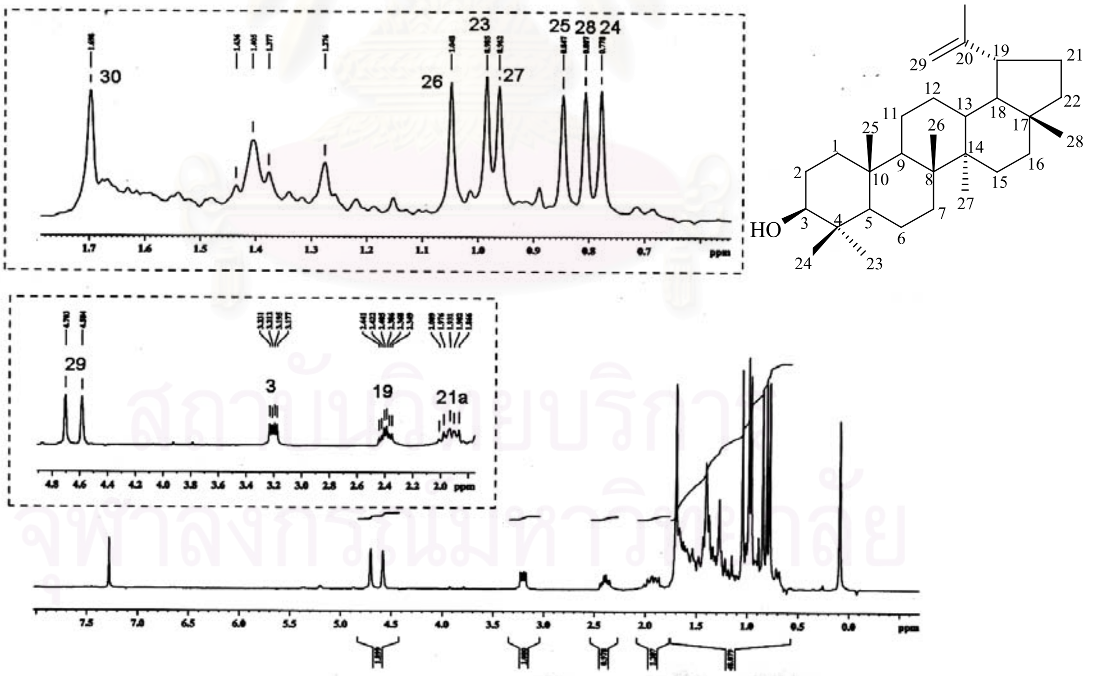

Figure 53 ^{1}H NMR (300 MHz) Spectrum of compound ME4 ($CDCl_3$)

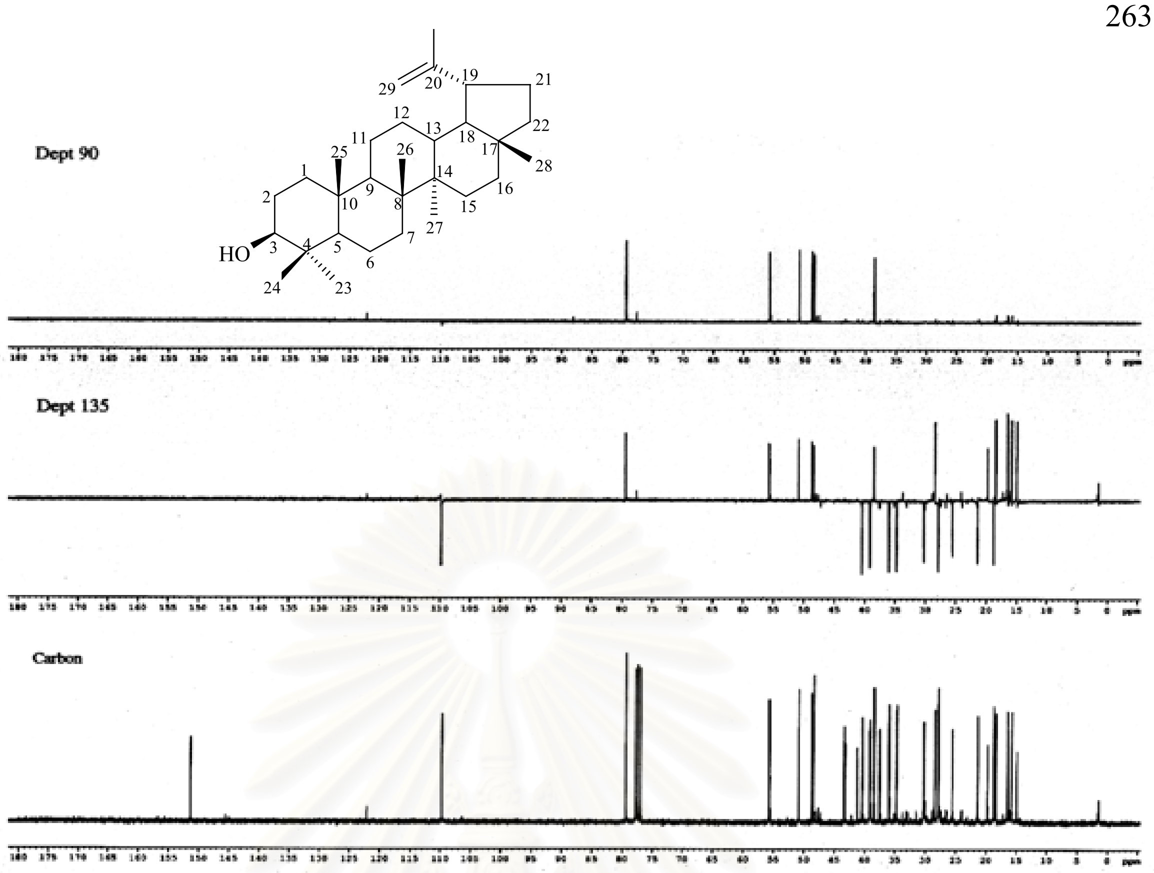

Figure 54 DEPT 90 and DEPT 135 Spectra of compound ME4 ($CDCl_3$)

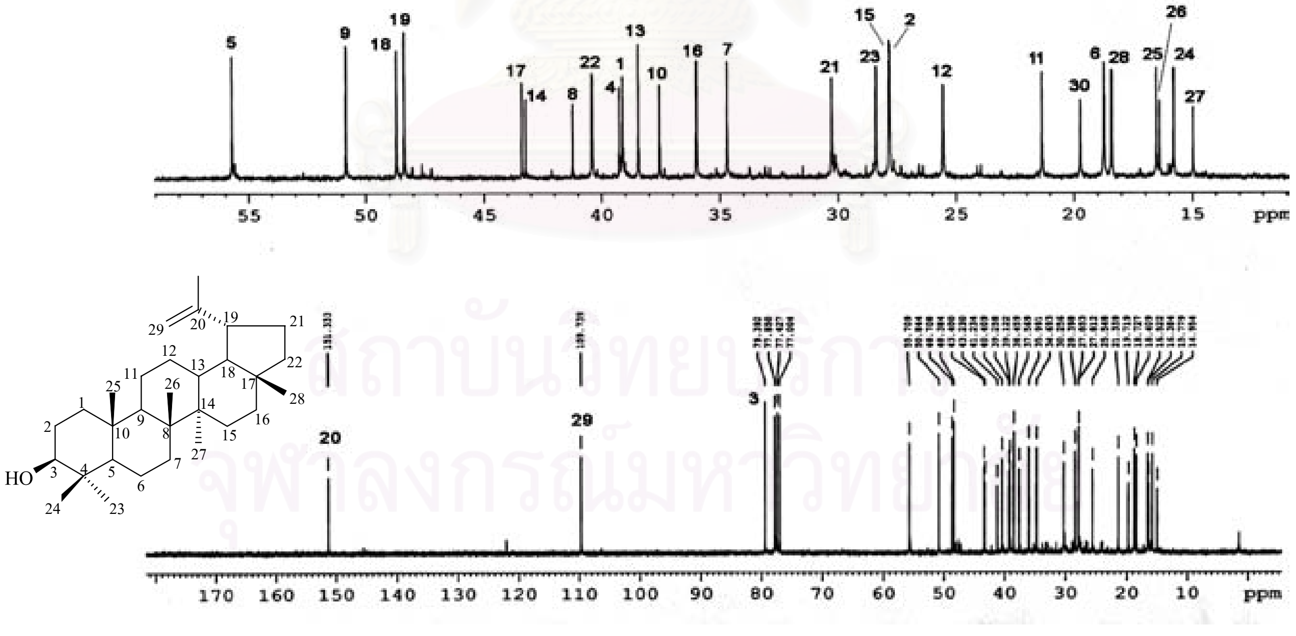

Figure 55 ^{13}C NMR (75 MHz) Spectrum of compound ME4 ($CDCl_3$)

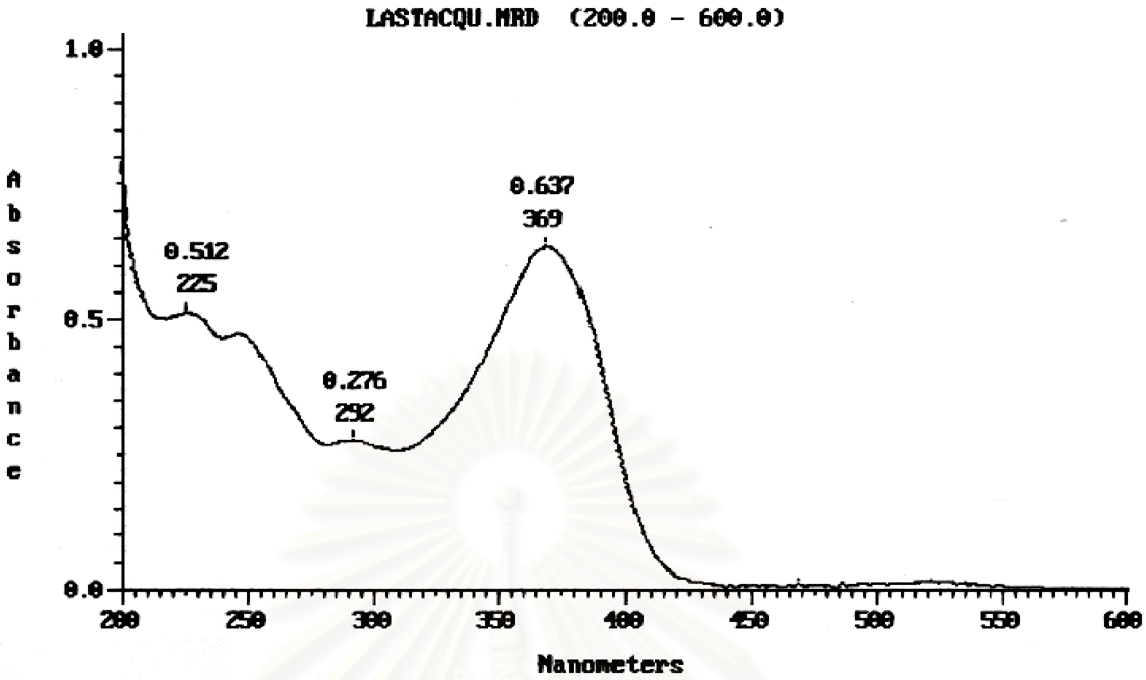

Figure 56 UV Spectrum of compound ME5 (methanol)

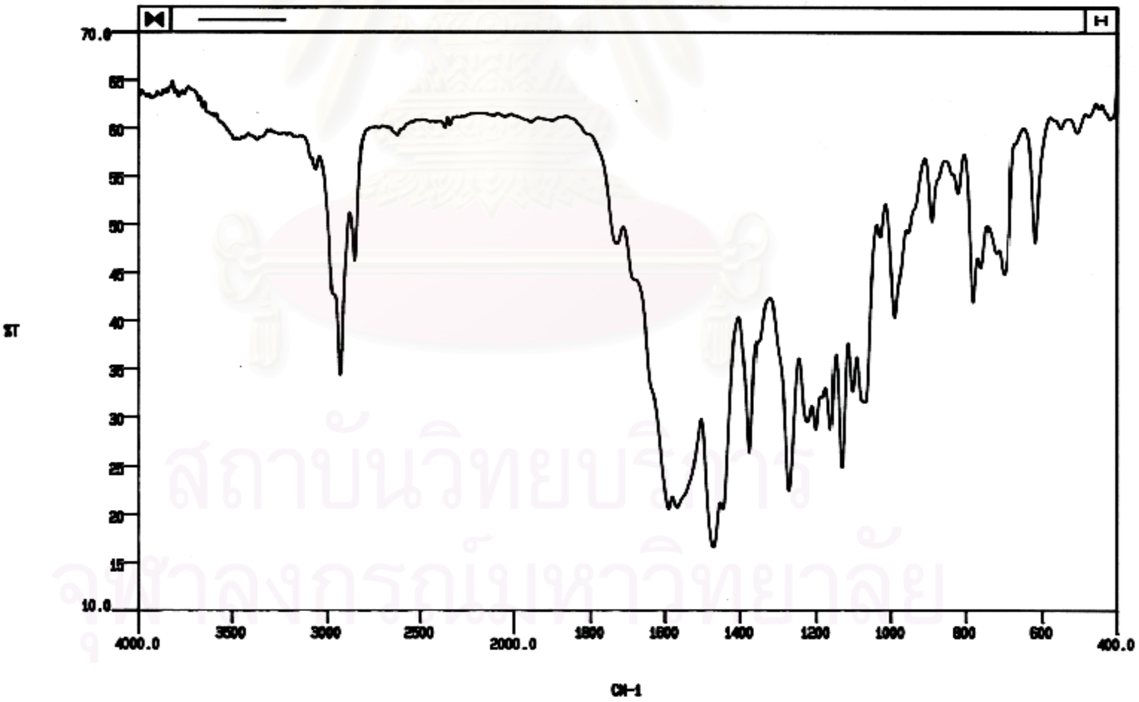

Figure 57 IR Spectrum of compound ME5 (KBr disc)

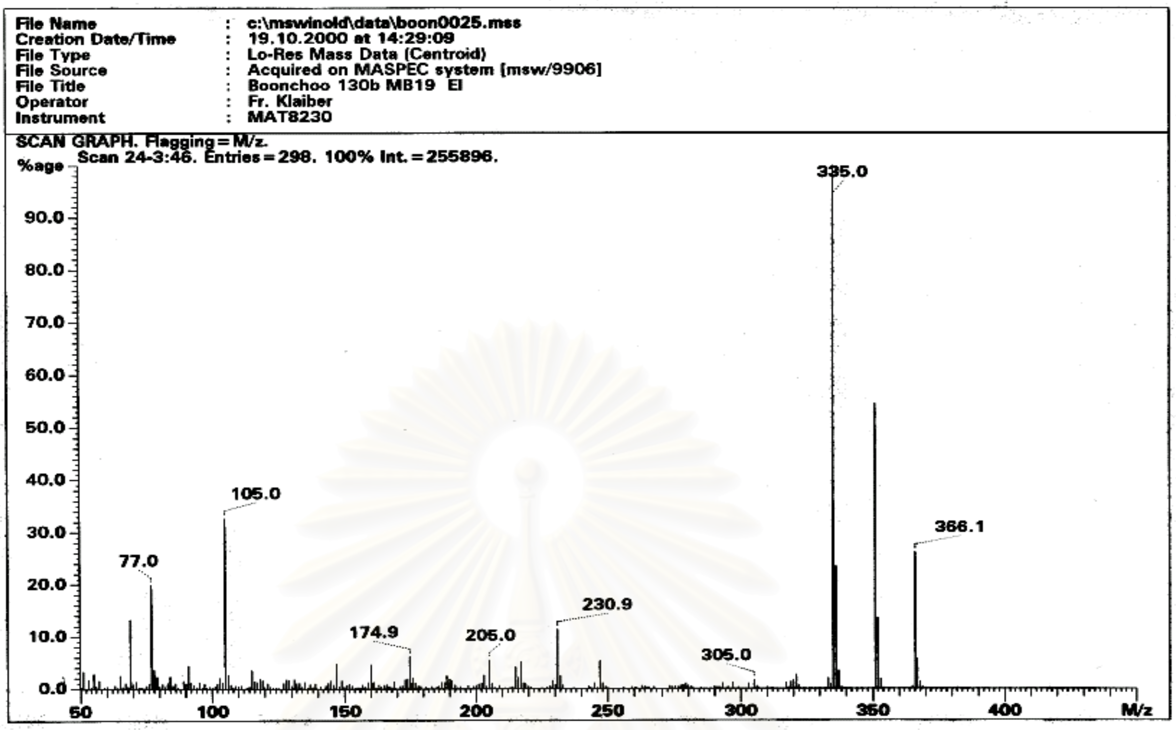

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Figure 58 EI Mass spectrum of compound ME5

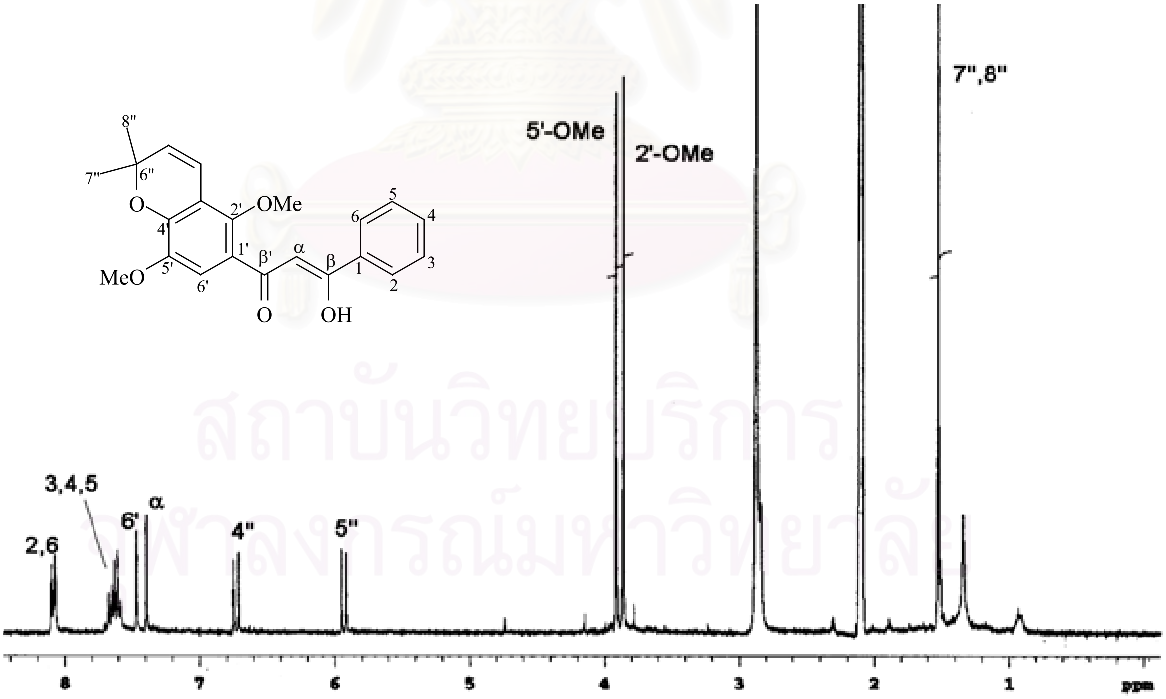

Figure 59 ^{1}H NMR (300 MHz) Spectrum of compound ME5 (acetone-d_6)

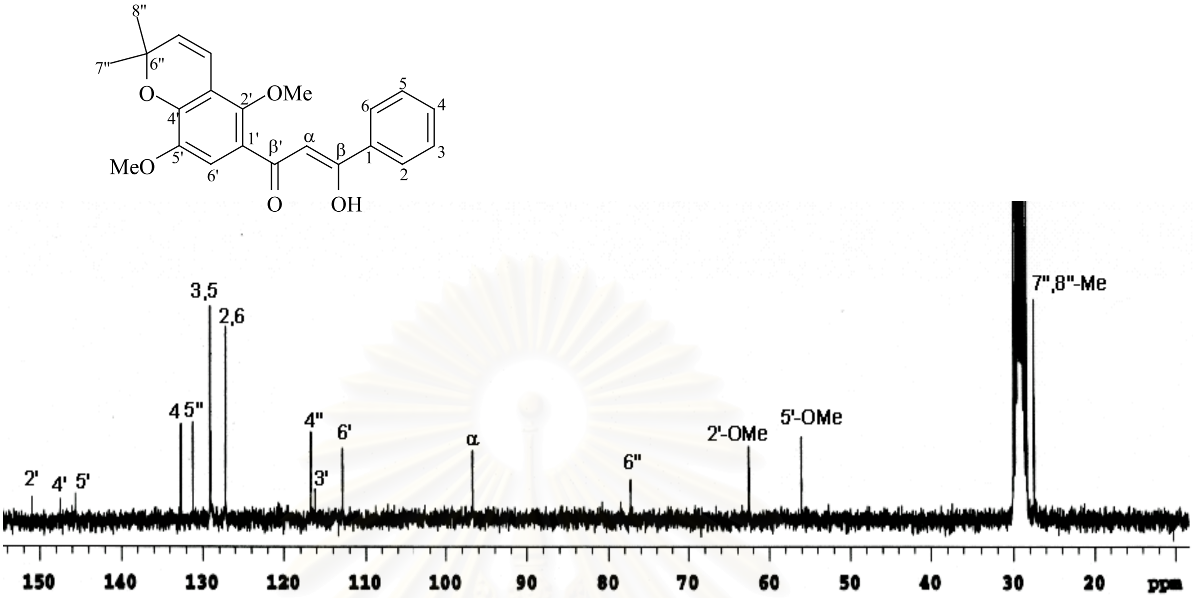

Figure 60 ^{13}C NMR (75 MHz) Spectrum of compound ME5 (acetone-d_6)

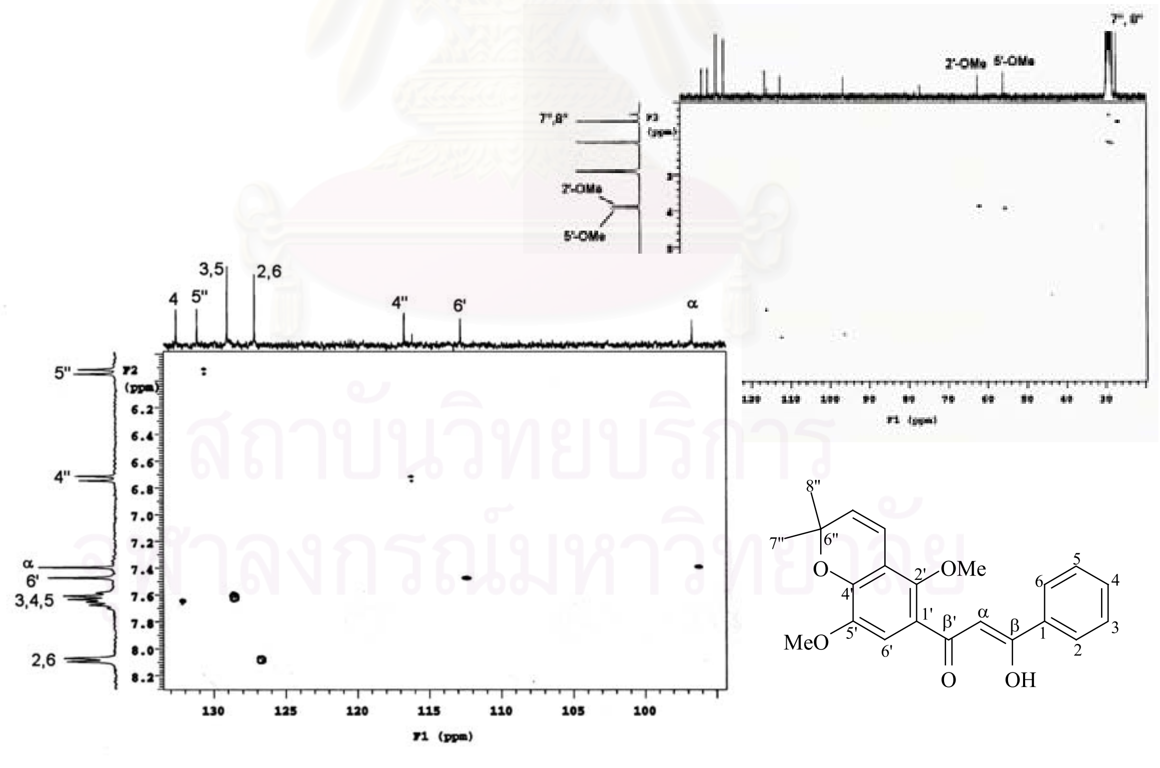

Figure 61 HSQC Spectrum of compound ME5 (acetone-d_6)

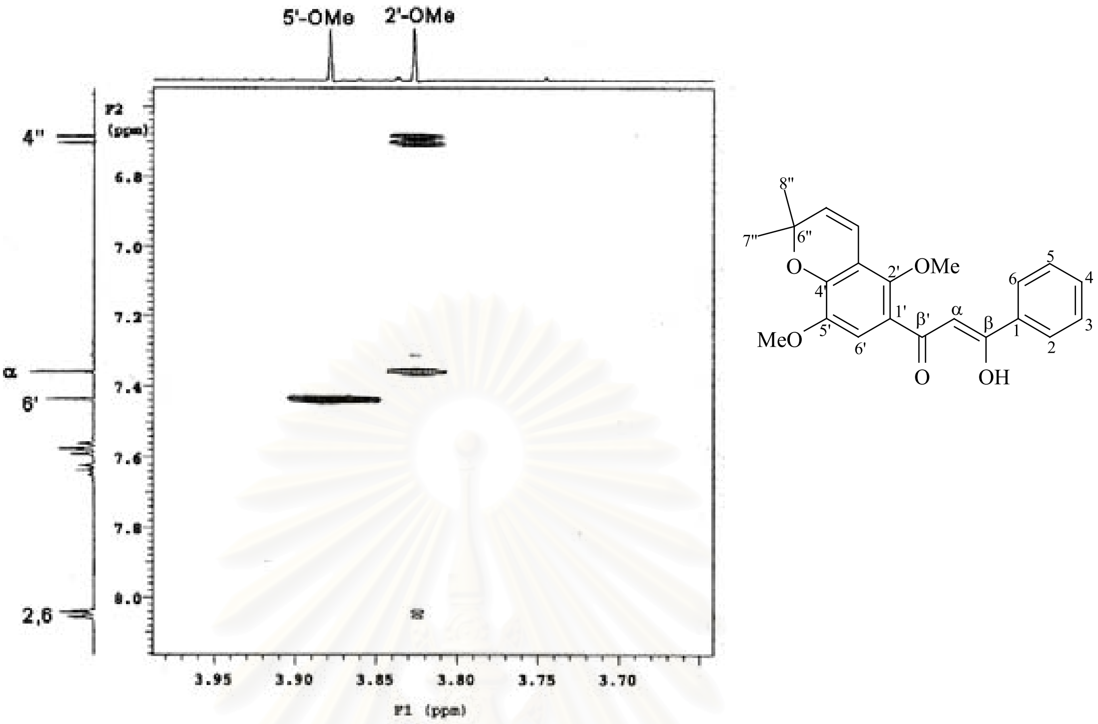

Figure 62 NOESY Spectrum of compound ME5 (acetone-d_6)

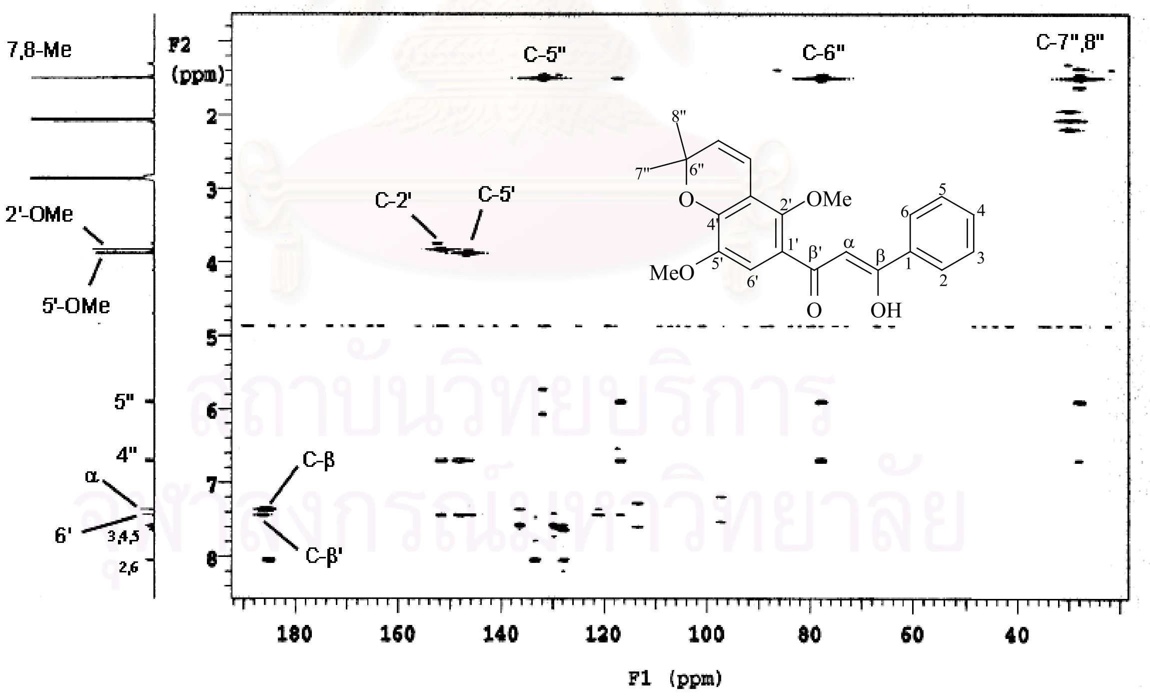

Figure 63 HMBC Spectrum of compound ME5 (acetone-d_6) [δ_H 0.8-8.6 ppm, δ_C 29-186 ppm]

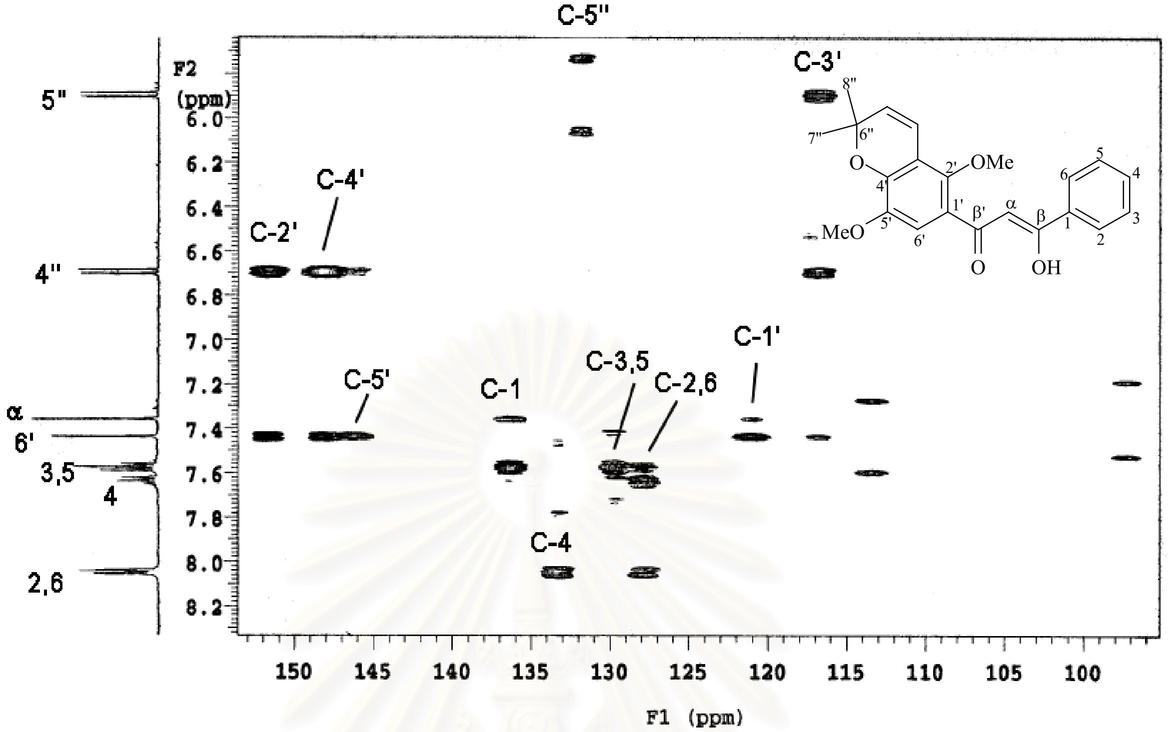

Figure 64 HMBC Spectrum of compound ME5 (acetone-d_6) [δ_H 5.6-8.3 ppm, δ_C 95-154 ppm]

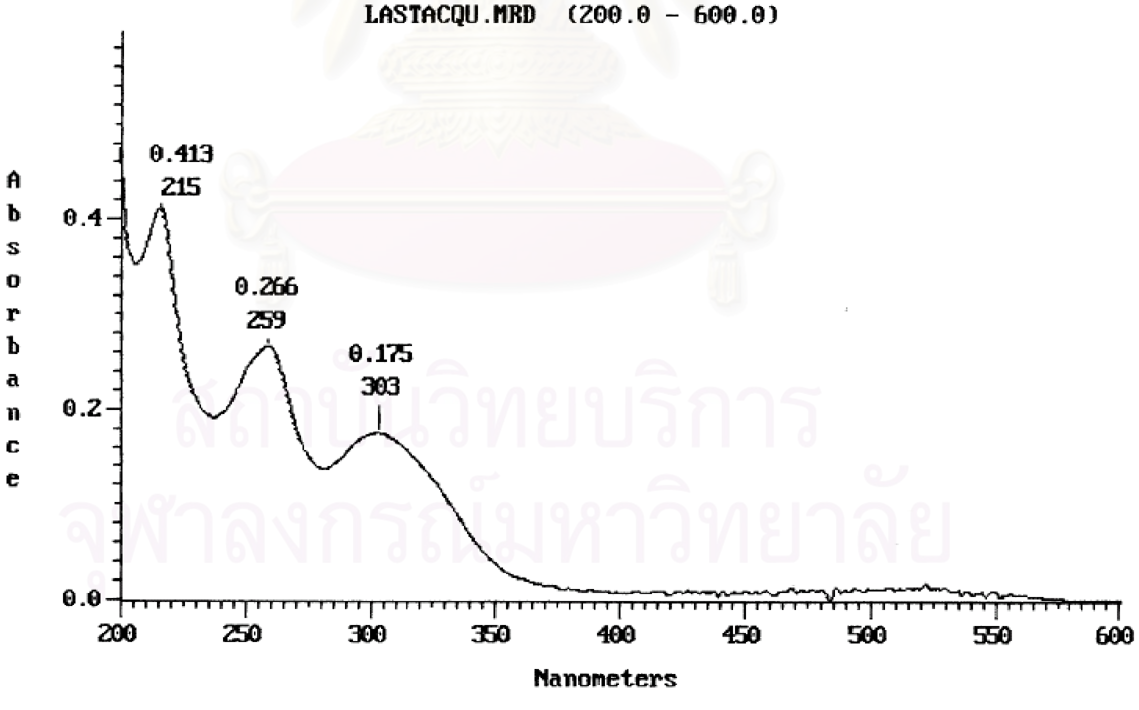

Figure 65 UV Spectrum of compound ME6 (methanol)

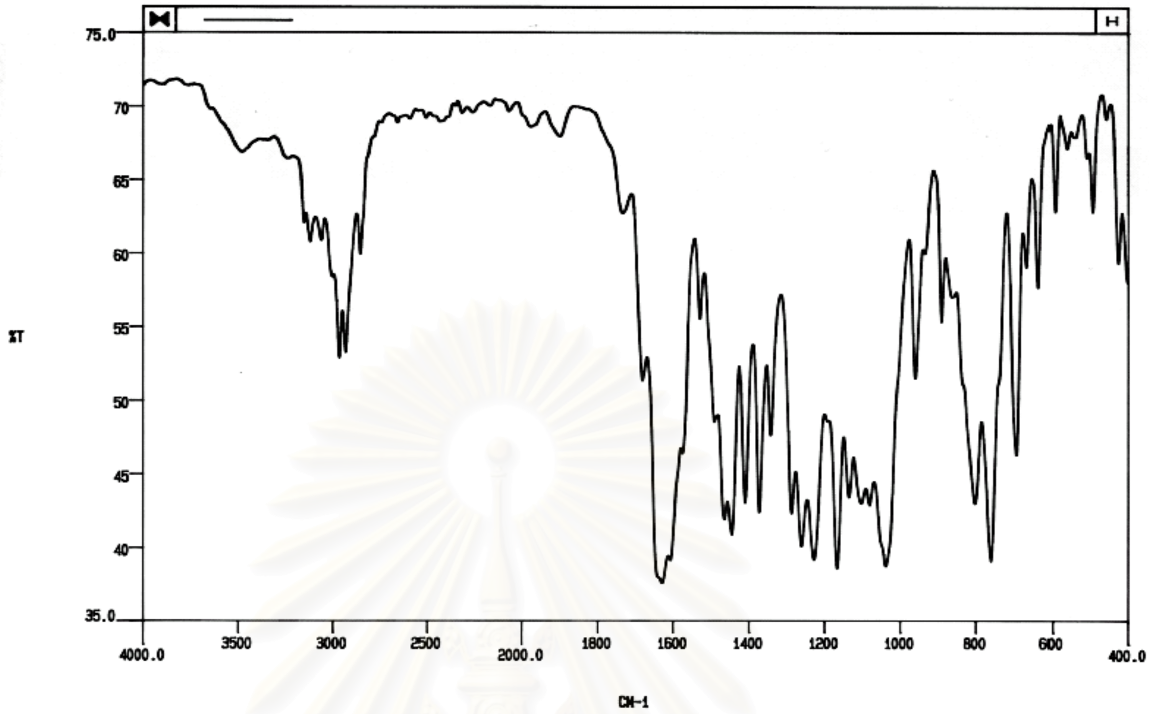

Figure 66 IR Spectrum of compound ME6 (KBr disc)

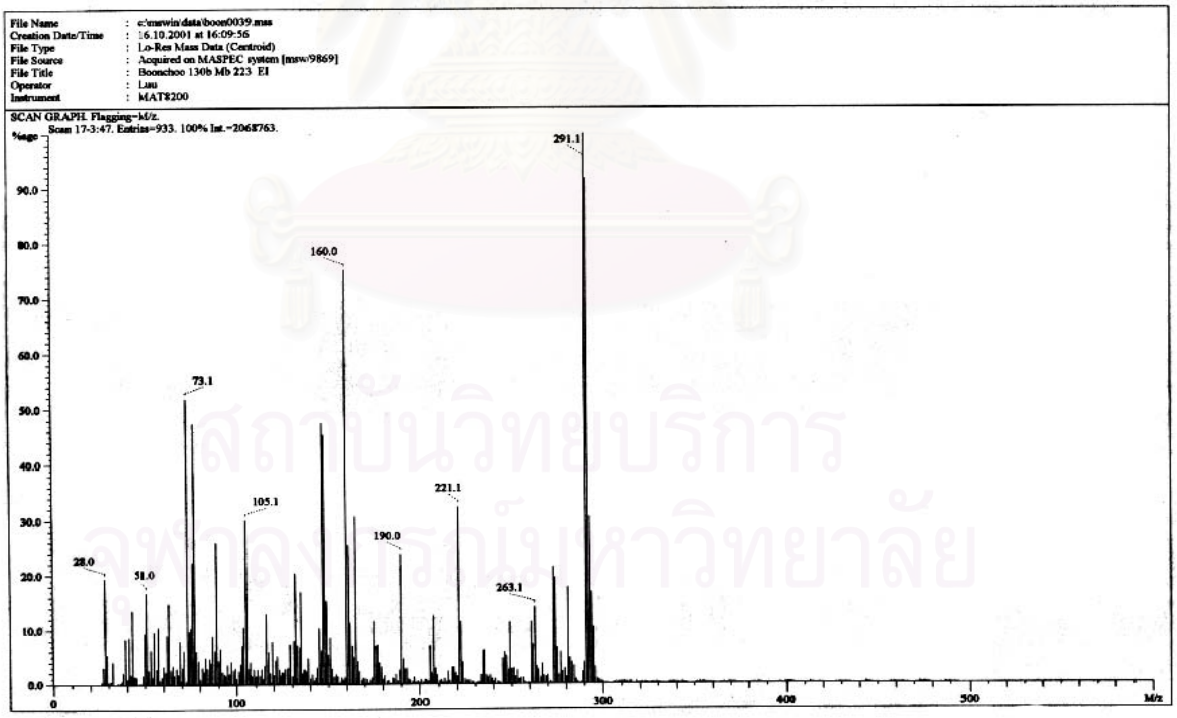

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Figure 67 EI Mass spectrum of compound ME6

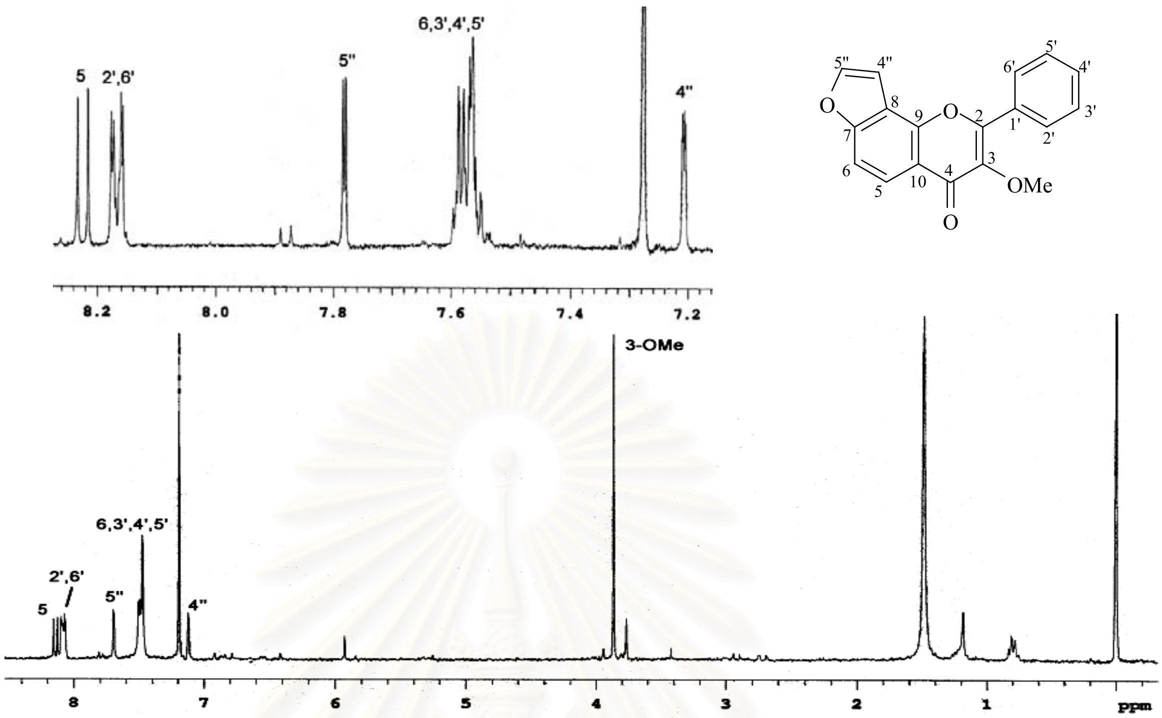

Figure 68 ^{1}H NMR (500 MHz) Spectrum of compound ME6 ($CDCl_3$)

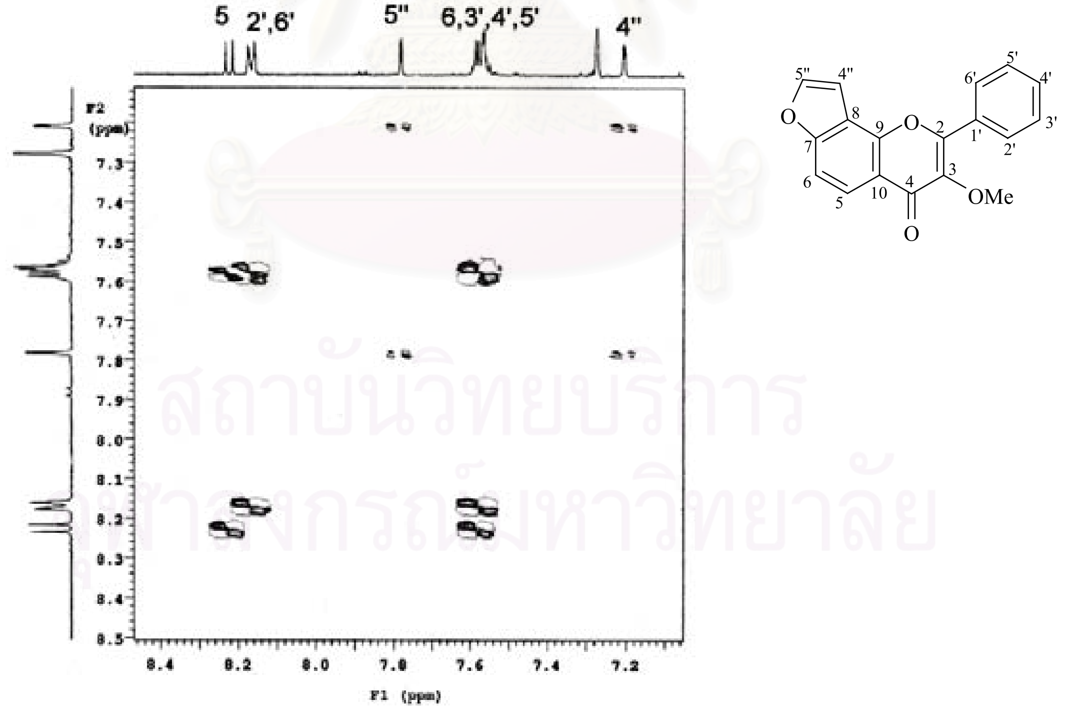

Figure 69 ^{1}H-^{1}H COSY Spectrum of compound ME6 ($CDCl_3$)

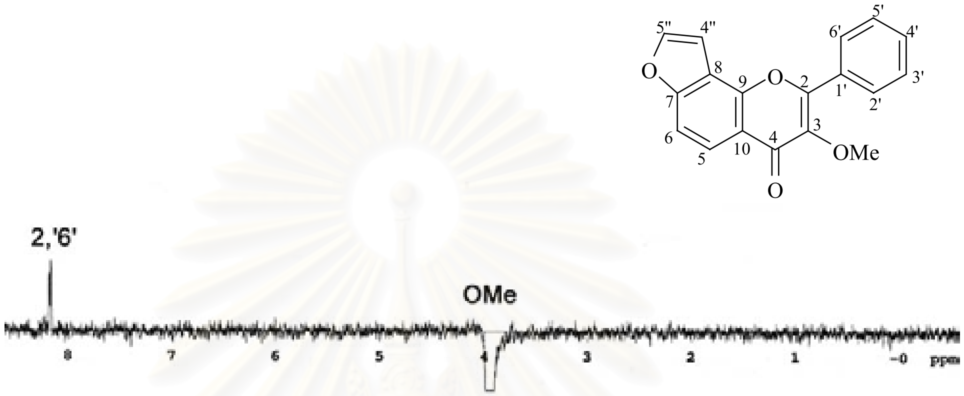

Figure 70 NOE Difference spectrum of compound ME6 ($CDCl_3$)

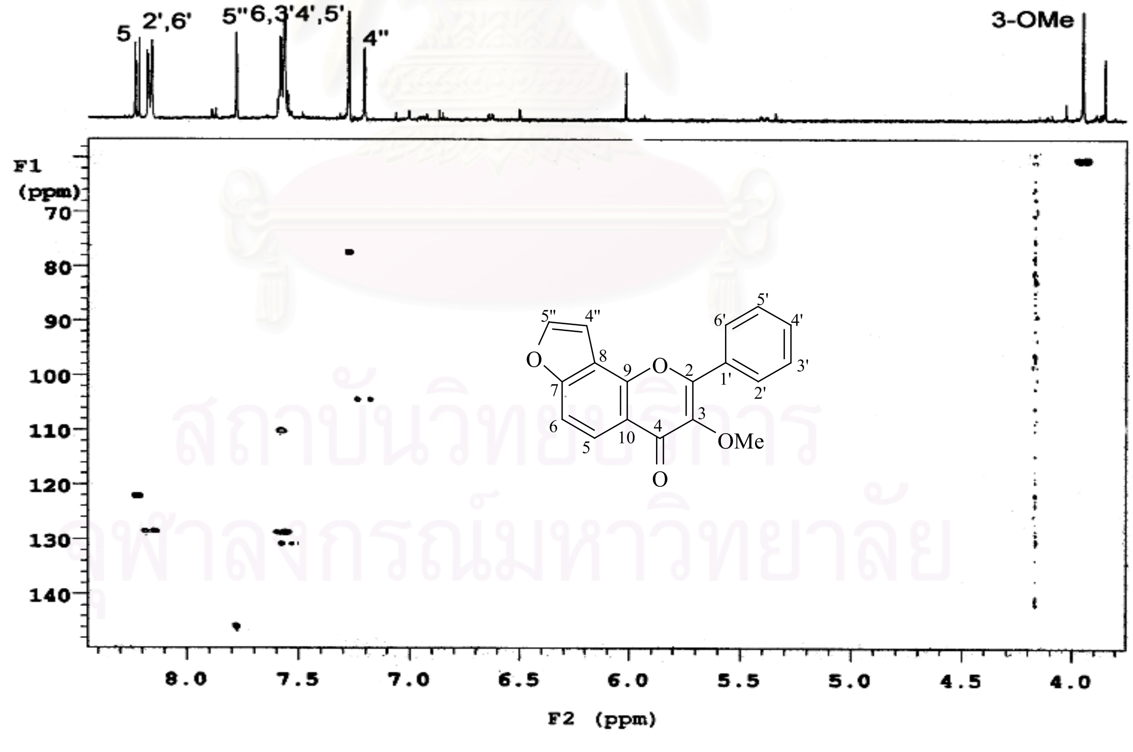

Figure 71 HSQC Spectrum of compound ME6 ($CDCl_3$)

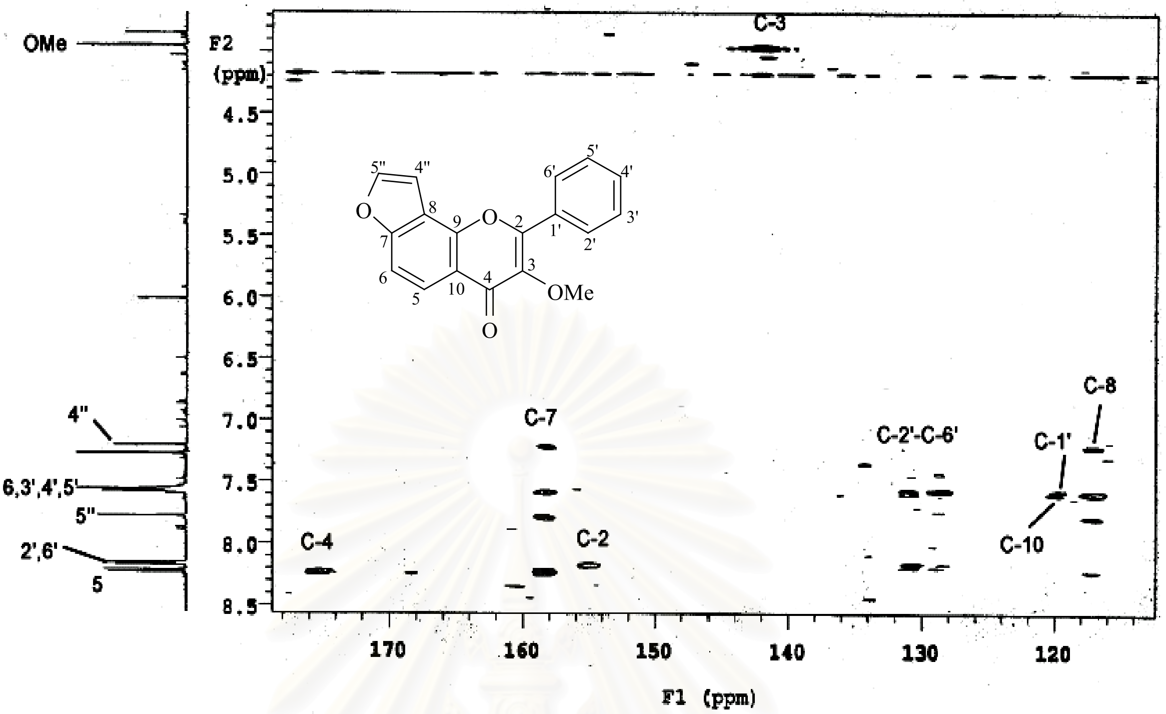

Figure 72 HMBC Spectrum of compound ME6 ($CDCl_3$) [δ_H 3.6-8.5 ppm, δ_C 112-180 ppm]

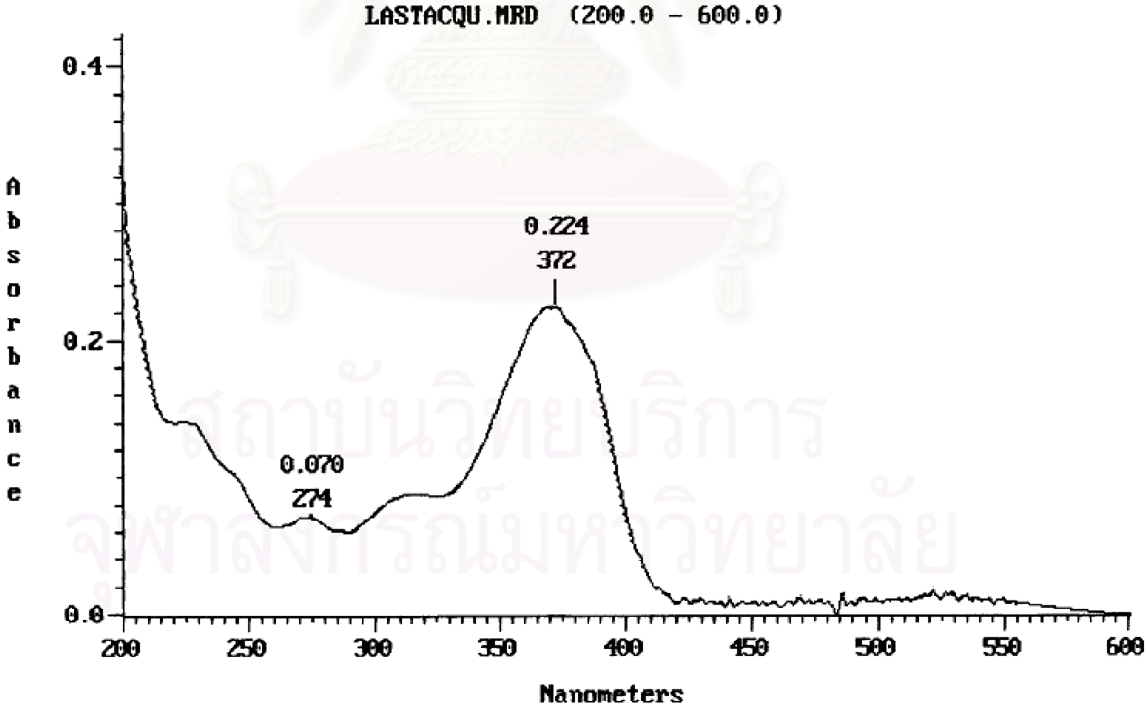

Figure 73 UV Spectrum of compound ME7 (methanol)

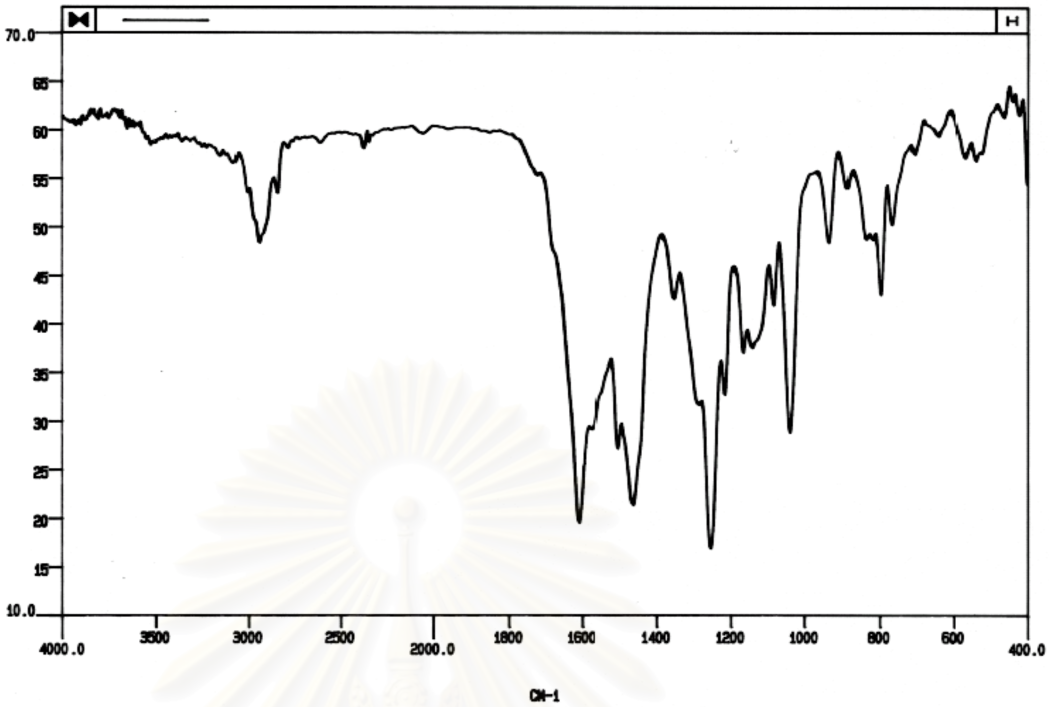

Figure 74 IR Spectrum of compound ME7 (KBr disc)

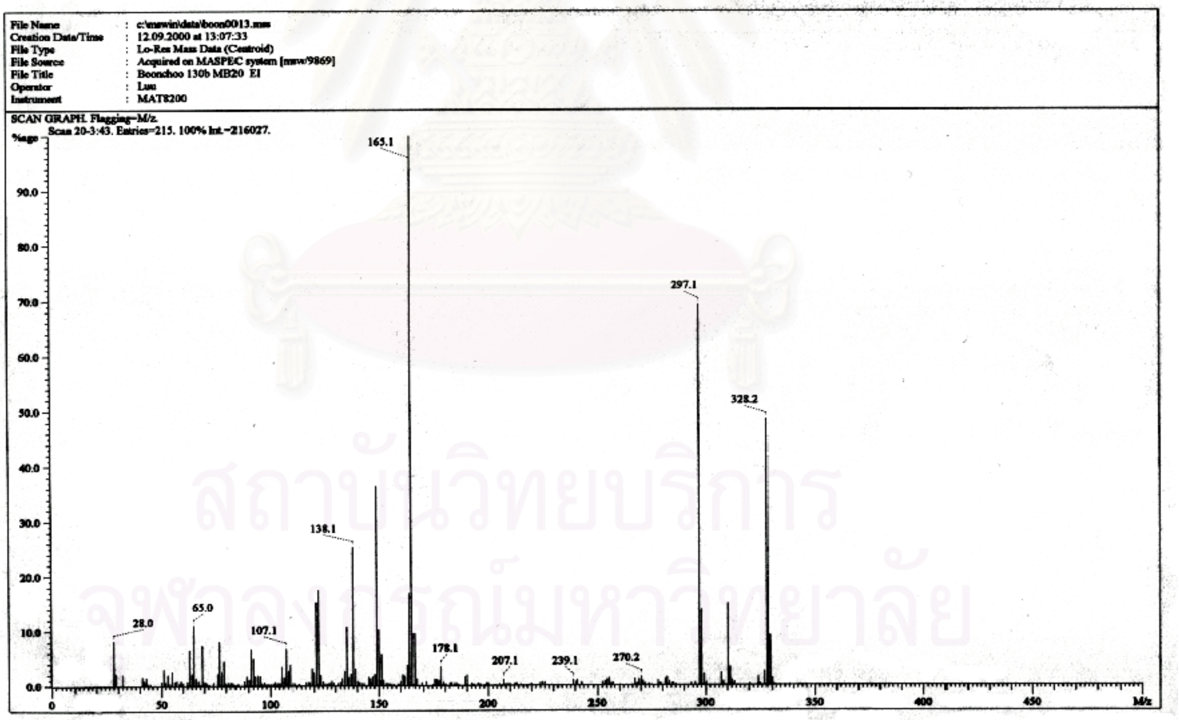
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Figure 75 EI Mass spectrum of compound ME7

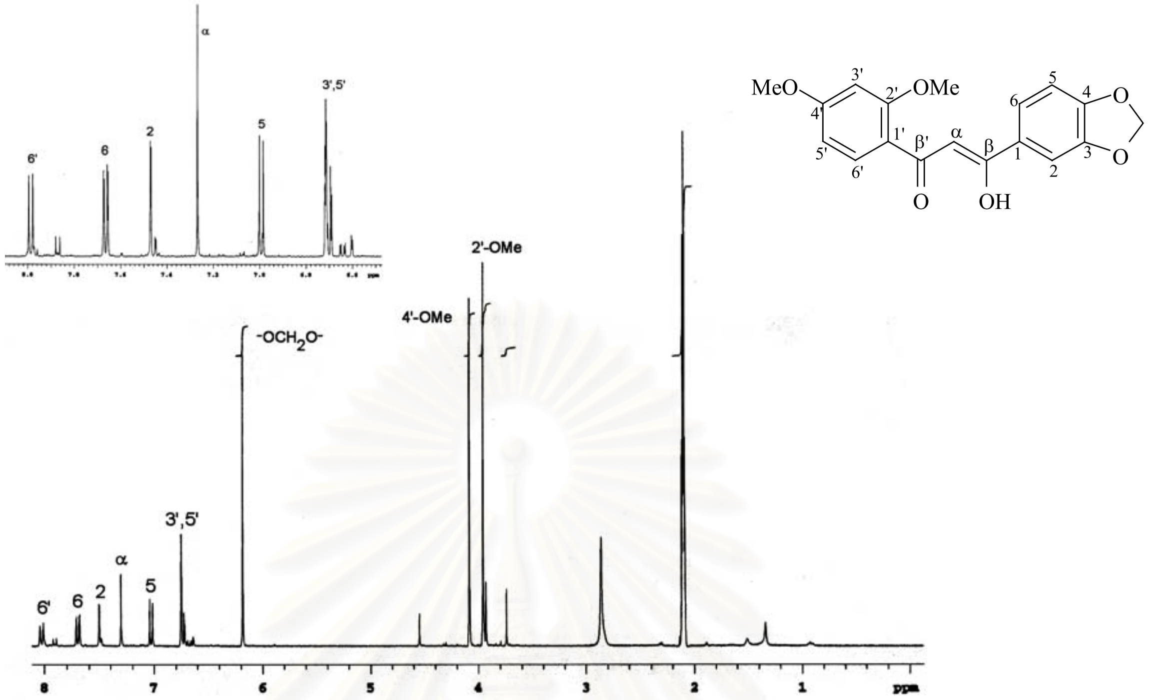

Figure 76 ^{1}H NMR (500 MHz) Spectrum of compound ME7 (acetone-d_6)

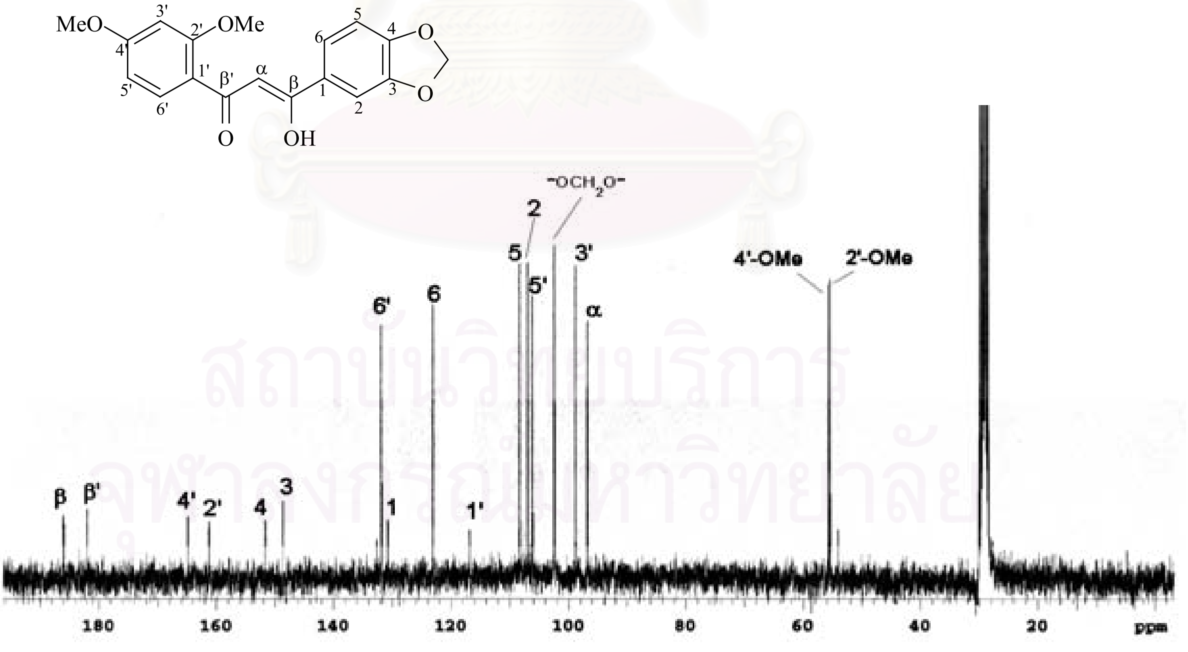

Figure 77 ^{13}C NMR (125 MHz) Spectrum of compound ME7 (acetone-d_6)

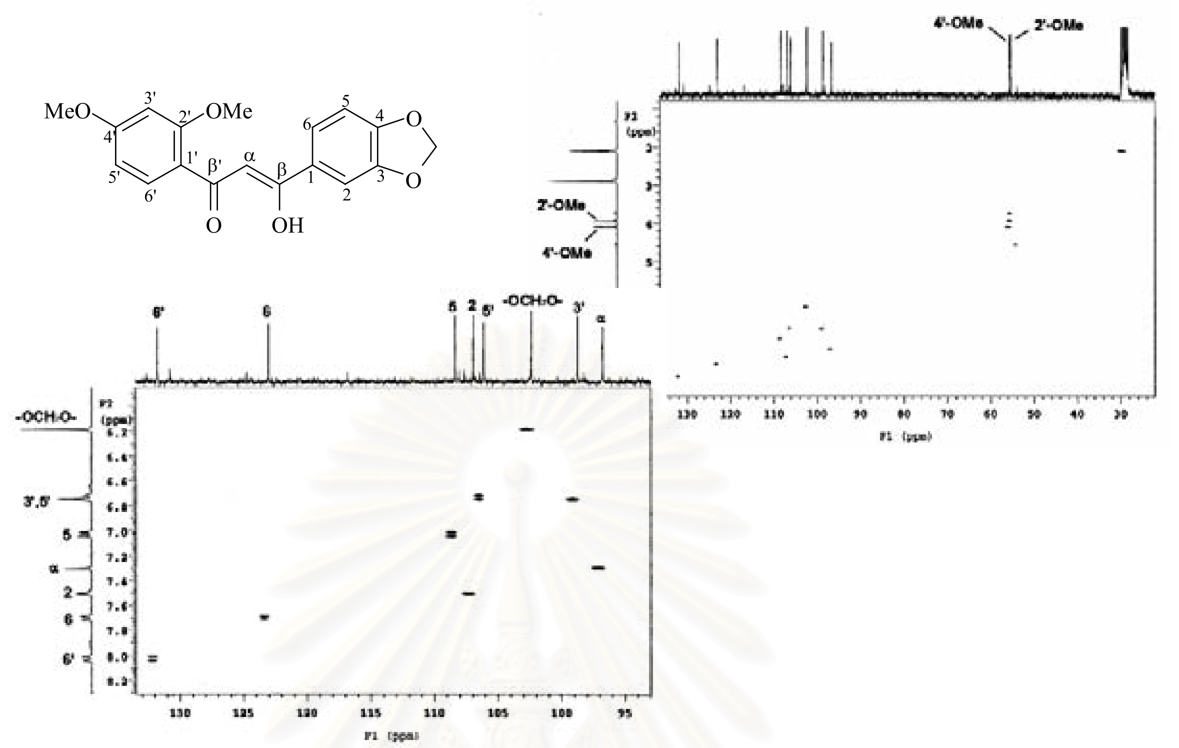

Figure 78 HSQC Spectrum of compound ME7 (acetone-d_6)

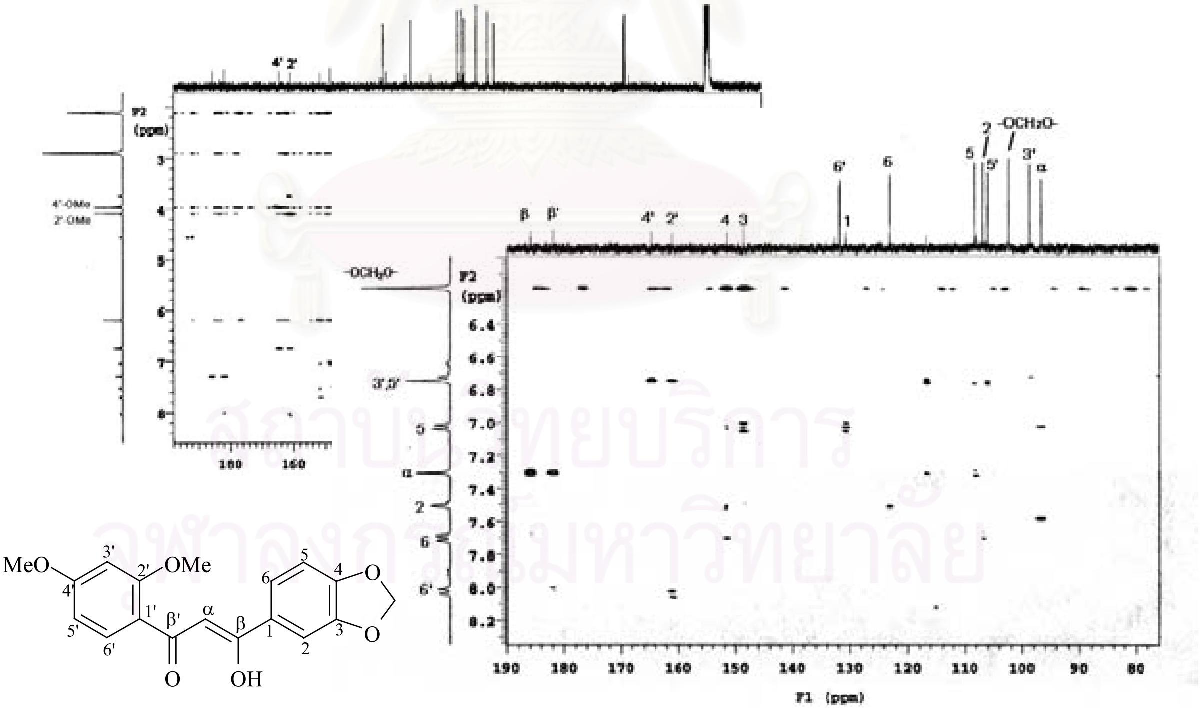

Figure 79 HMBC Spectrum of compound ME7 (acetone-d_6)

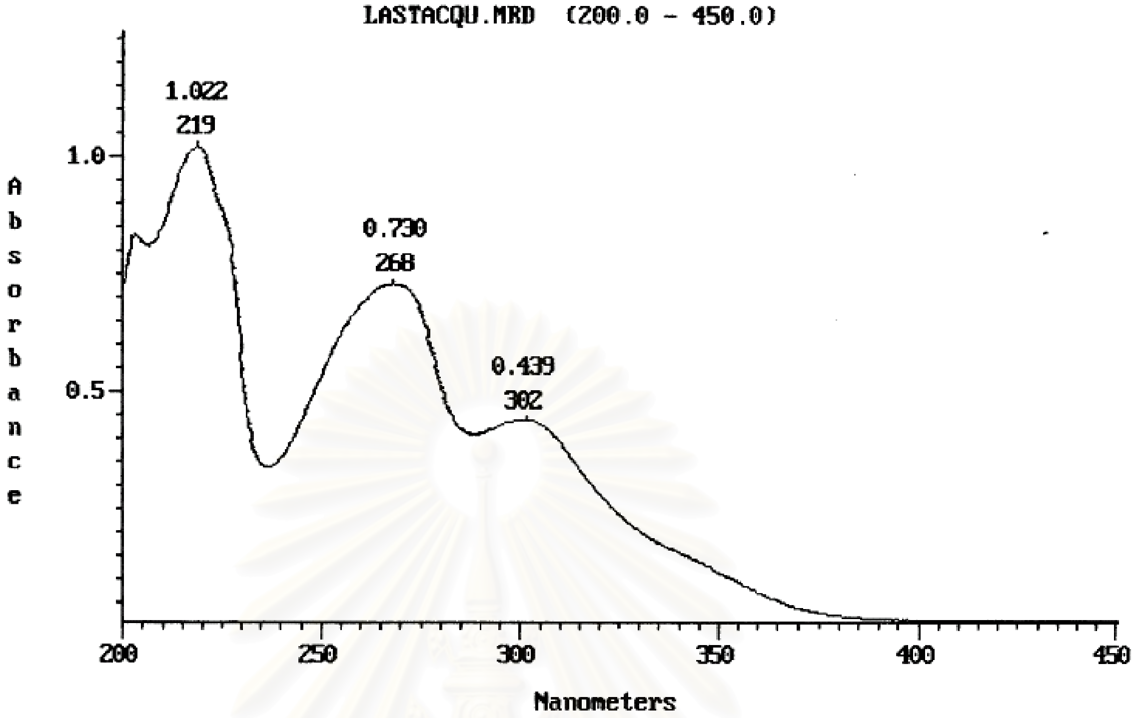

Figure 80 UV Spectrum of compound ME8 (methanol)

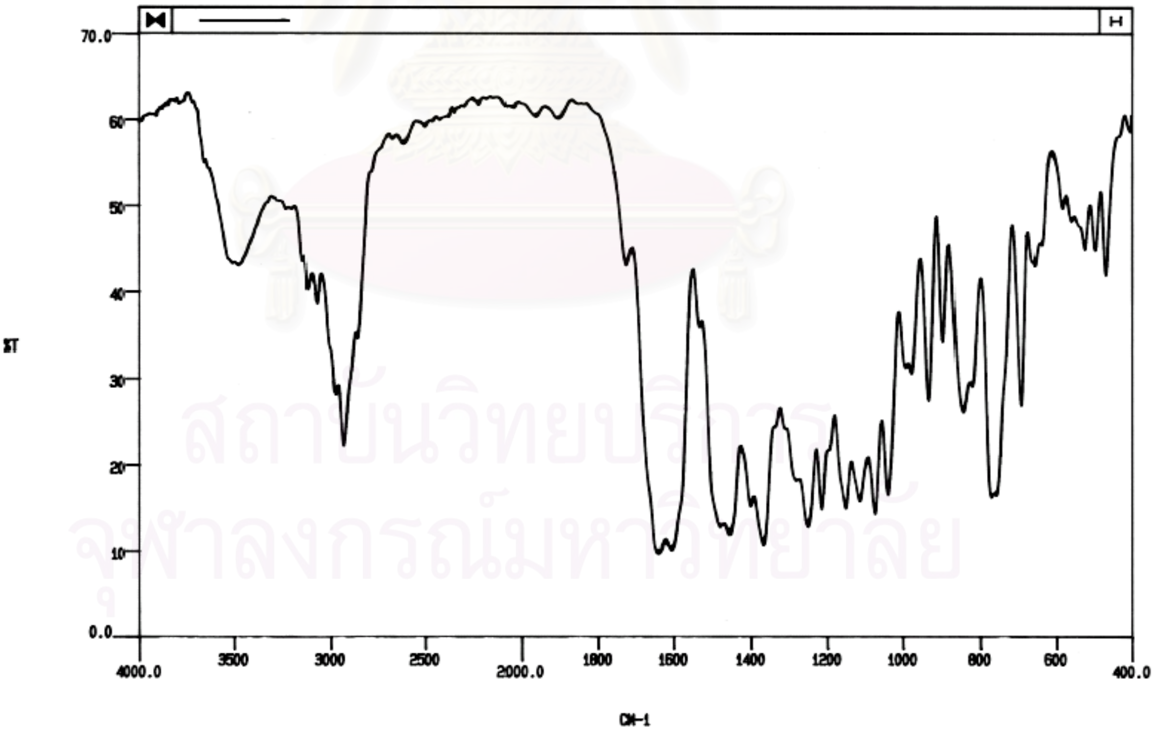

Figure 81 IR Spectrum of compound ME8 (KBr disc)

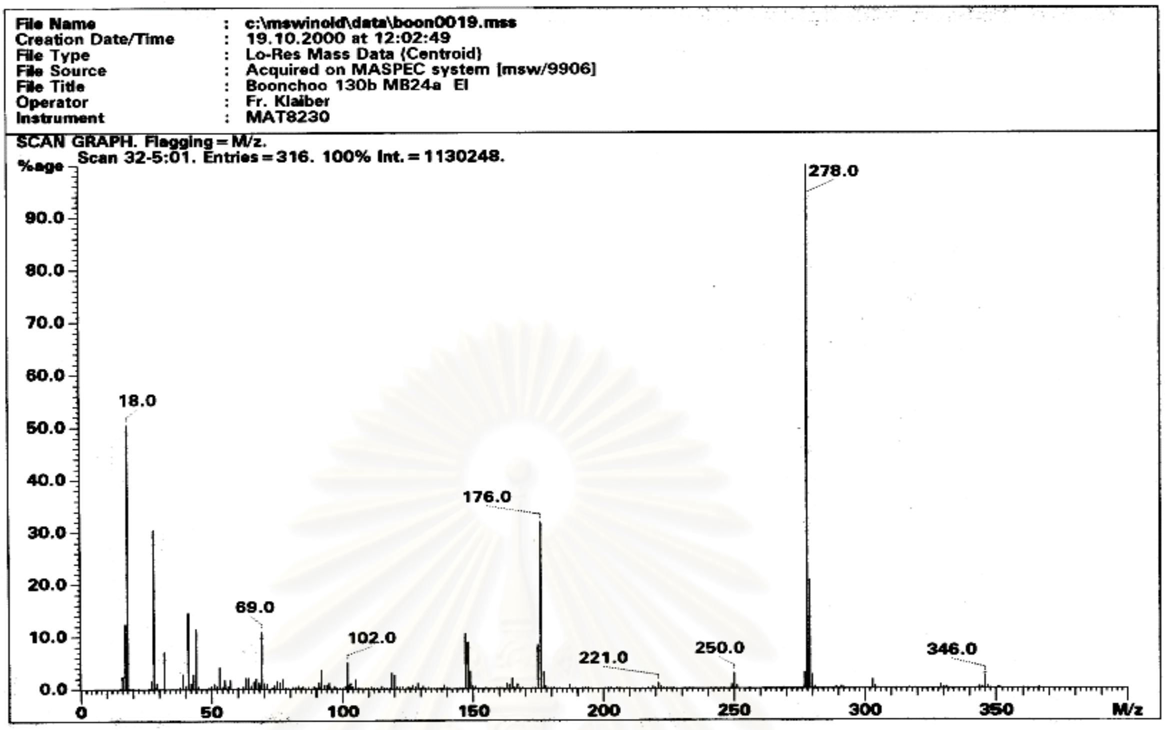

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Figure 82 EI Mass spectrum of compound ME8

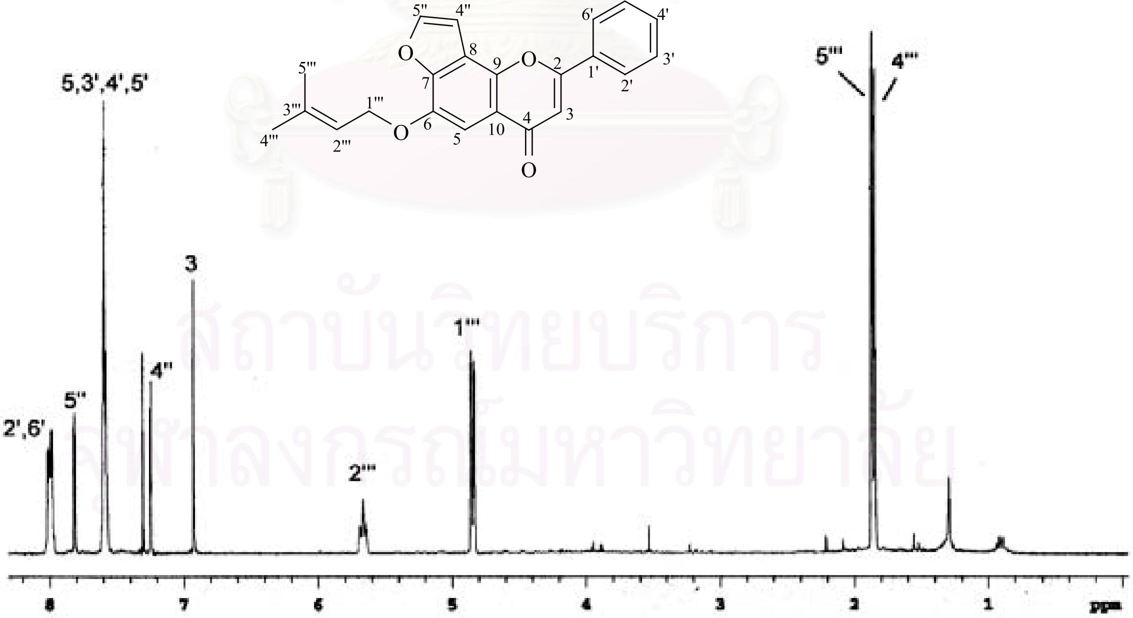

Figure 83 ^{1}H NMR (500 MHz) Spectrum of compound ME8 ($CDCl_3$)

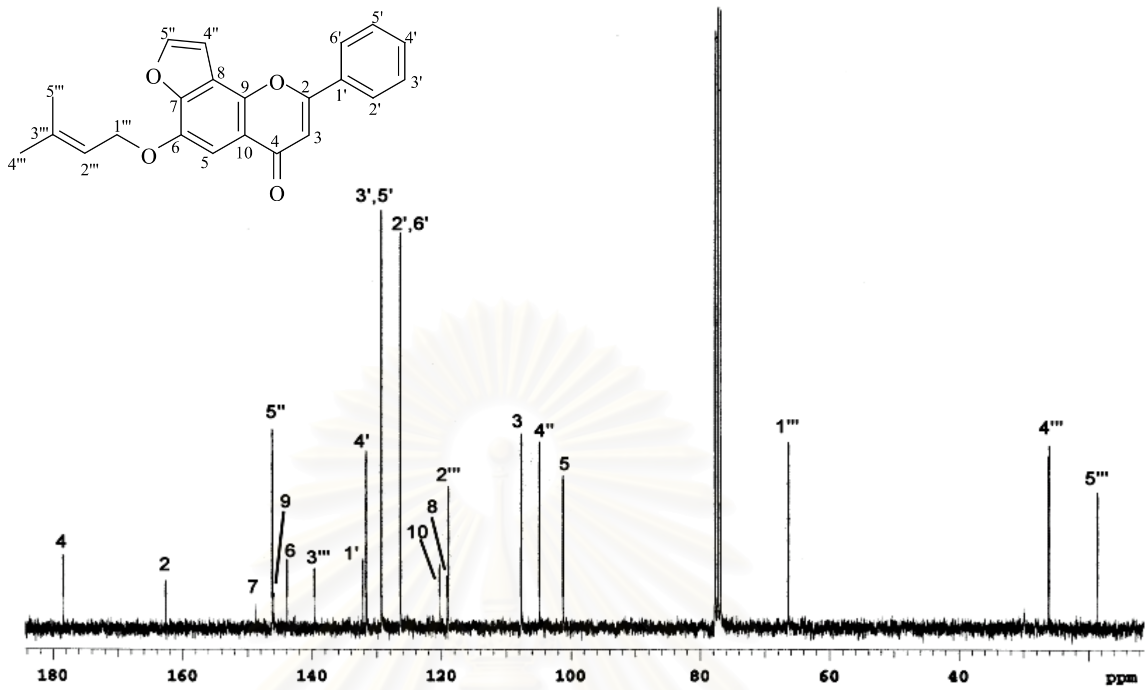

Figure 84 ^{13}C NMR (125 MHz) Spectrum of compound ME8 ($CDCl_3$)

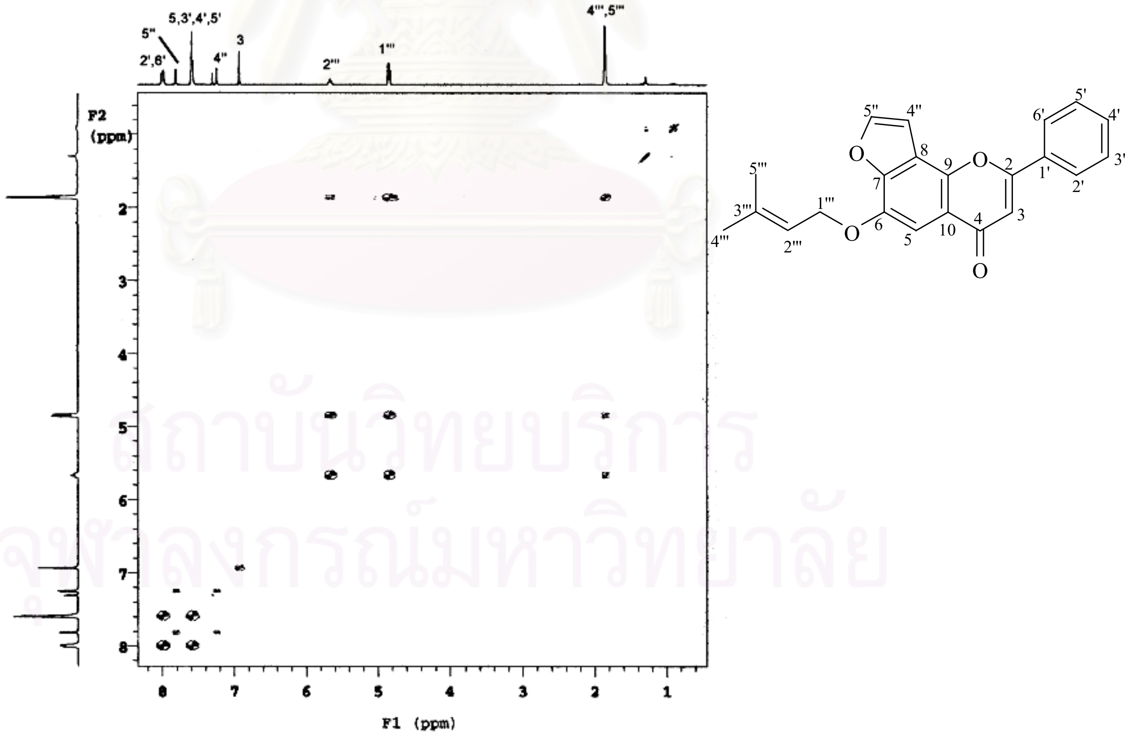

Figure 85 ^{1}H-^{1}H COSY Spectrum of compound ME8 ($CDCl_3$)

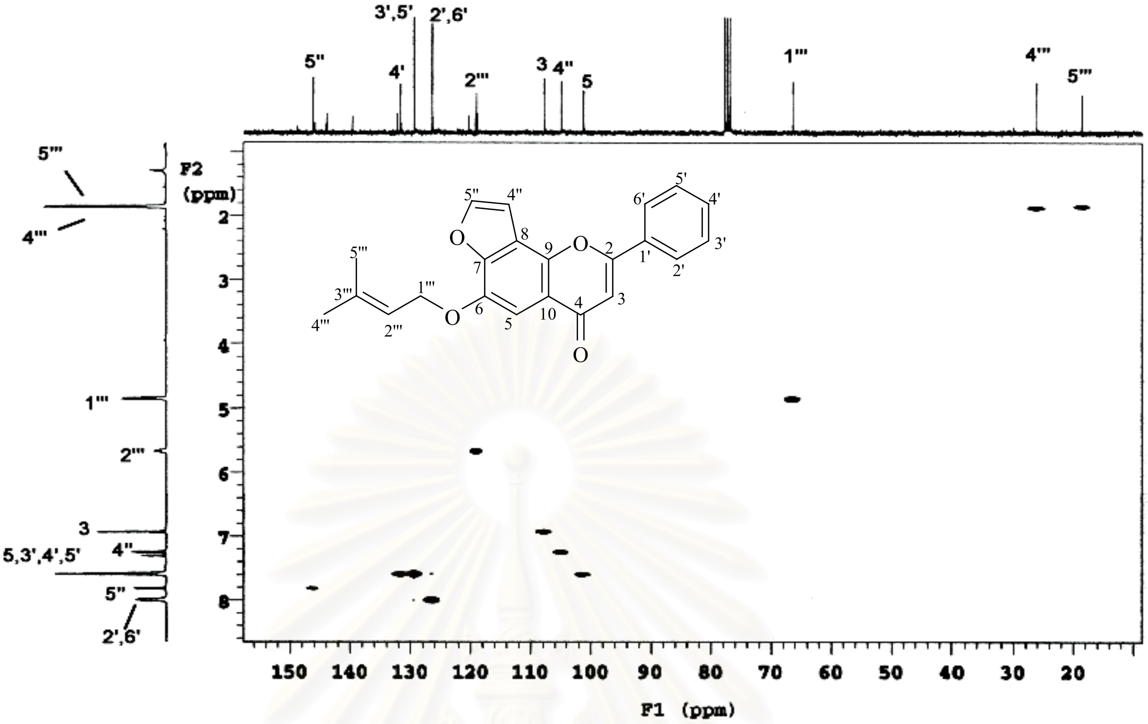

Figure 86 HSQC Spectrum of compound ME8 ($CDCl_3$)

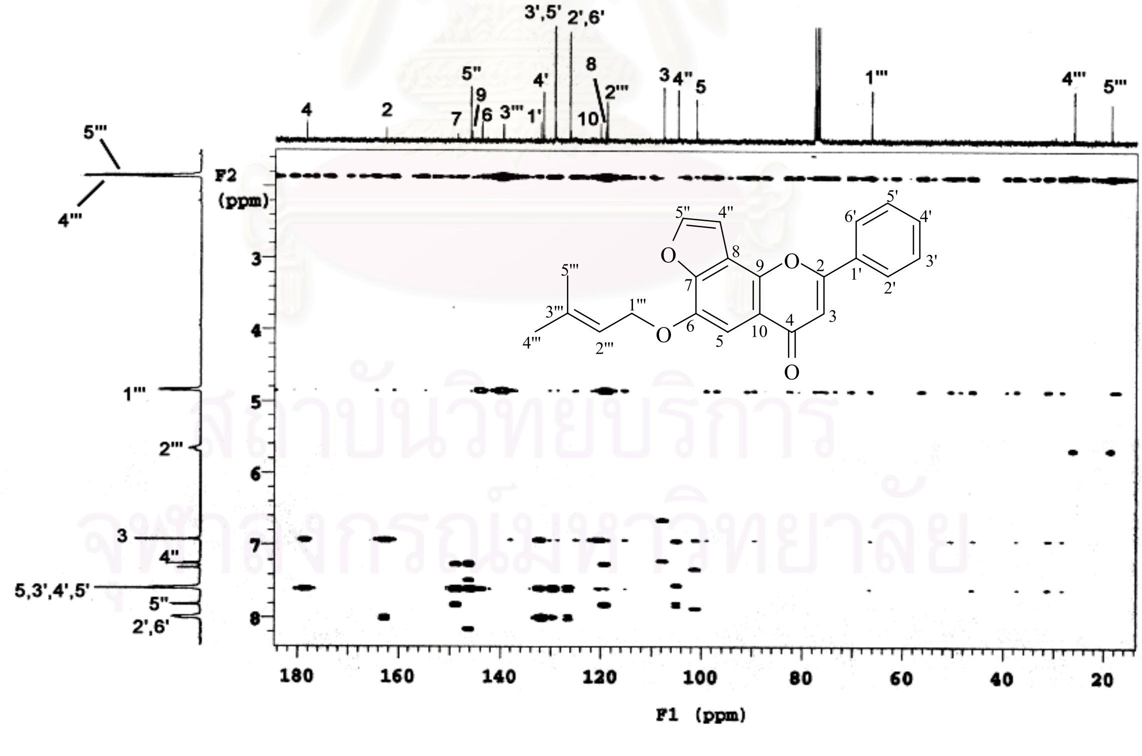

Figure 87 HMBC Spectrum of compound ME8 ($CDCl_3$)

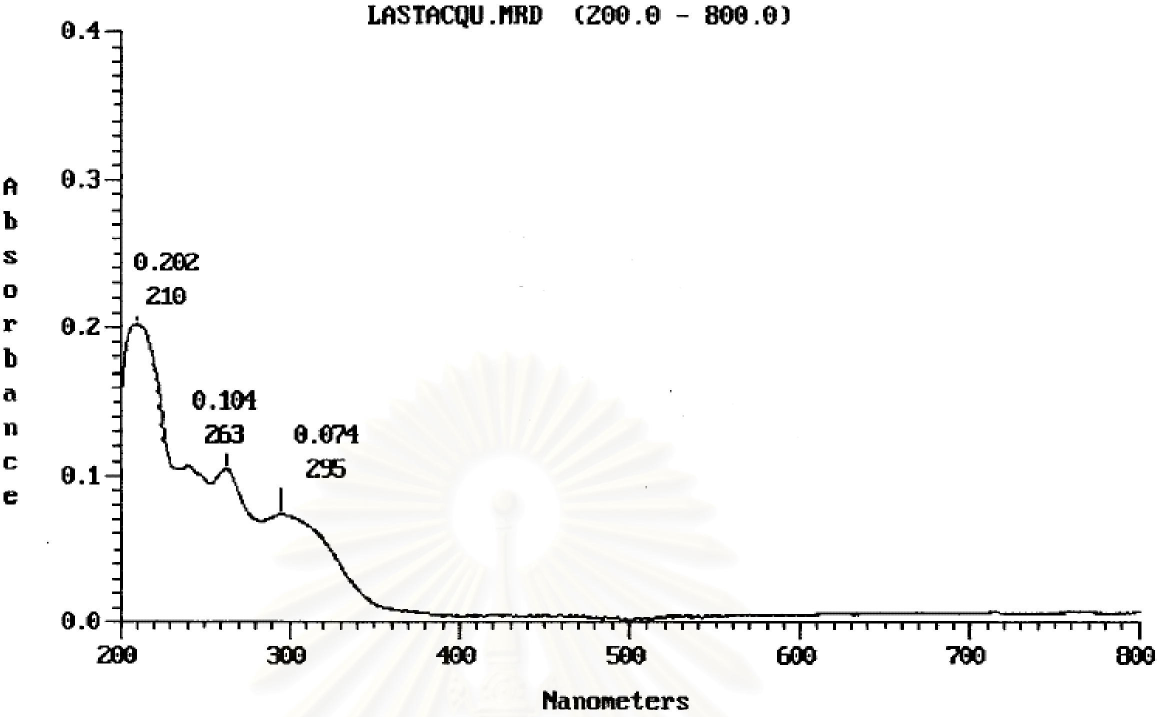

Figure 88 UV Spectrum of compound ME9 (methanol)

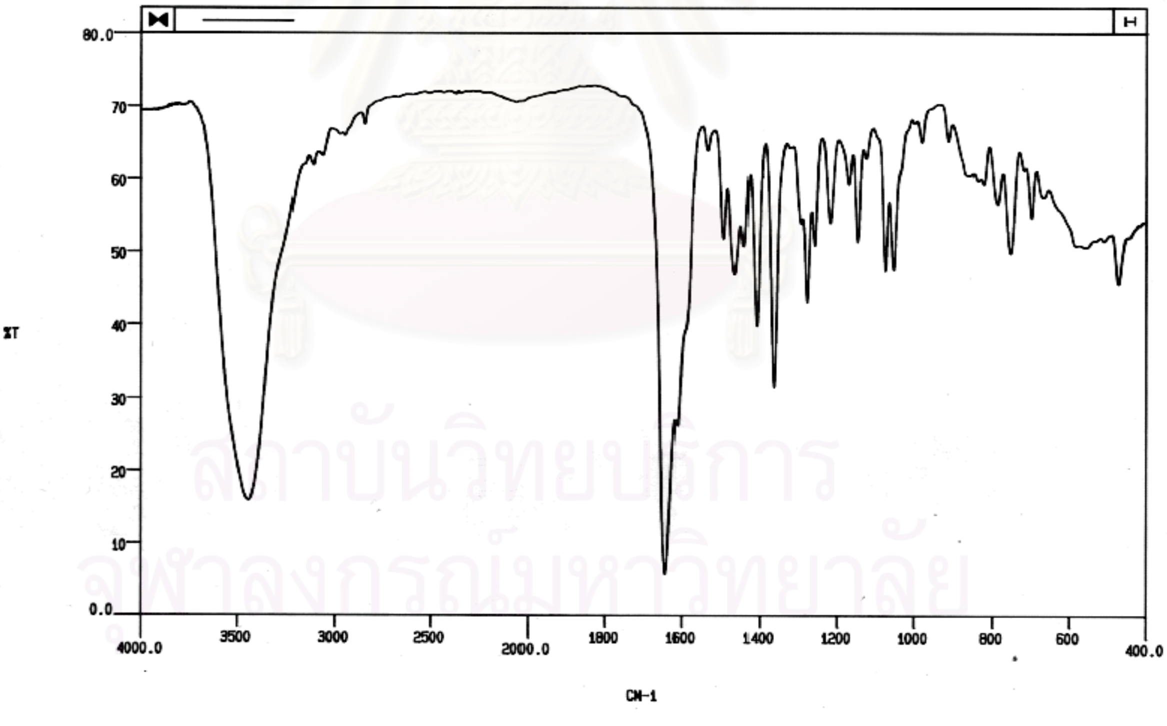

Figure 89 IR Spectrum of compound ME9 (KBr disc)

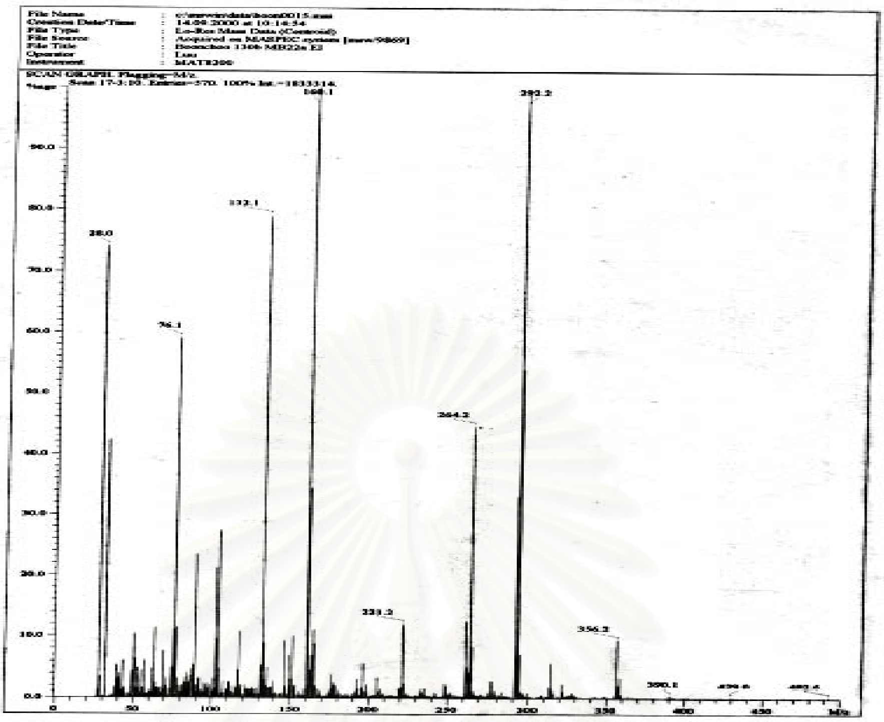

Figure 90 EI Mass spectrum of compound ME9

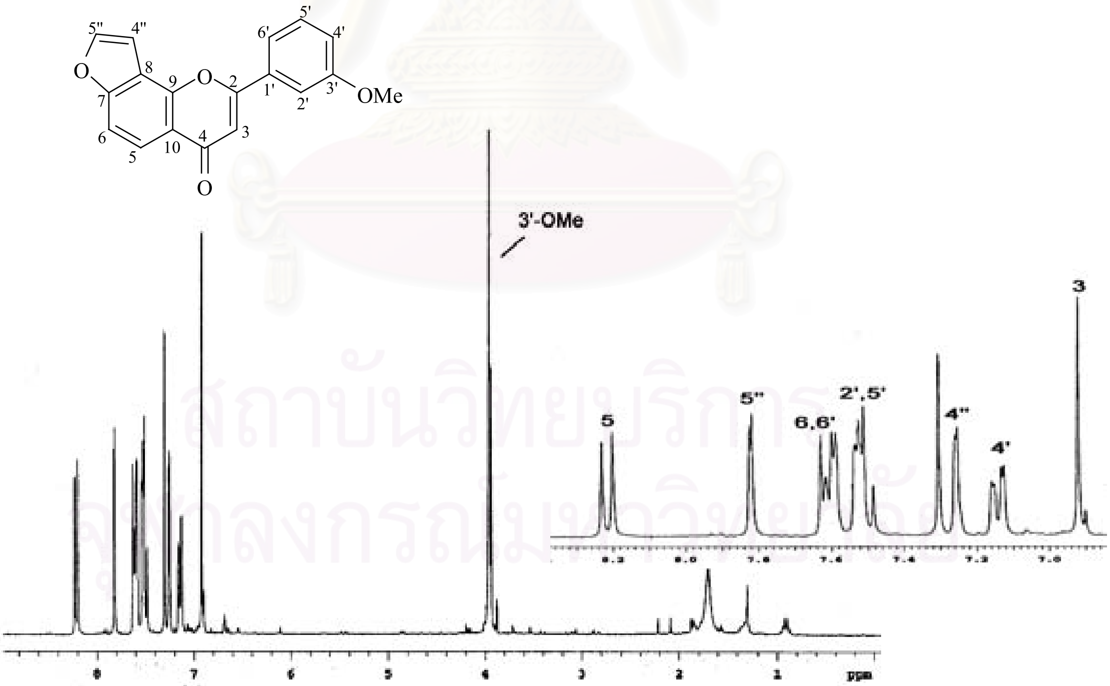

Figure 91 ^{1}H NMR (300 MHz) Spectrum of compound ME9 ($CDCl_3$)

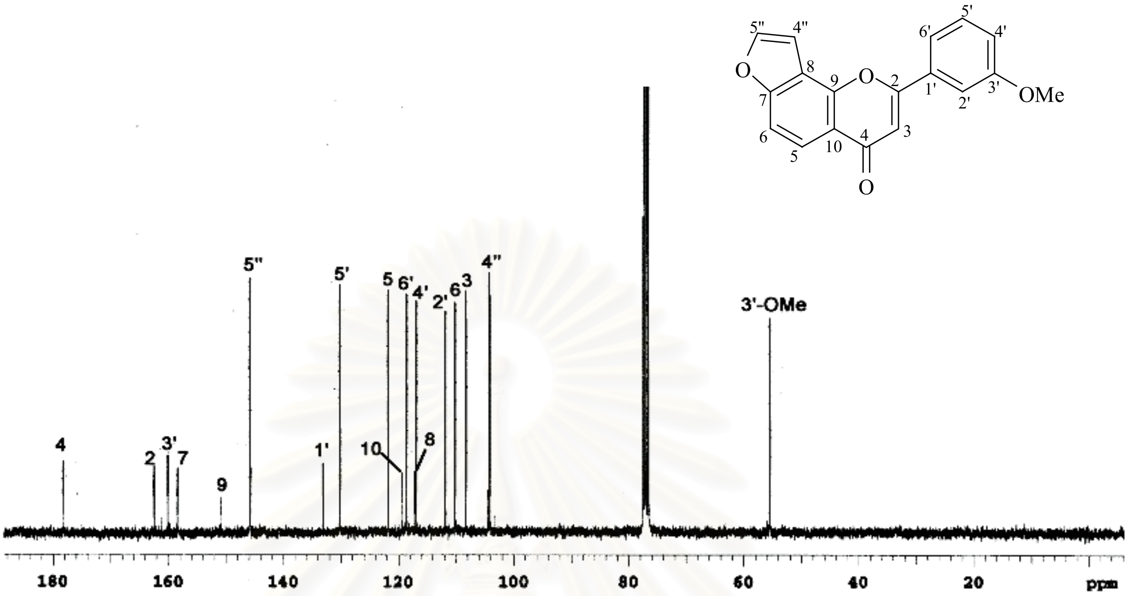

Figure 92 ^{13}C NMR (75 MHz) Spectrum of compound ME9 ($CDCl_3$)

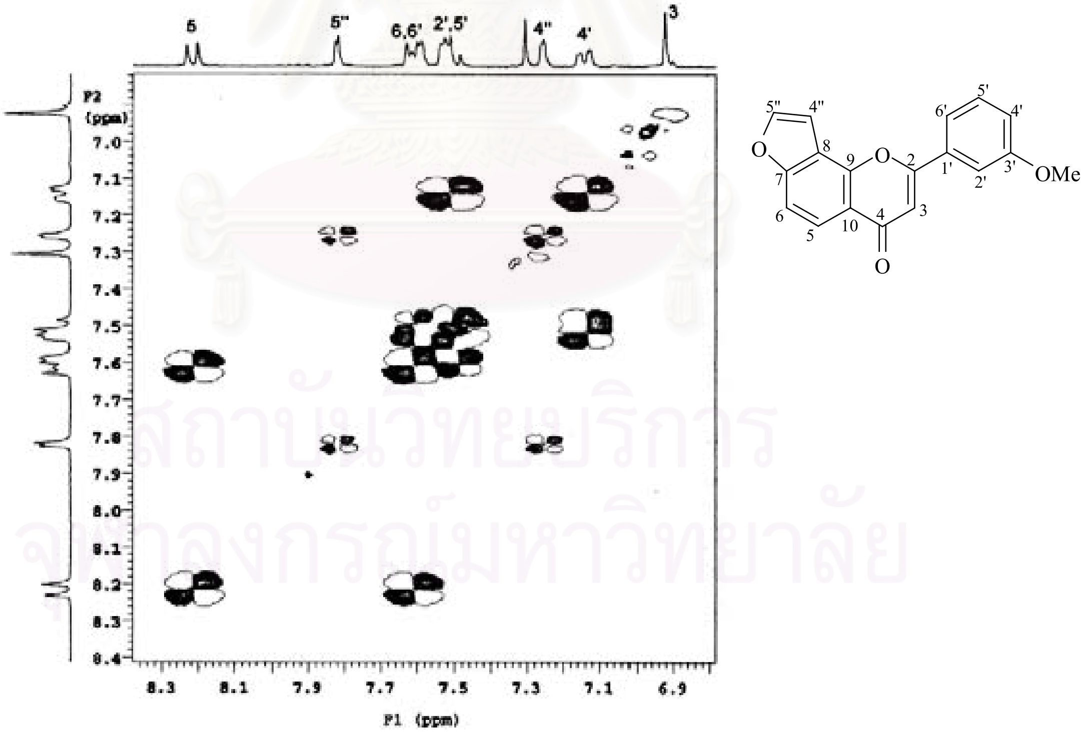

Figure 93 ^{1}H-^{1}H COSY Spectrum of compound ME9 ($CDCl_3$)

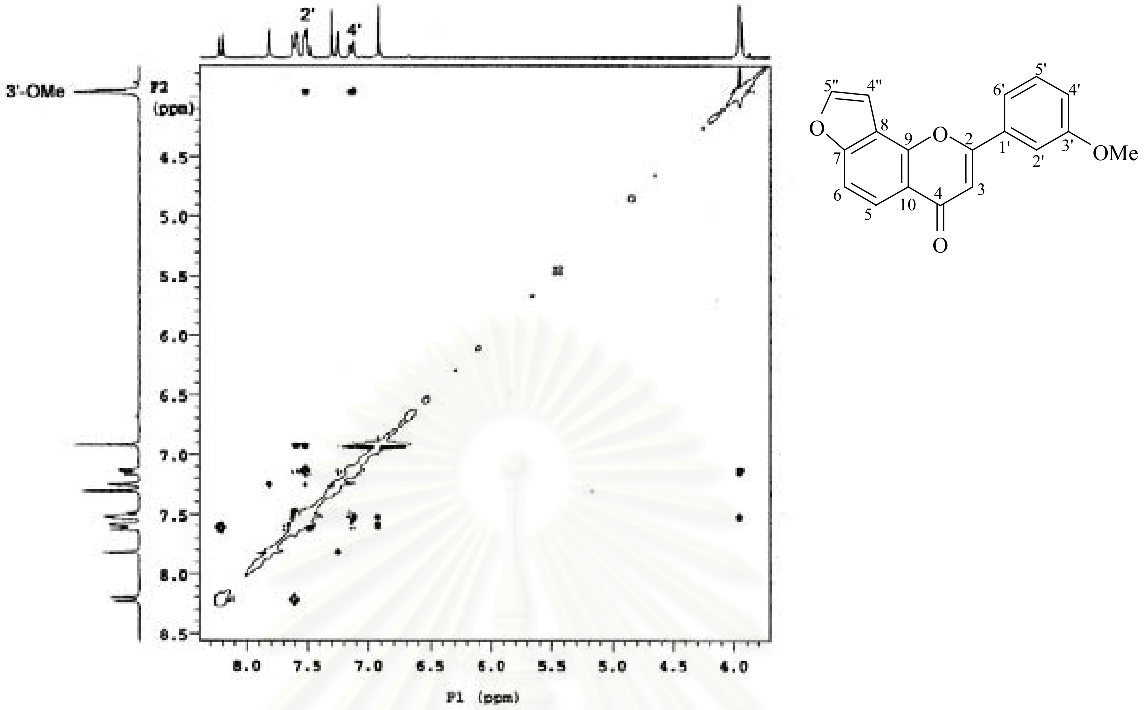

Figure 94 NOESY Spectrum of compound ME9 ($CDCl_3$)

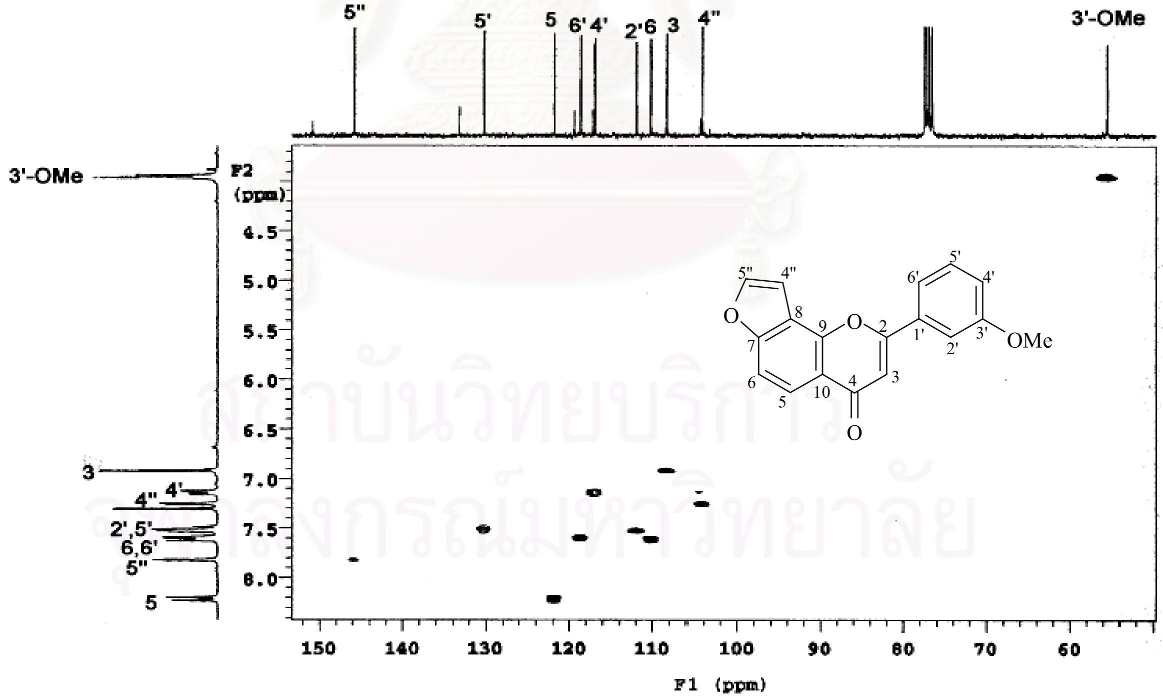

Figure 95 HSQC Spectrum of compound ME9 ($CDCl_3$)

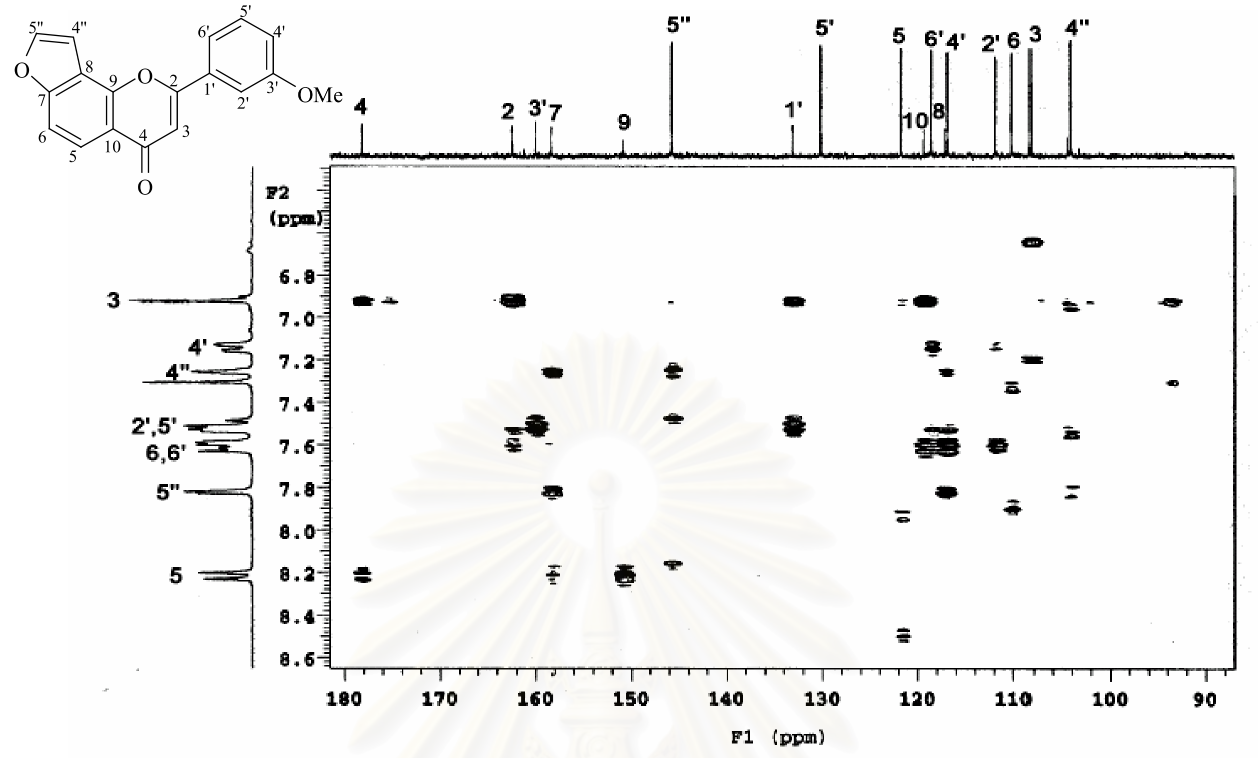

Figure 96 HMBC Spectrum of compound ME9 ($CDCl_3$)

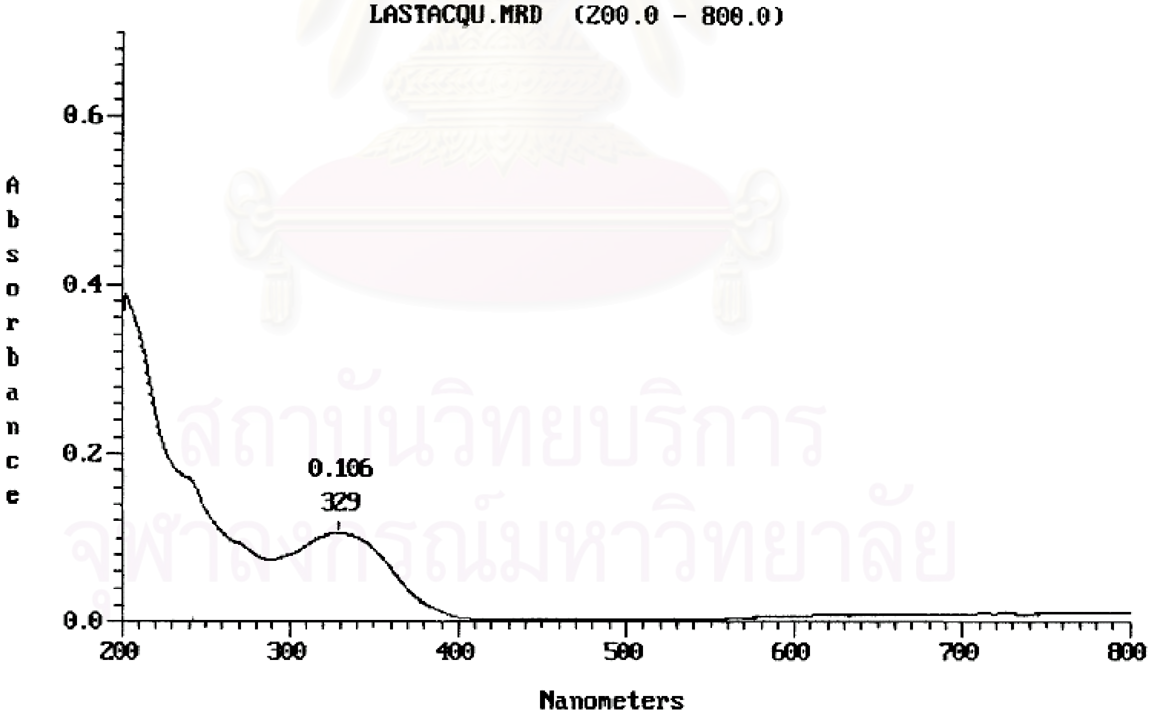

Figure 97 UV Spectrum of compound ME10 (methanol)

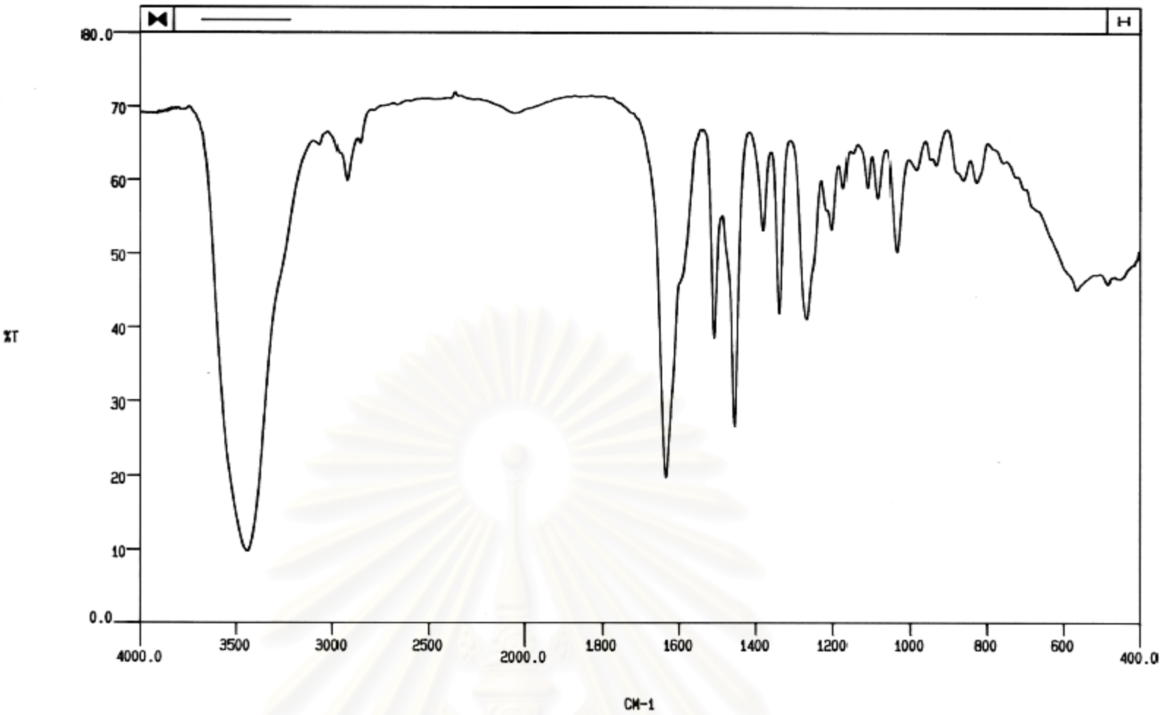

Figure 98 IR Spectrum of compound ME10 (KBr disc)

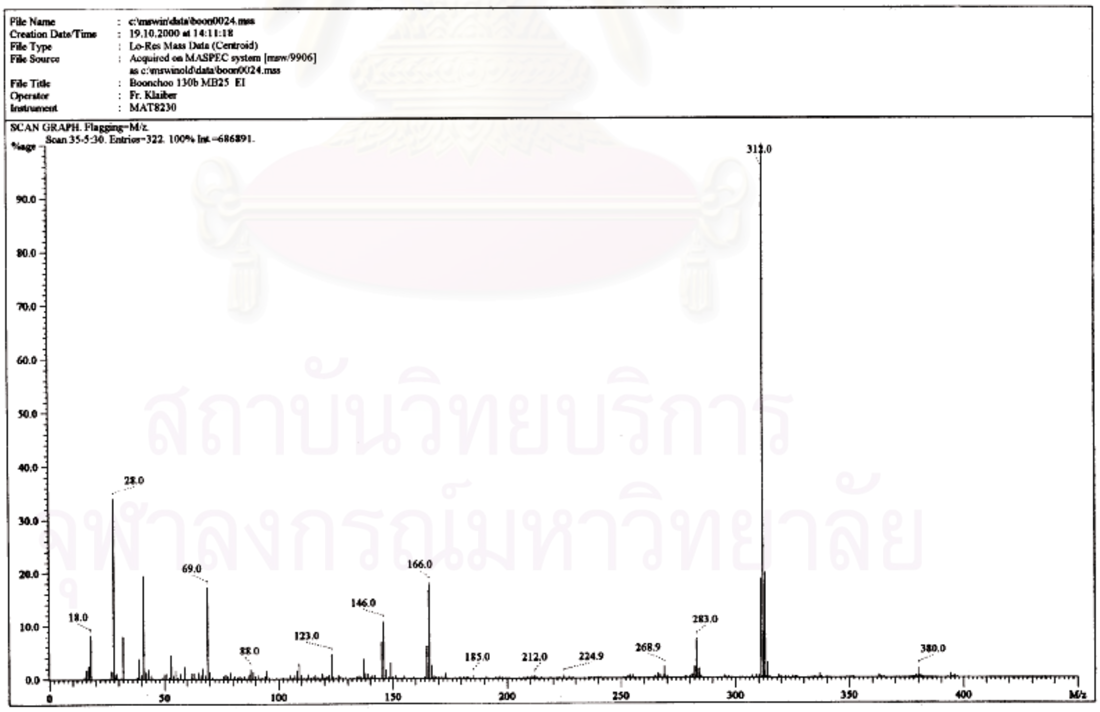

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Figure 99 EI Mass spectrum of compound ME10

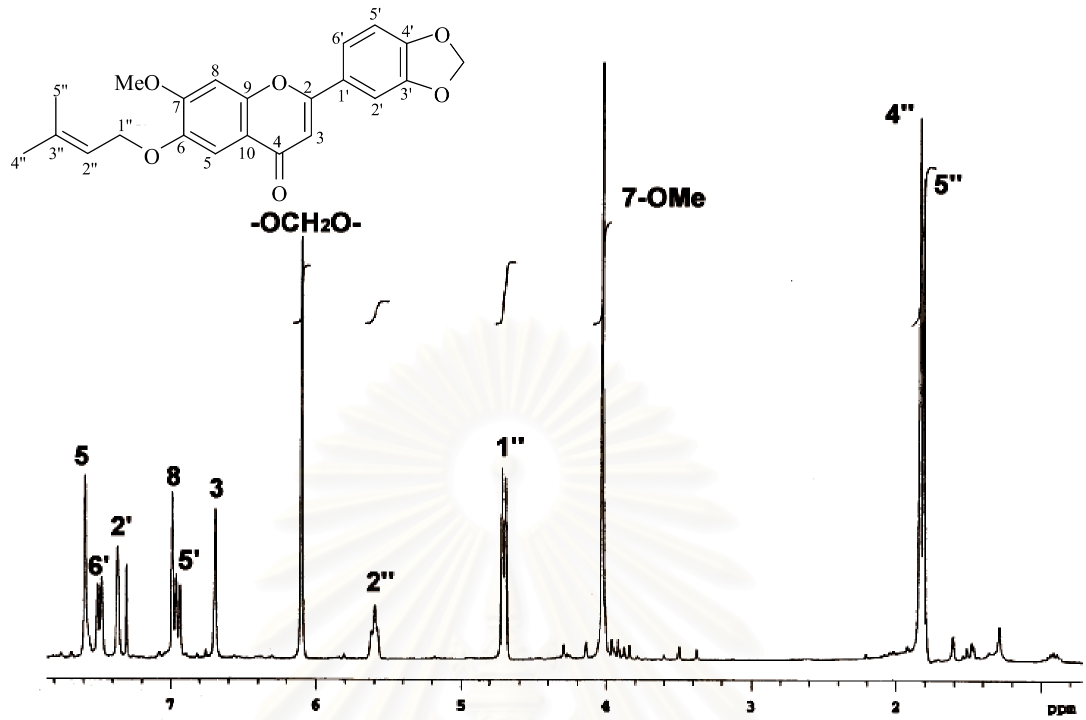

Figure 100 ^{1}H NMR (300 MHz) Spectrum of compound ME10 ($CDCl_3$)

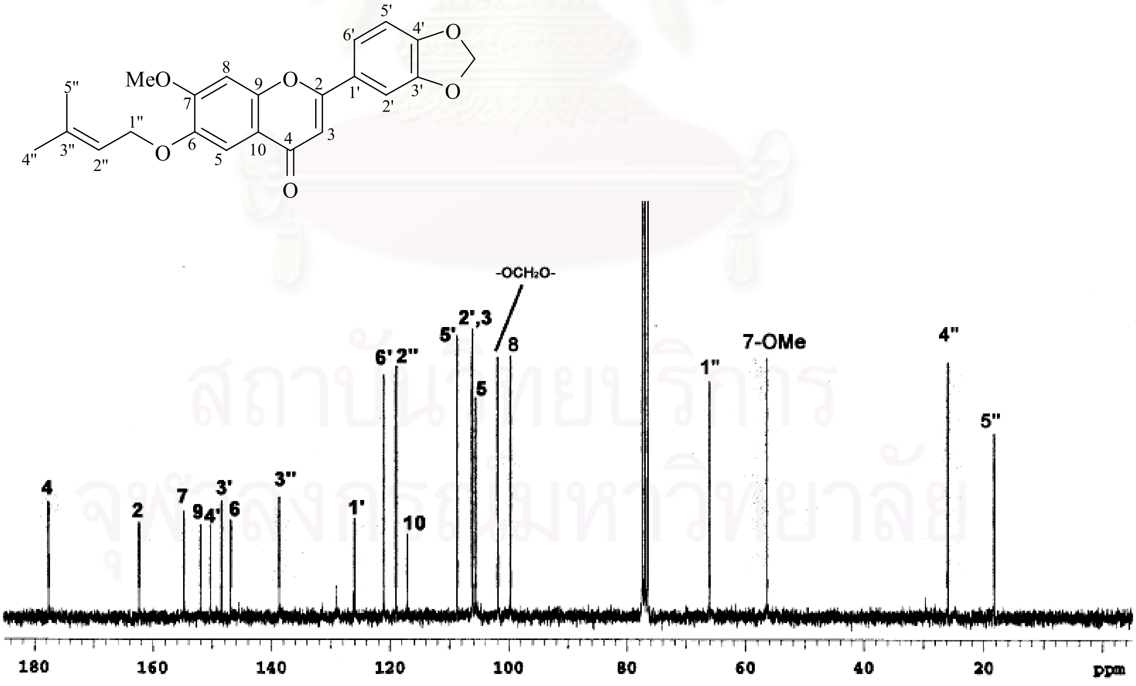

Figure 101 ^{13}C NMR (75 MHz) Spectrum of compound ME10 ($CDCl_3$)

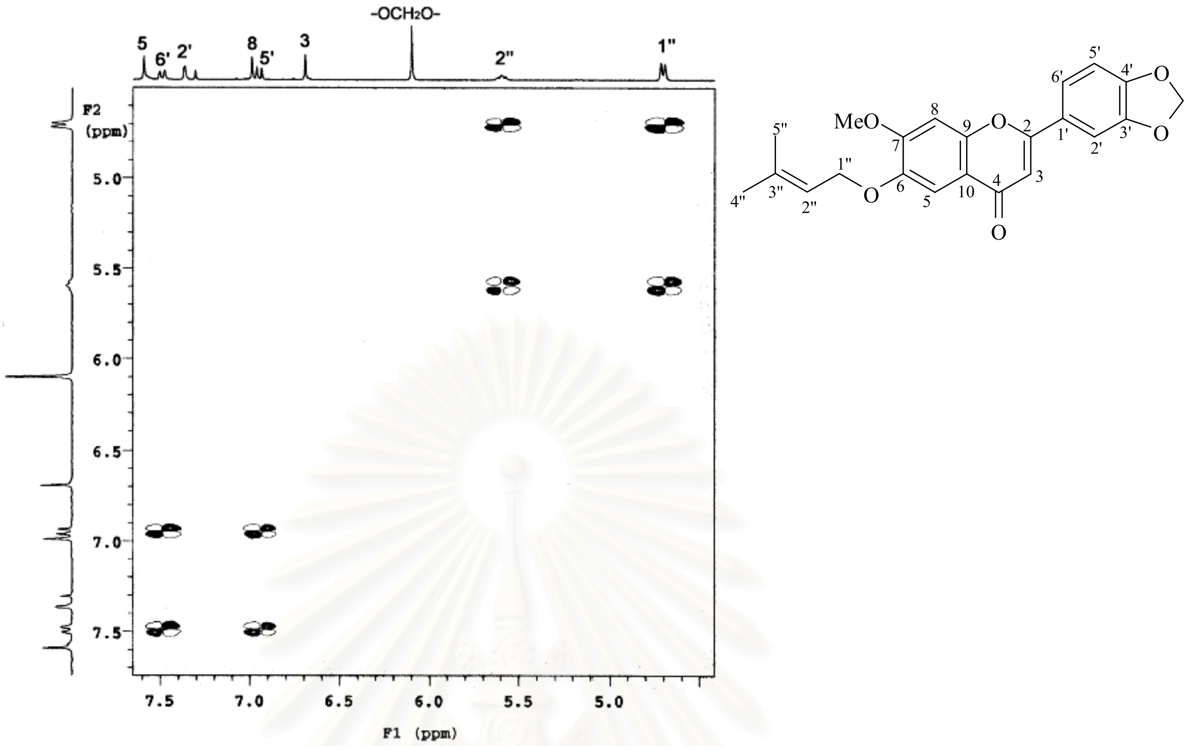

Figure 102 ^1H-^1H COSY Spectrum of compound ME10 ($CDCl_3$)

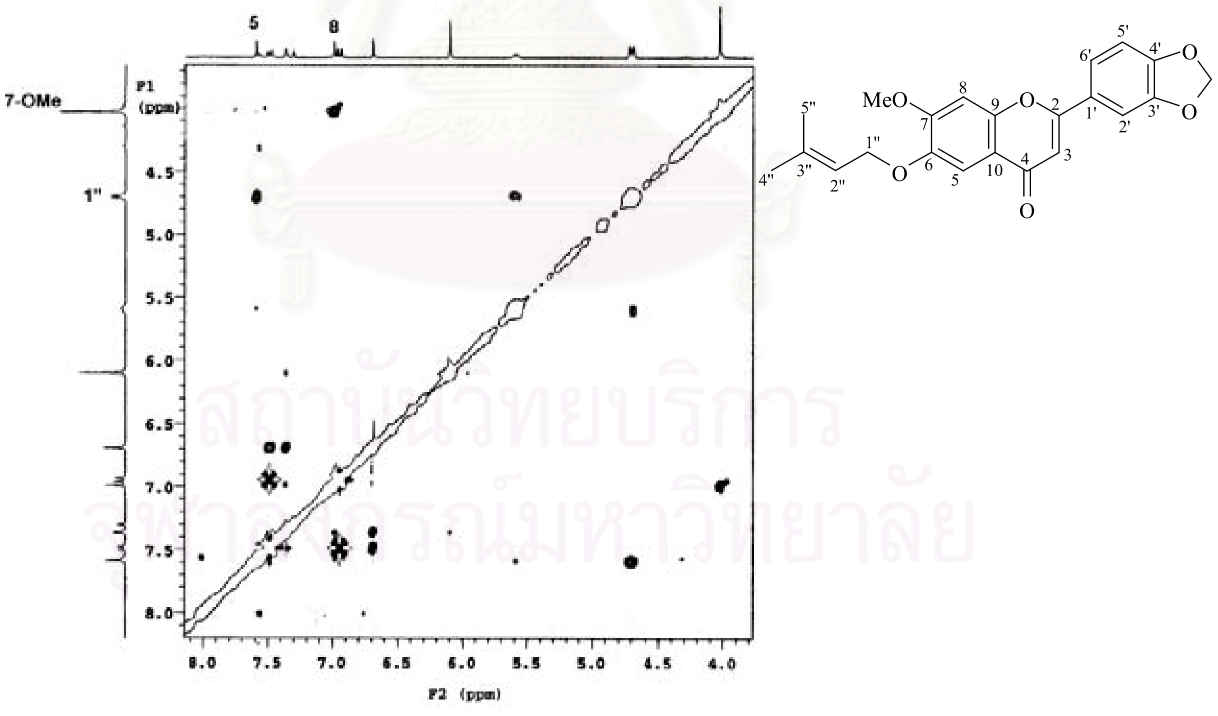

Figure 103 NOESY Spectrum of compound ME10 ($CDCl_3$)

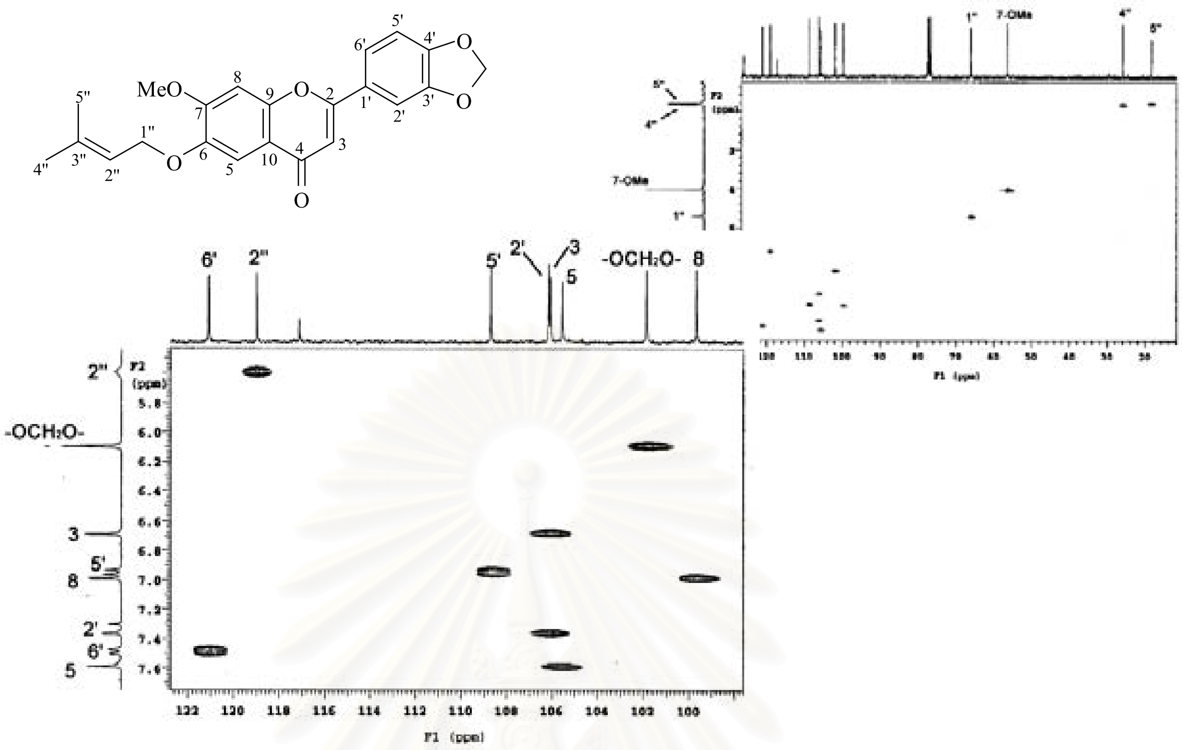

Figure 104 HSQC Spectrum of compound ME10 ($CDCl_3$)

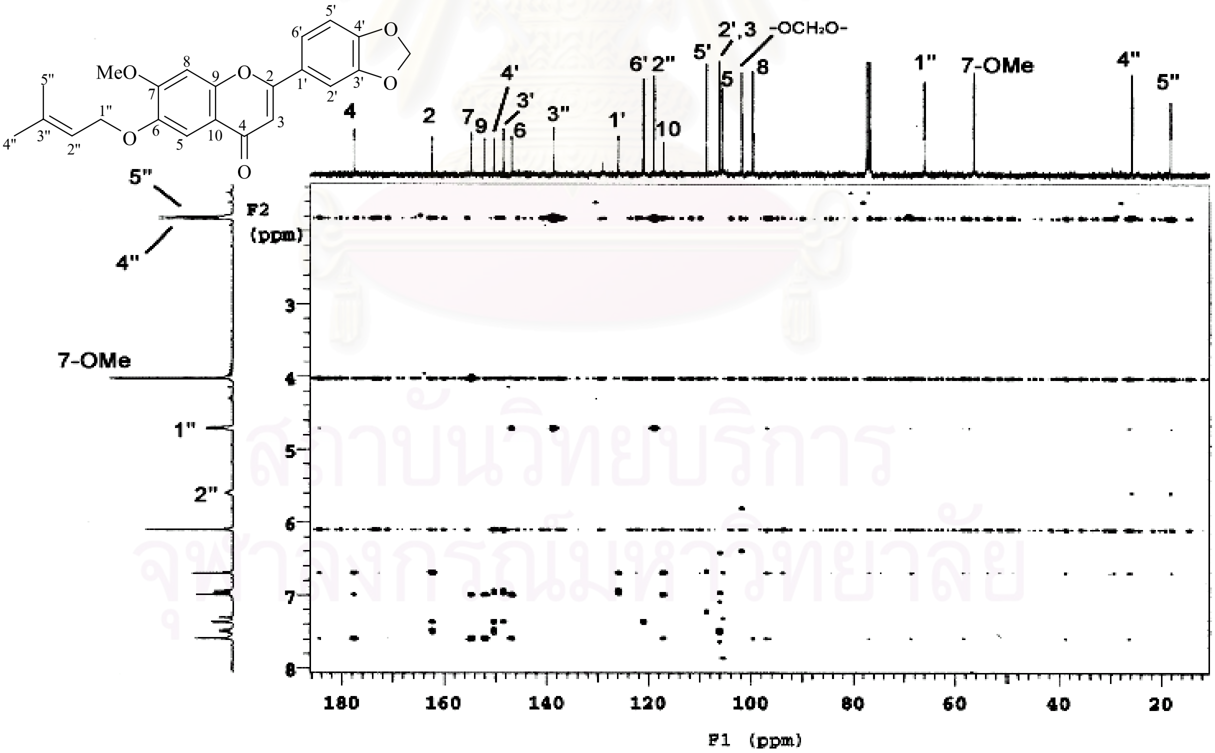

Figure 105 HMBC Spectrum of compound ME10 ($CDCl_3$) [δ_H 1.4-8.0 ppm, δ_C 10-186 ppm]

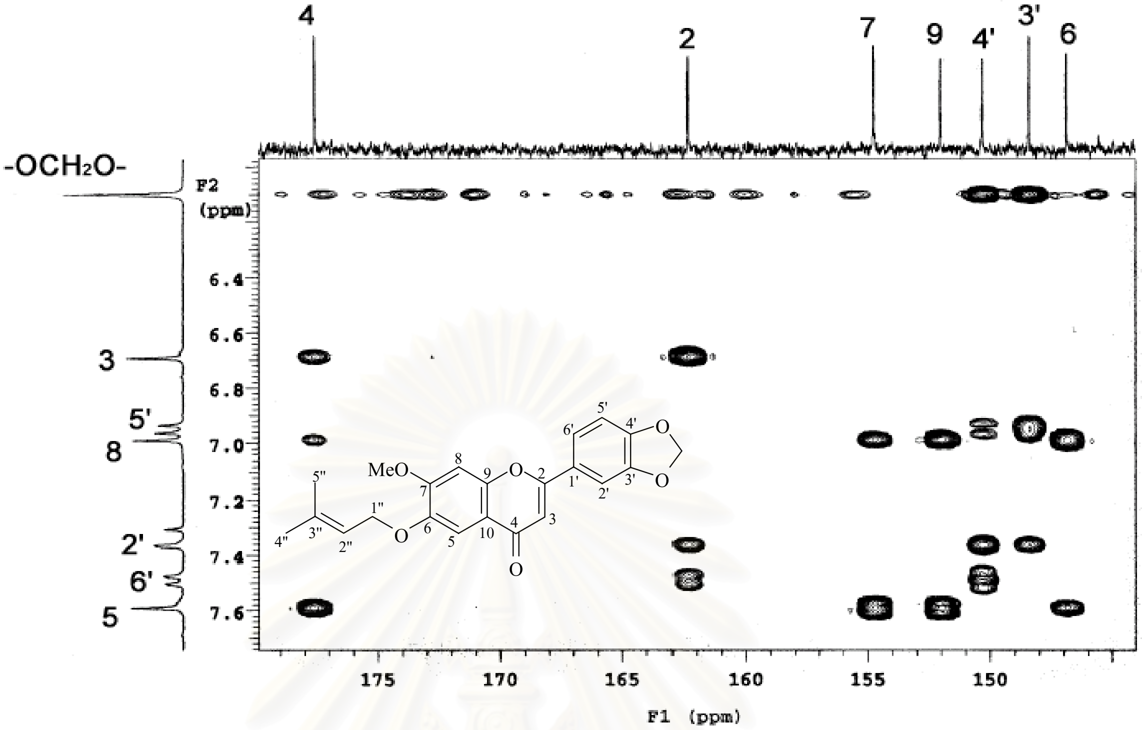

Figure 106 HMBC Spectrum of compound ME10 (CDCl$_3$) [δ_H 6.2-7.7 ppm, δ_C 144-180 ppm]

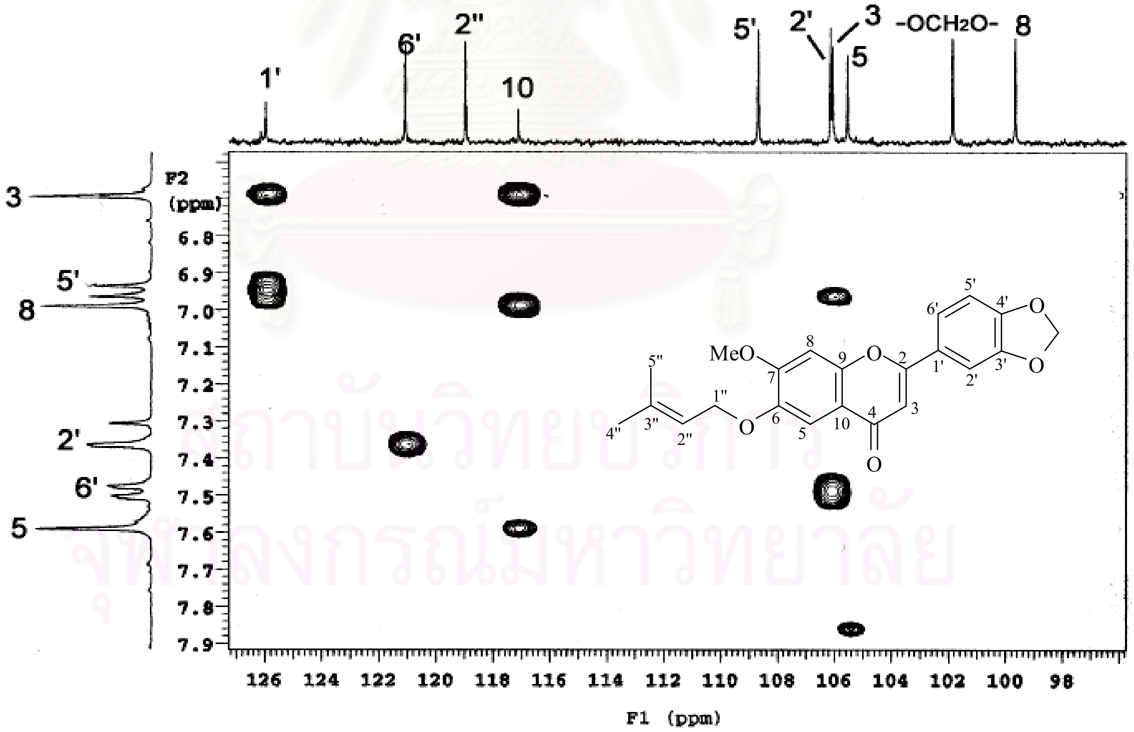

Figure 107 HMBC Spectrum of compound ME10 (CDCl$_3$) [δ_H 6.6-7.9 ppm, δ_C 97-127 ppm]

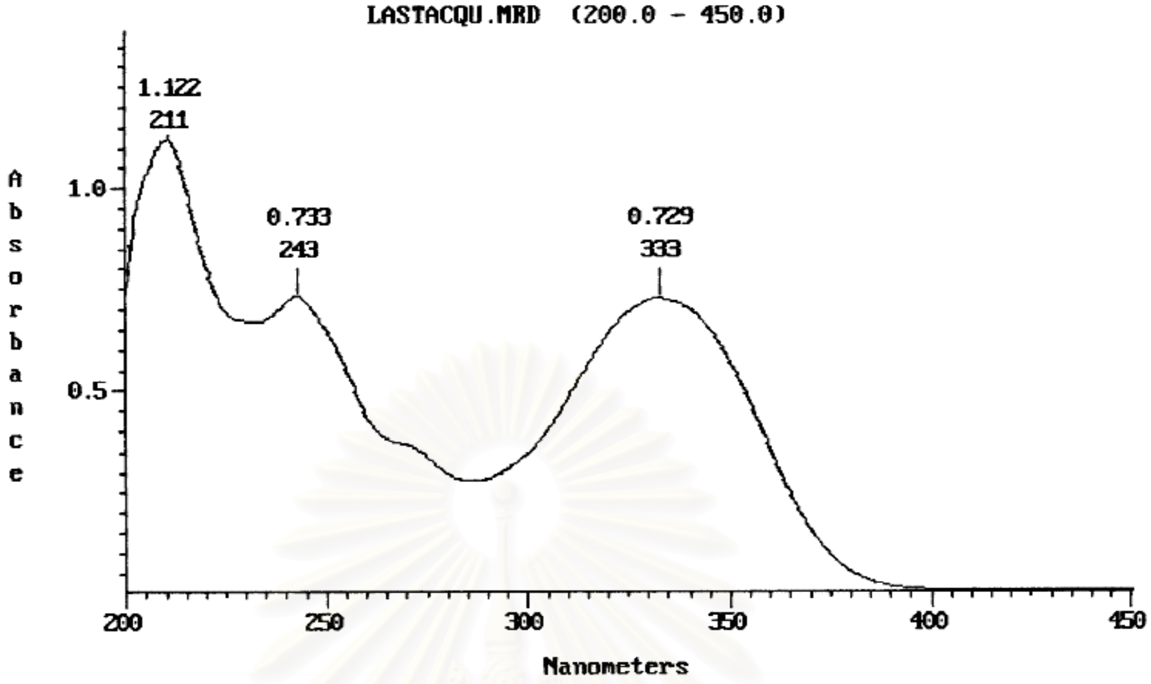

Figure 108 UV Spectrum of compound ME11 (methanol)

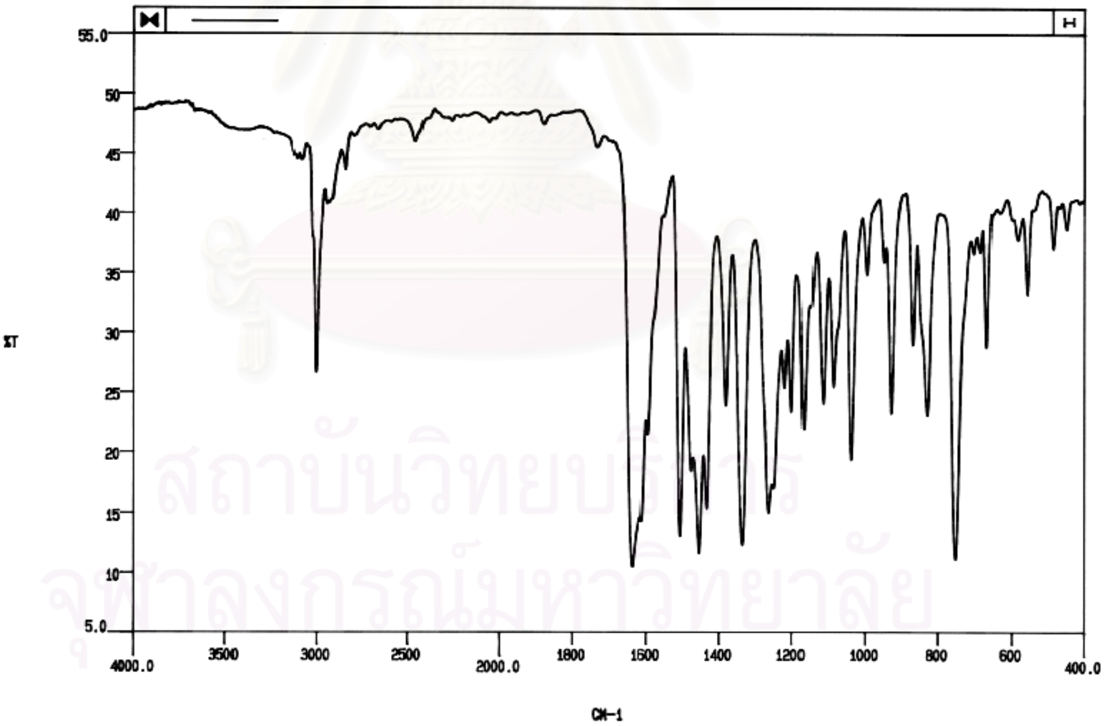

Figure 109 IR Spectrum of compound ME11 (KBr disc)

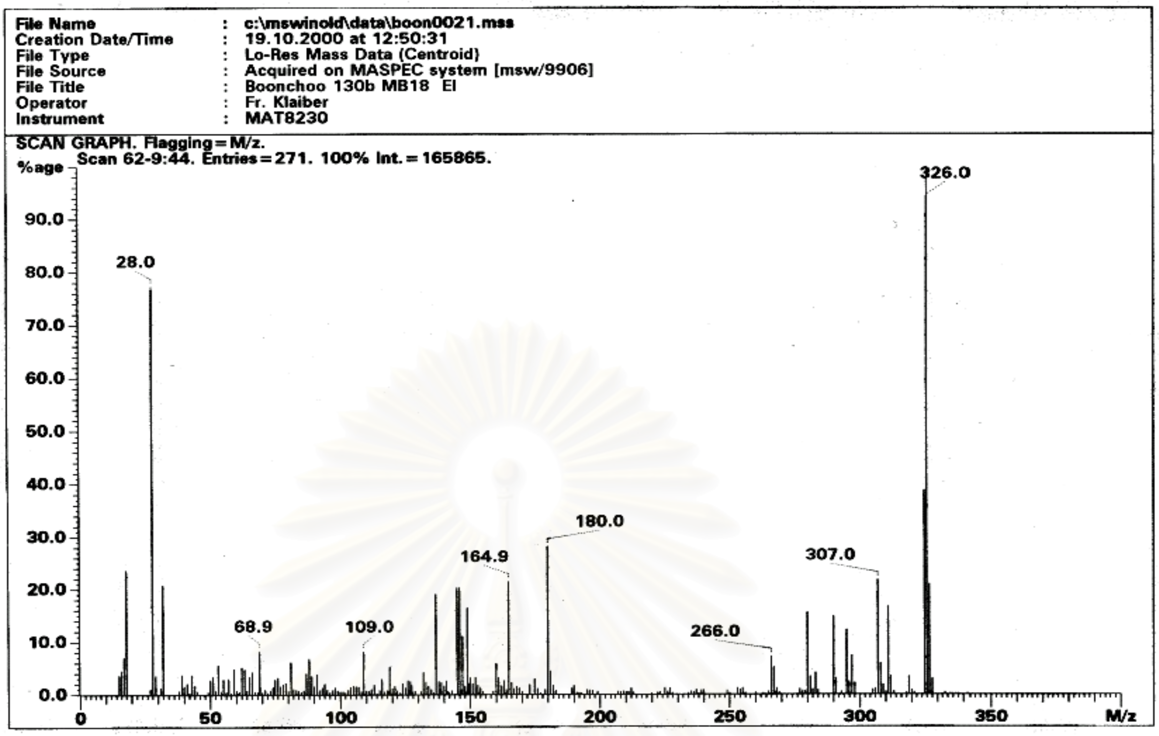

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Figure 110 EI Mass spectrum of compound ME11

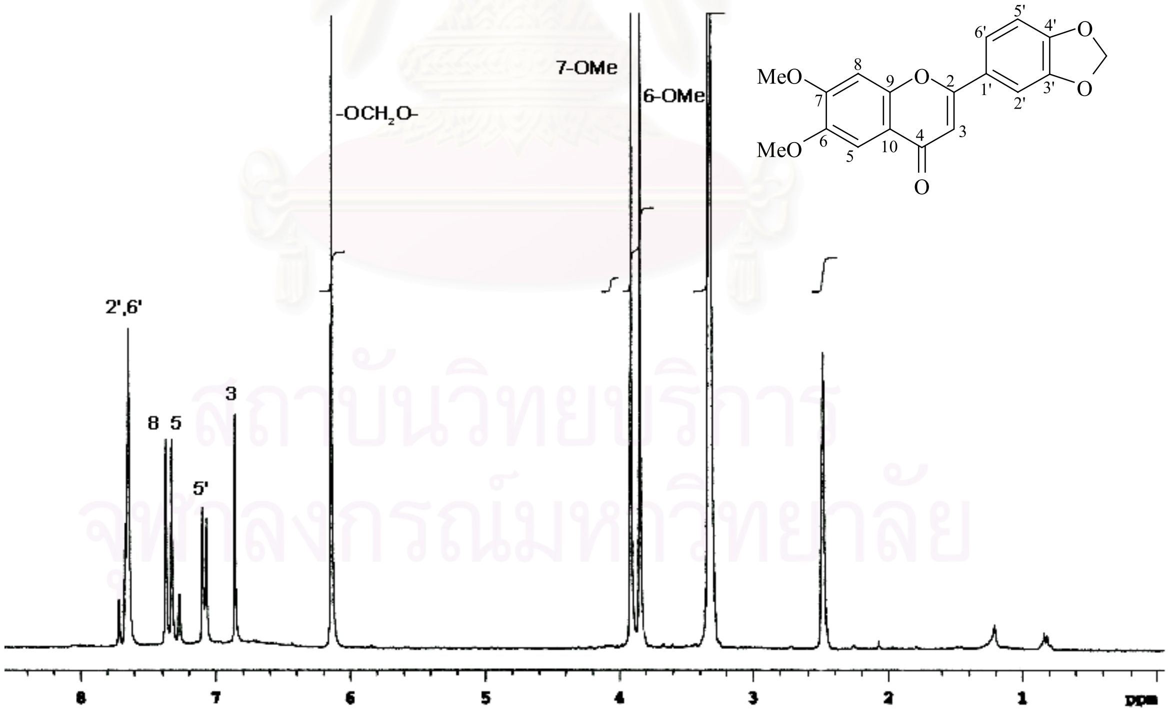

Figure 111 ^{1}H NMR (300 MHz) Spectrum of compound ME11 (DMSO-d_6)

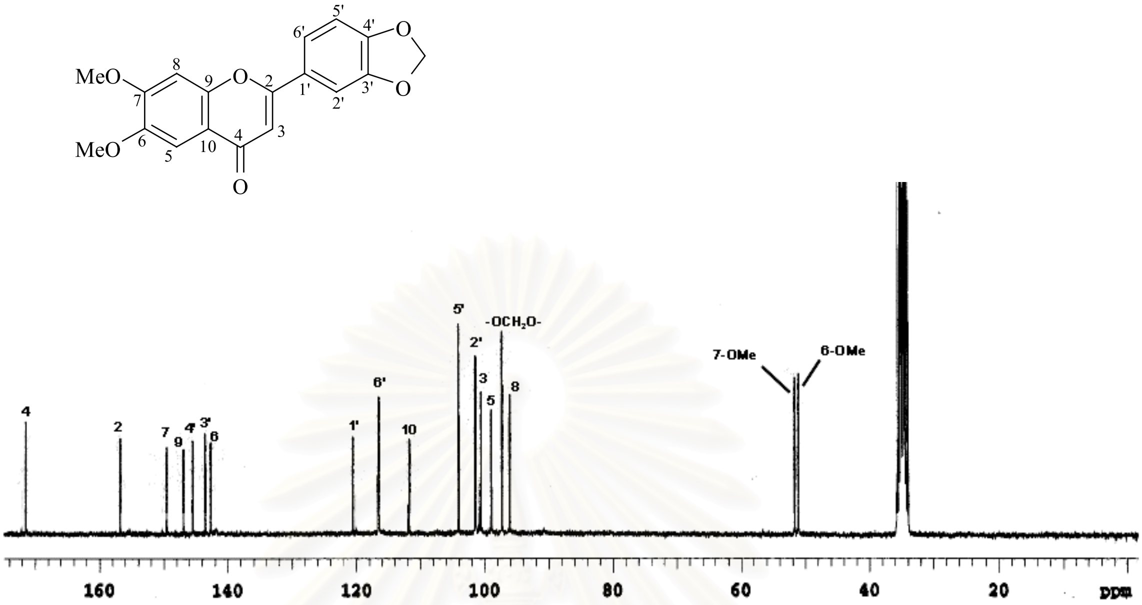

Figure 112 ^{13}C NMR (75 MHz) Spectrum of compound ME11 (DMSO-d_6)

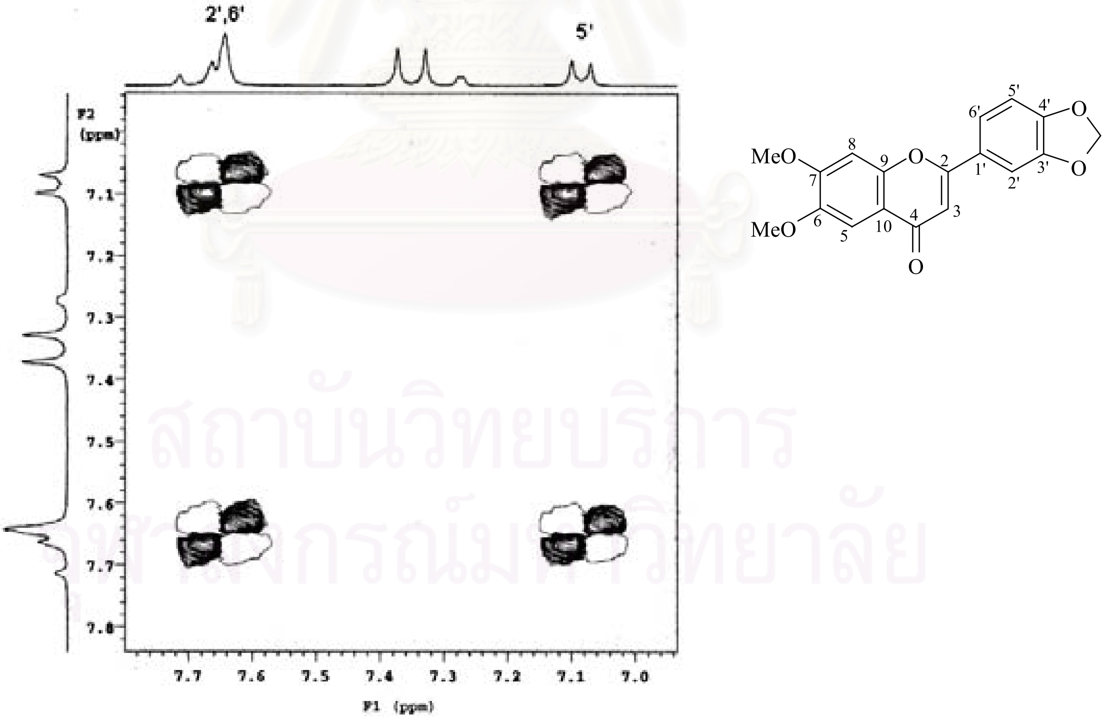

Figure 113 ^{1}H-^{1}H COSY Spectrum of compound ME11 (DMSO-d_6)

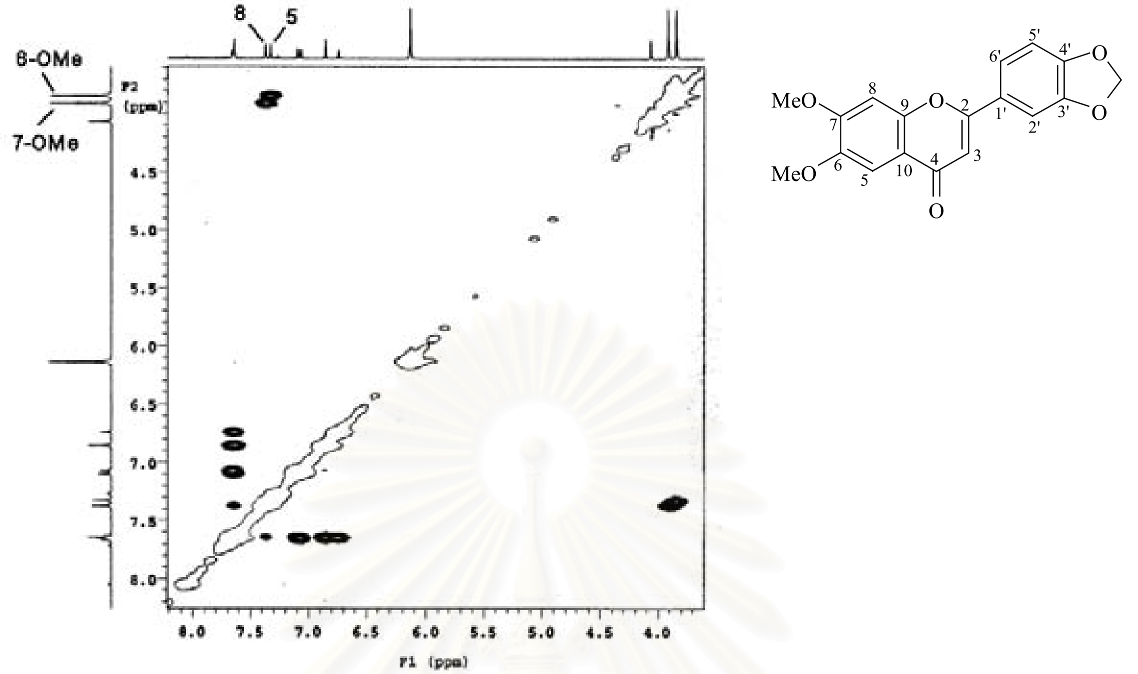

Figure 114 NOESY Spectrum of compound ME11 (DMSO-d_6)

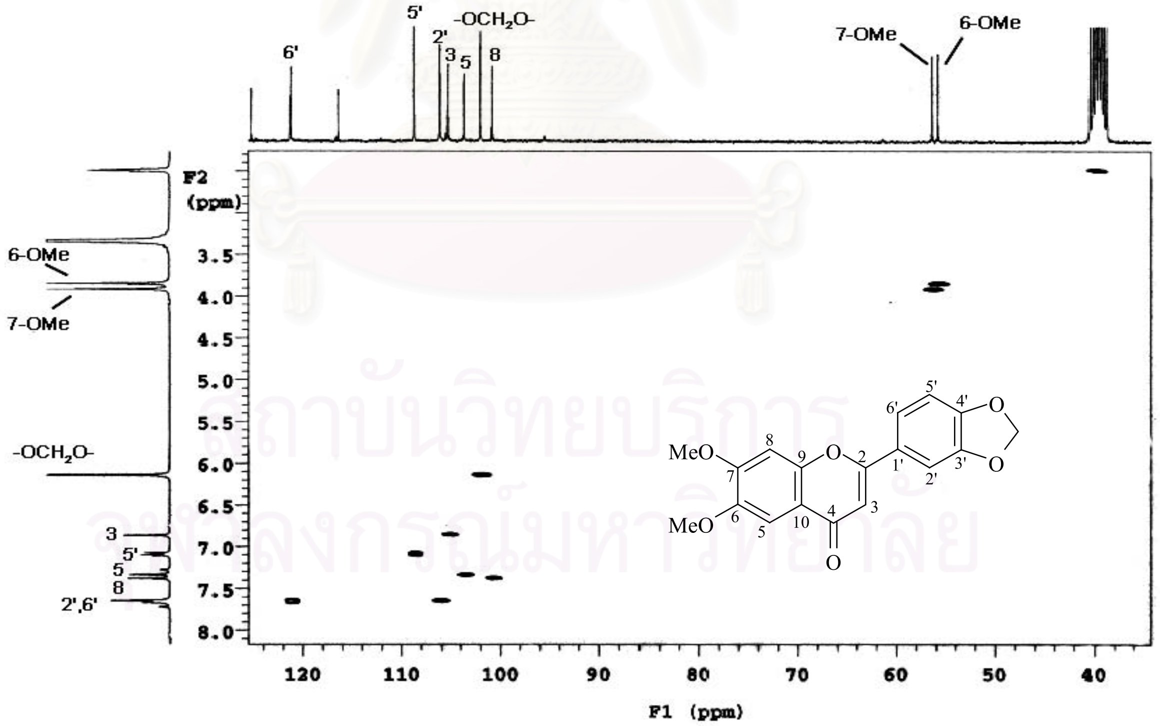

Figure 115 HSQC Spectrum of compound ME11 (DMSO-d_6)

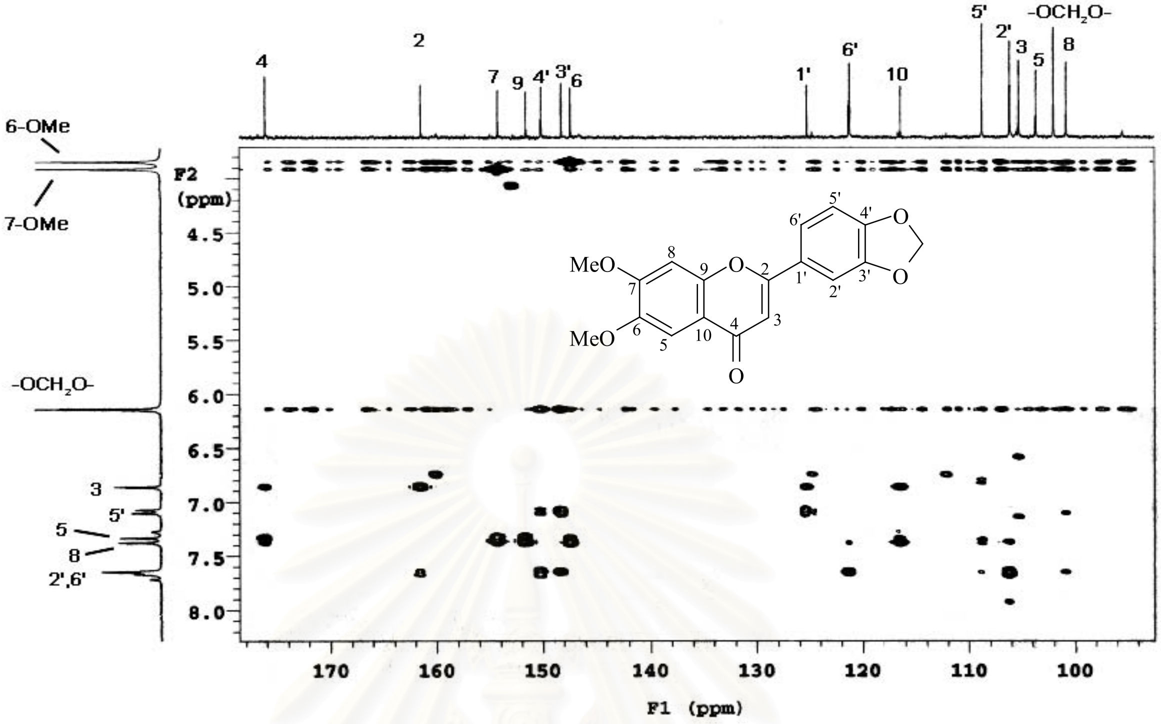

Figure 116 HMBC Spectrum of compound ME11 (DMSO-d_6)

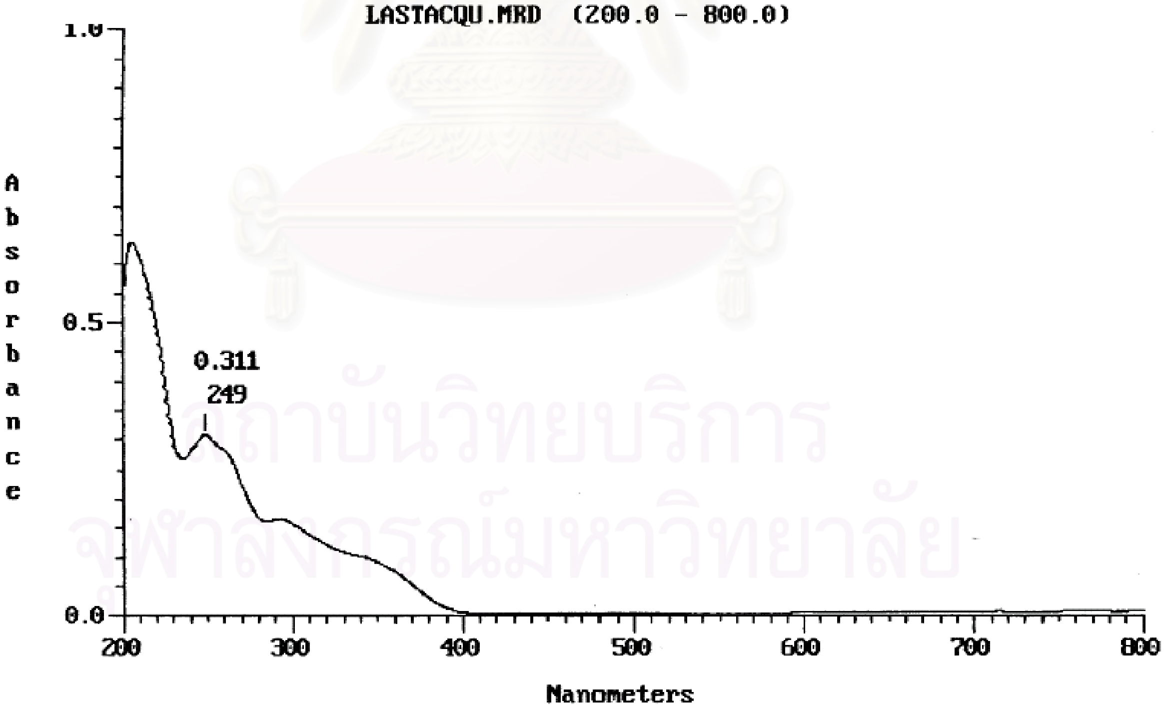

Figure 117 UV Spectrum of compound ME12 (methanol)

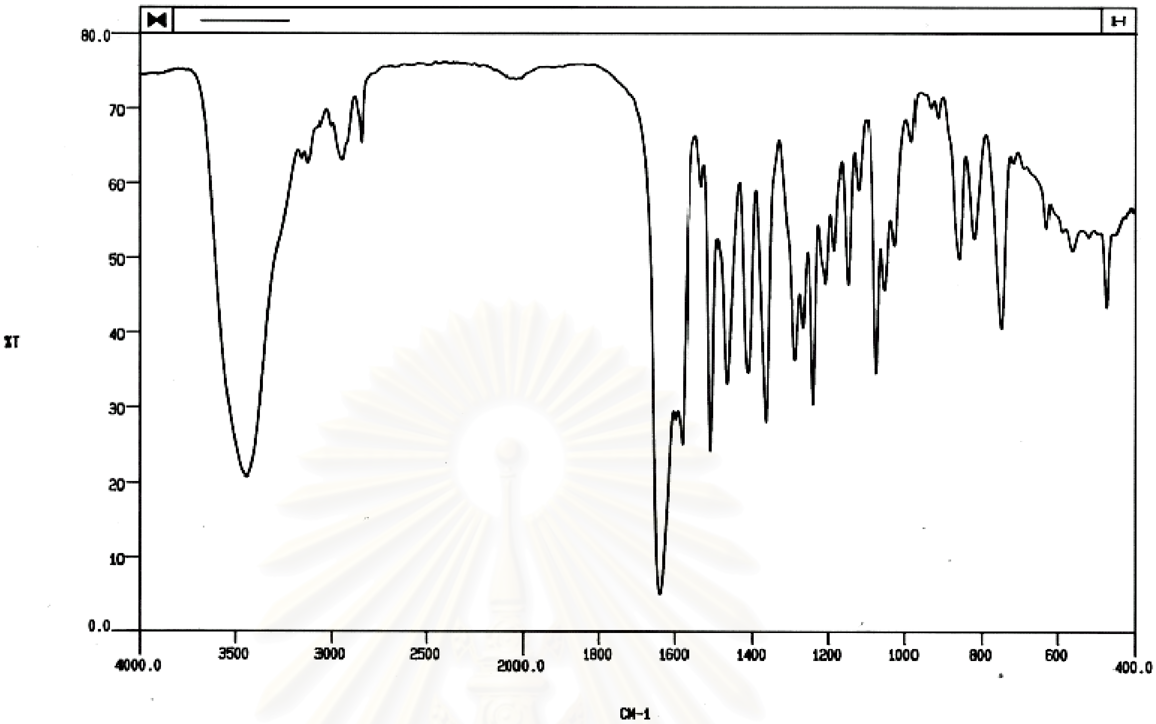

Figure 118 IR Spectrum of compound ME12 (KBr disc)

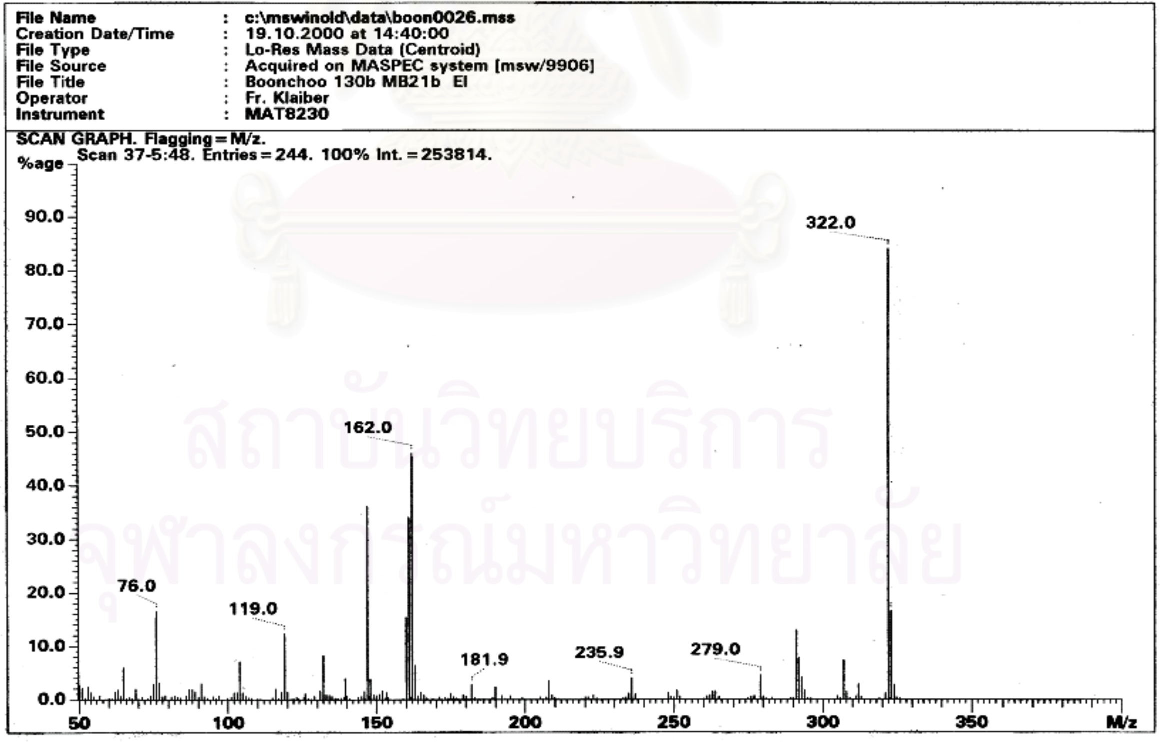
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Figure 119 EI Mass spectrum of compound ME12

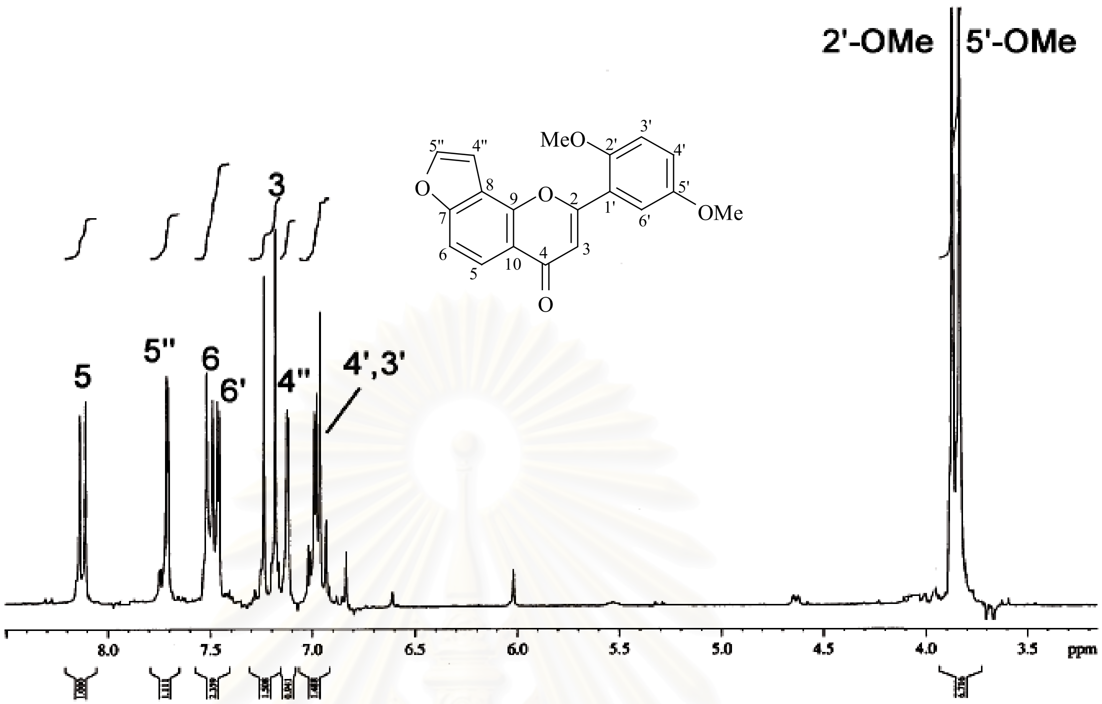

Figure 120 ^{1}H NMR (300 MHz) Spectrum of compound ME12 ($CDCl_3$)

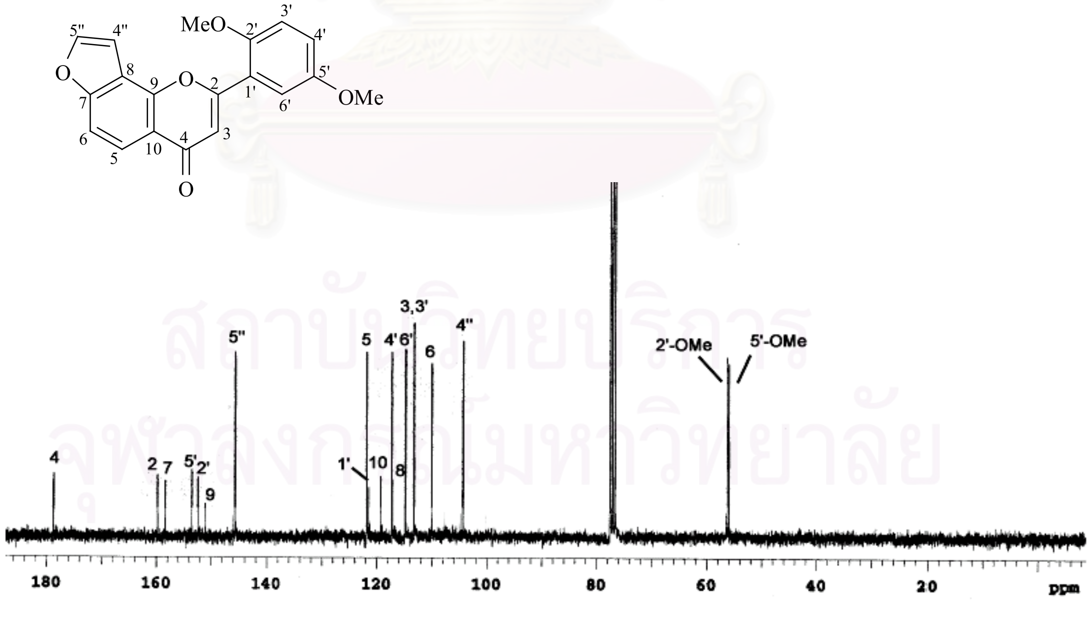

Figure 121 ^{13}C NMR (75 MHz) Spectrum of compound ME12 ($CDCl_3$)

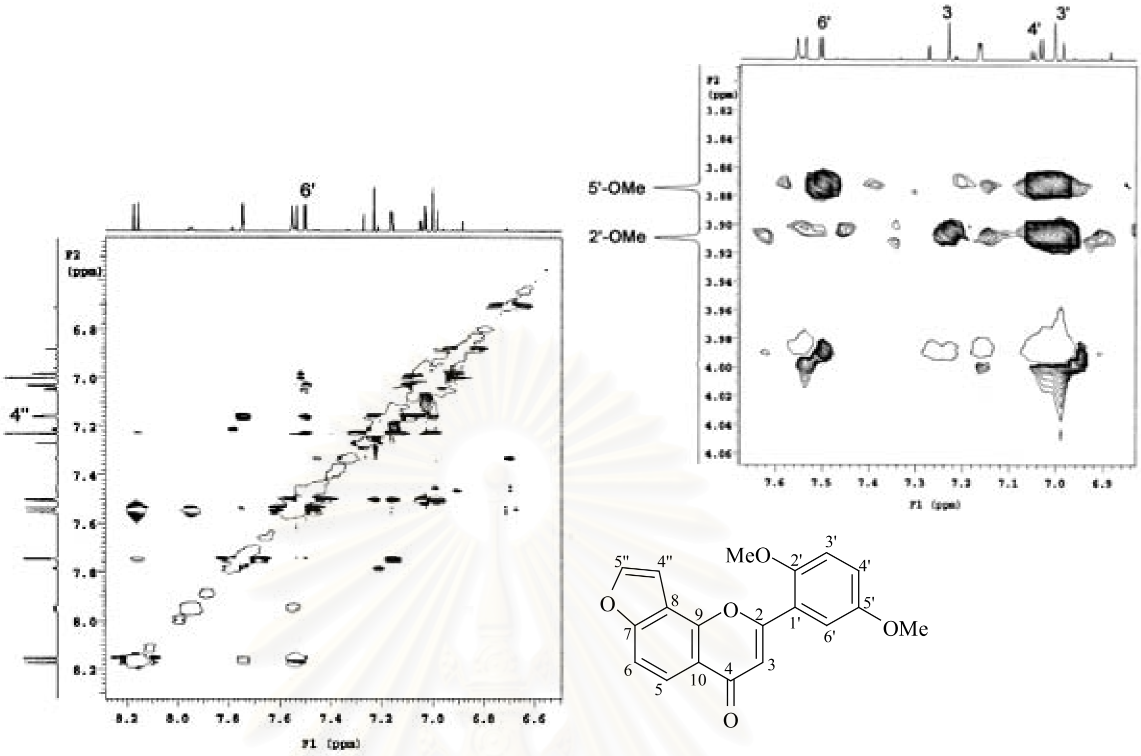

Figure 122 NOESY Spectrum of compound ME12 ($CDCl_3$)

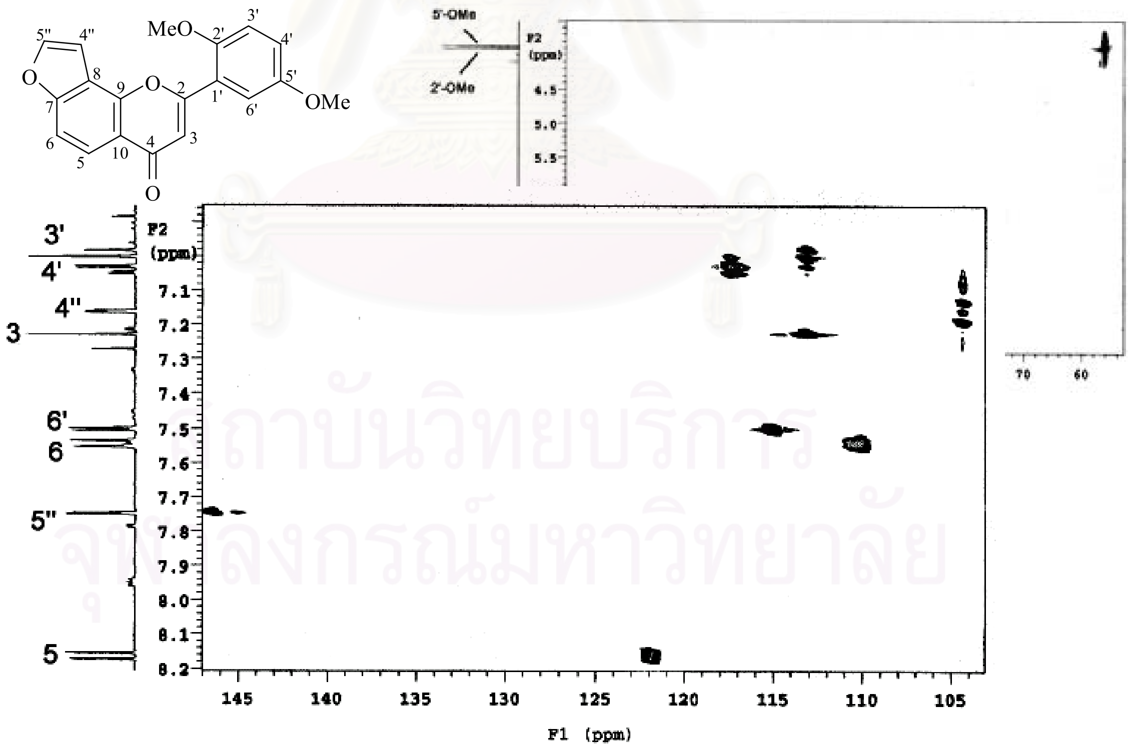

Figure 123 HSQC Spectrum of compound ME12 ($CDCl_3$)

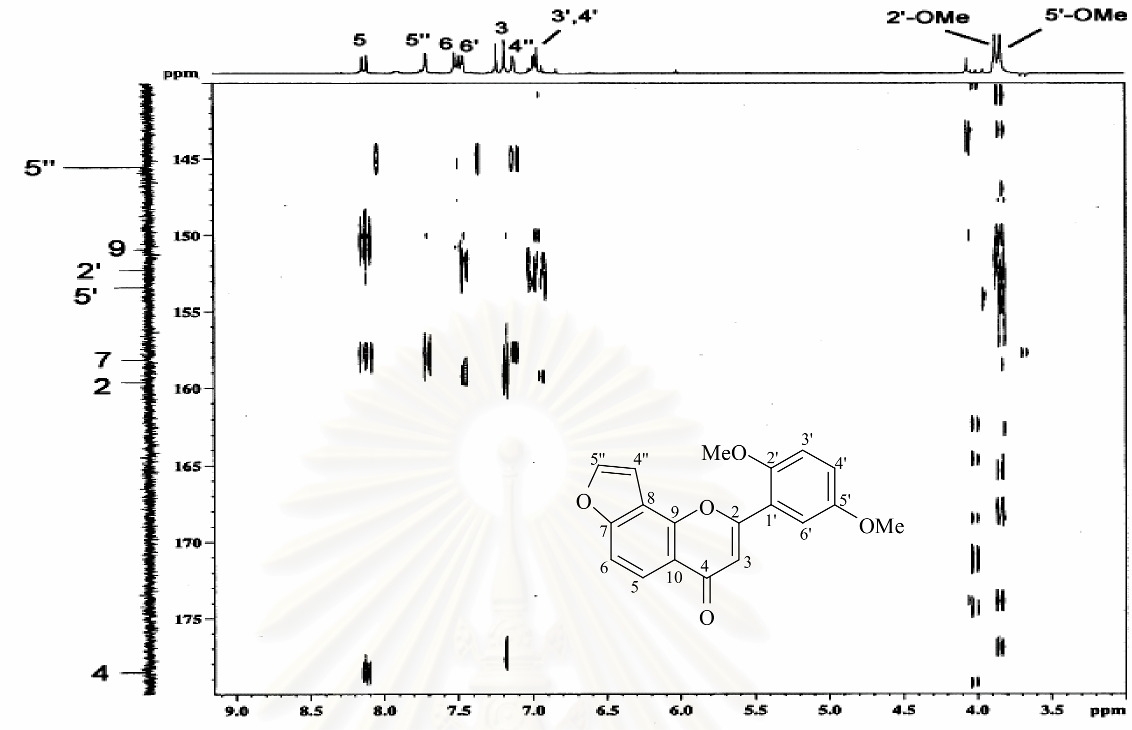

Figure 124 HMBC Spectrum of compound ME12 ($CDCl_3$) [δ_H 3.0-9.1 ppm, δ_C 140-180 ppm]

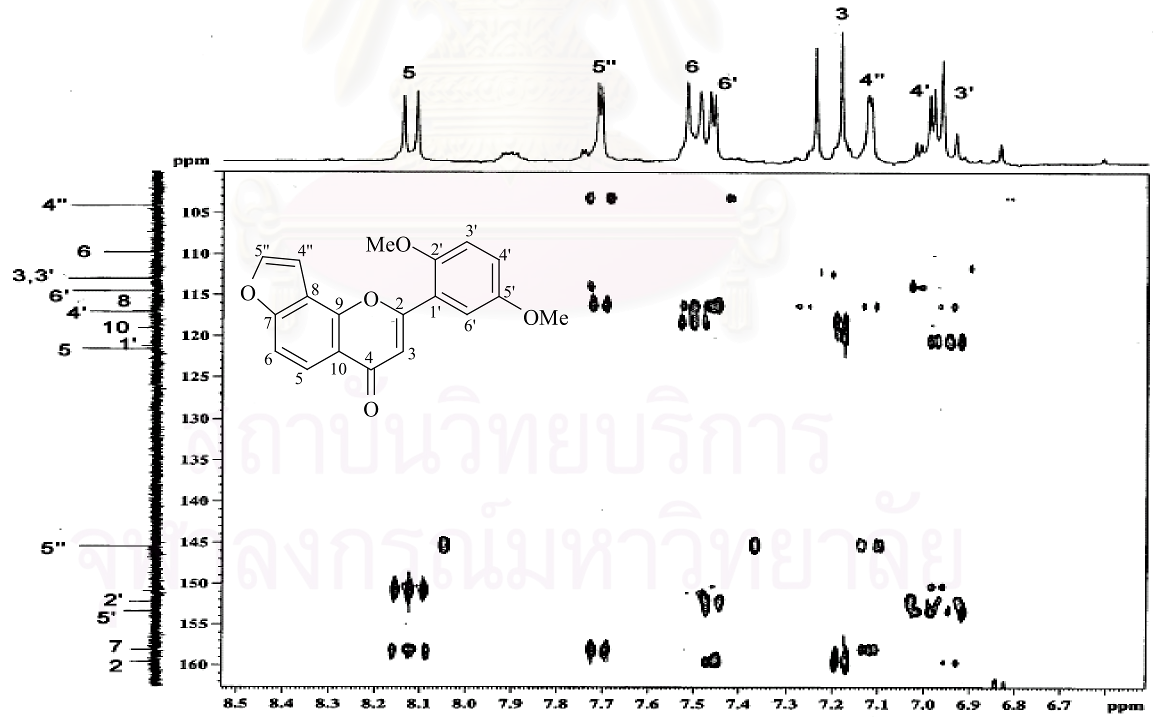

Figure 125 HMBC Spectrum of compound ME12 ($CDCl_3$) [δ_H 6.5-8.6 ppm, δ_C 100-163 ppm]

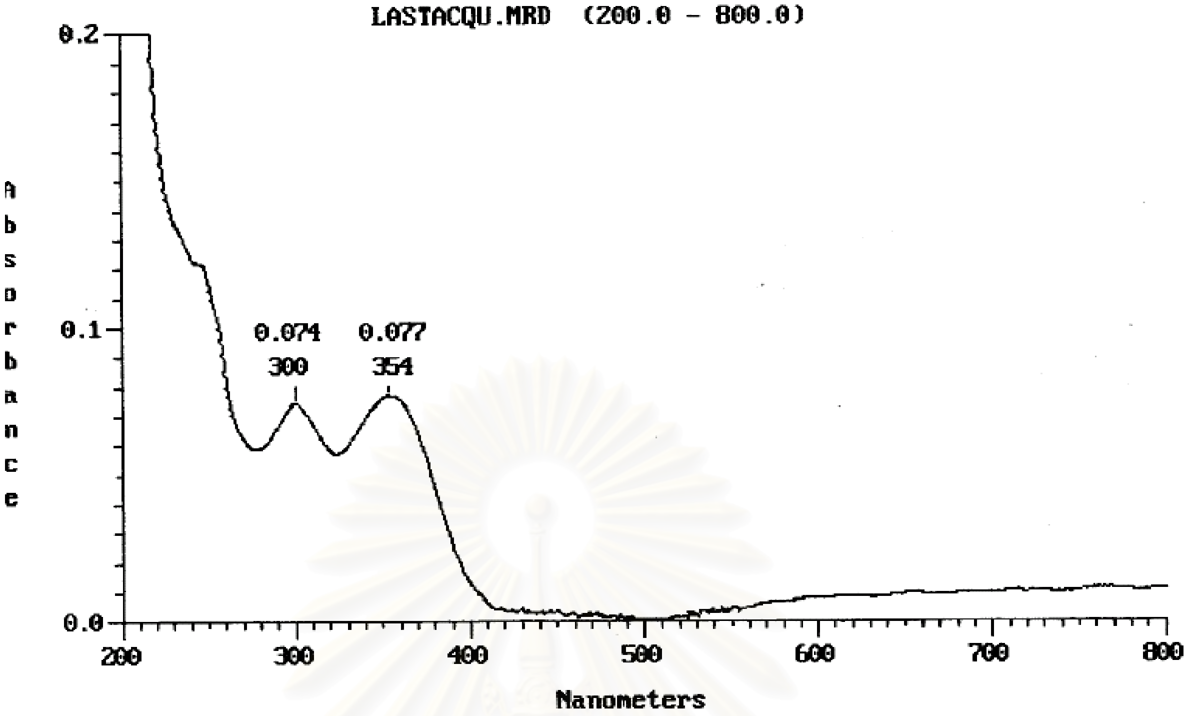

Figure 126 UV Spectrum of compound ME13 (methanol)

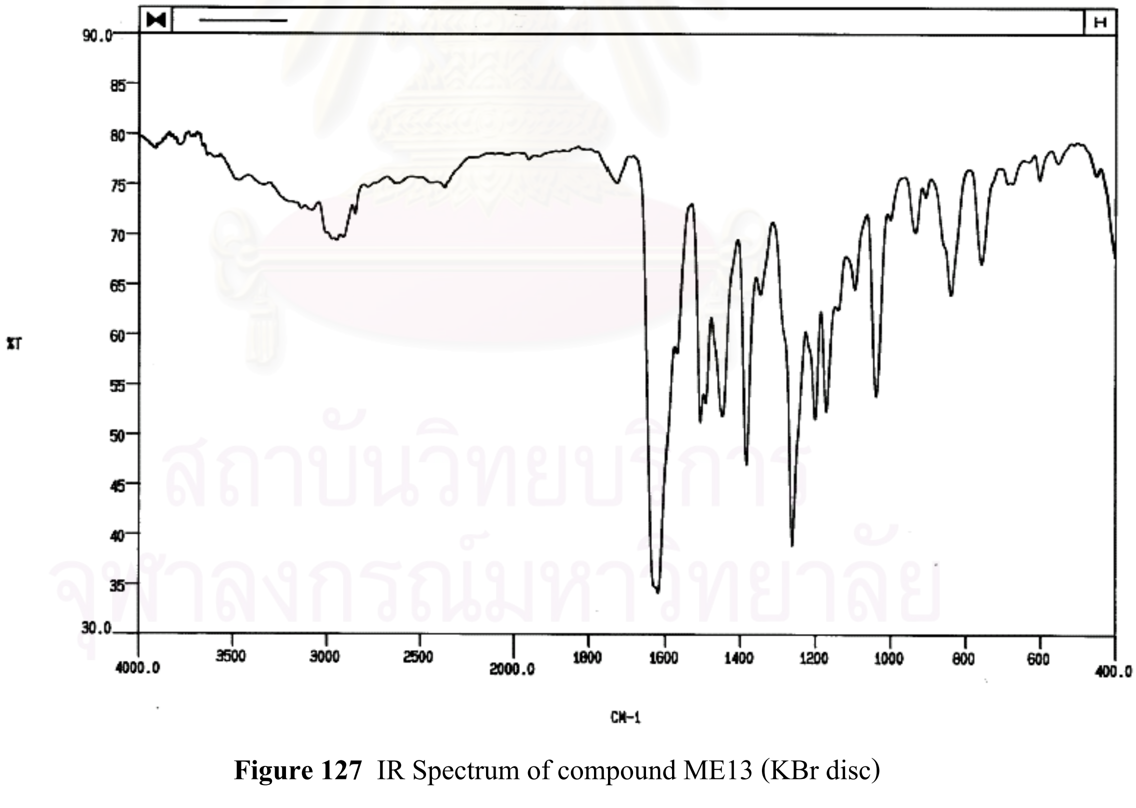

Figure 127 IR Spectrum of compound ME13 (KBr disc)

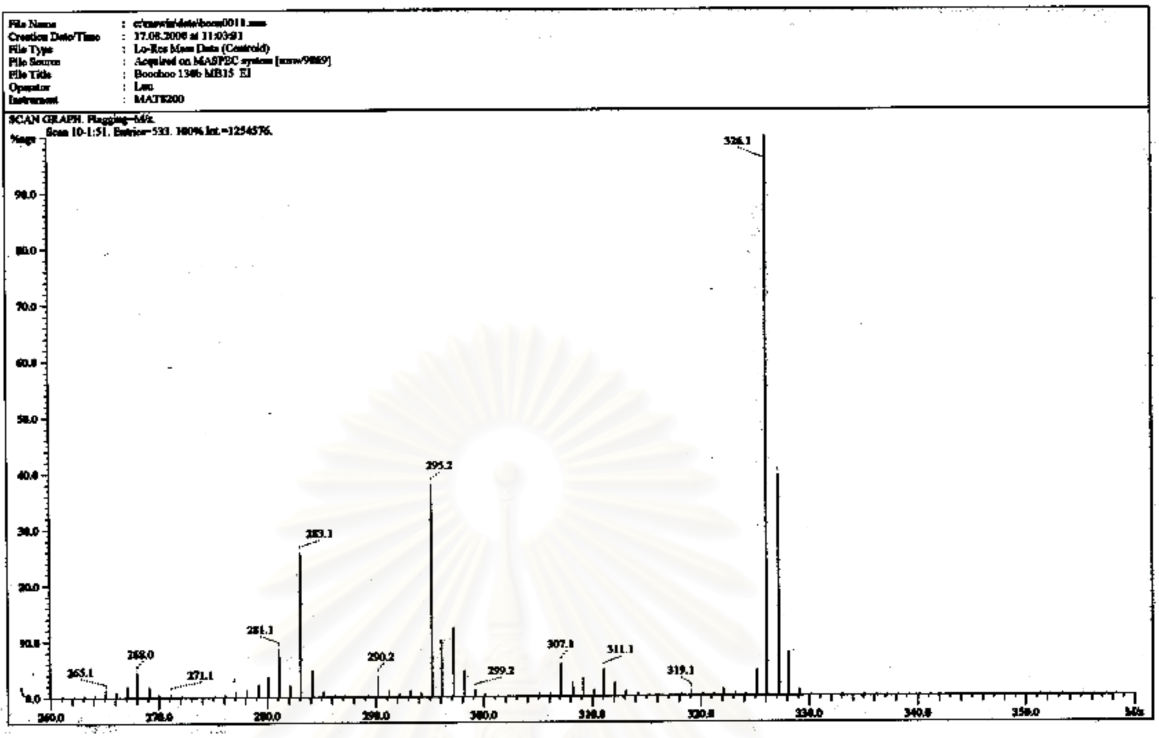

Figure 128 EI Mass spectrum of compound ME13

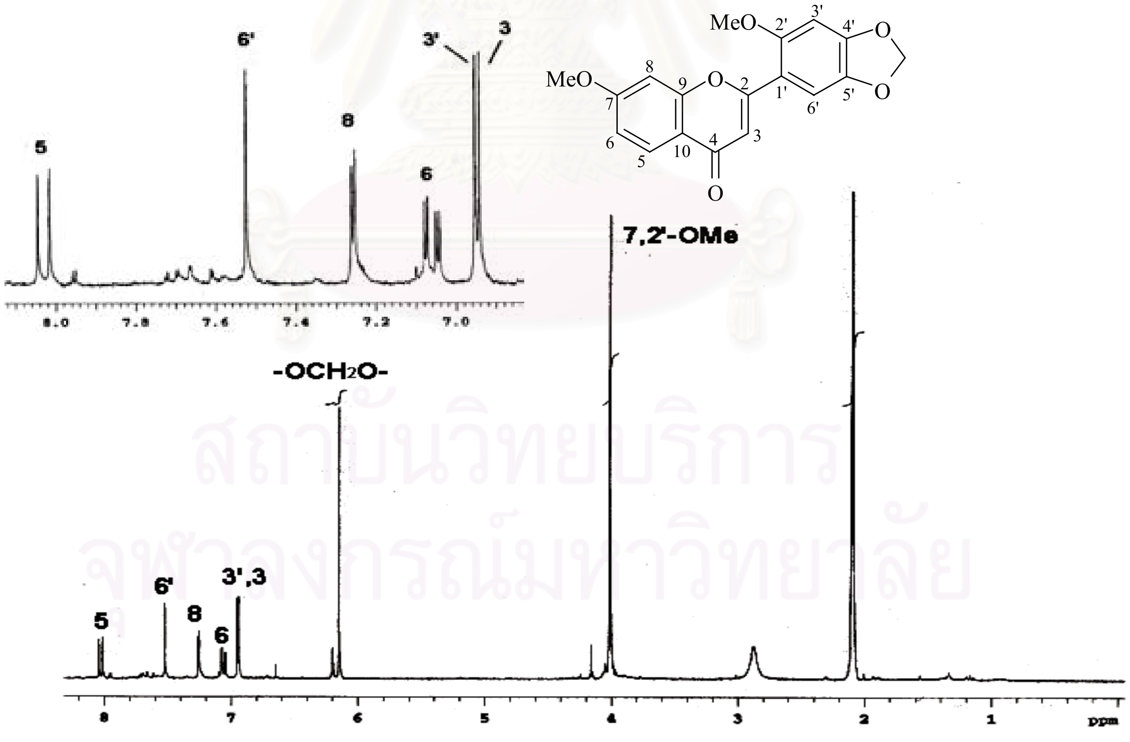

Figure 129 ^{1}H NMR (300 MHz) Spectrum of compound ME13 (acetone-d_6)

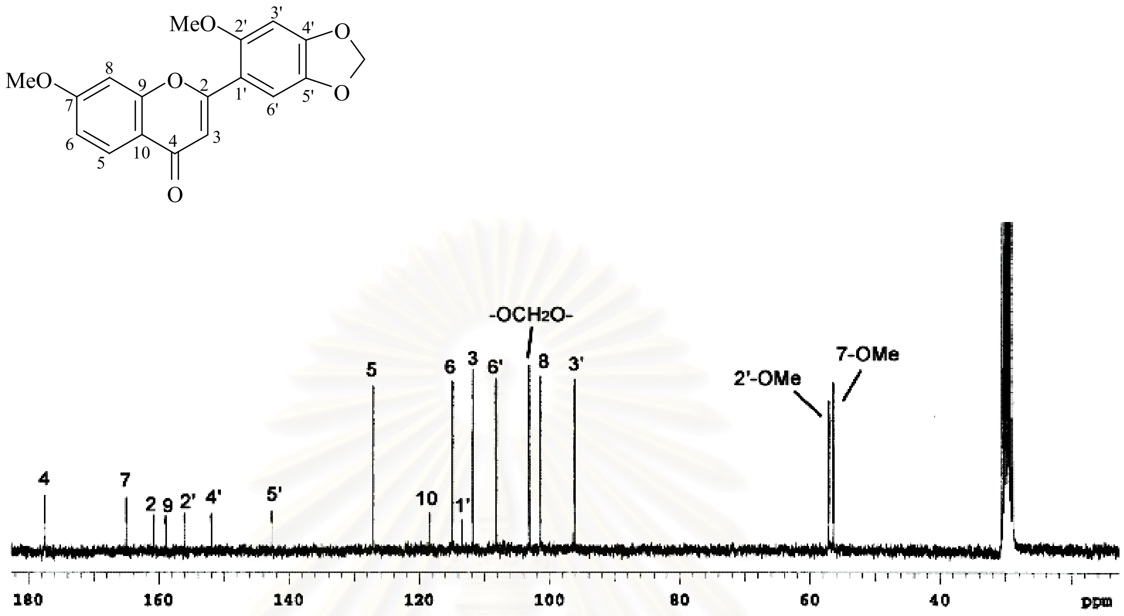

Figure 130 ^{13}C NMR (75 MHz) Spectrum of compound ME13 (acetone-d_6)

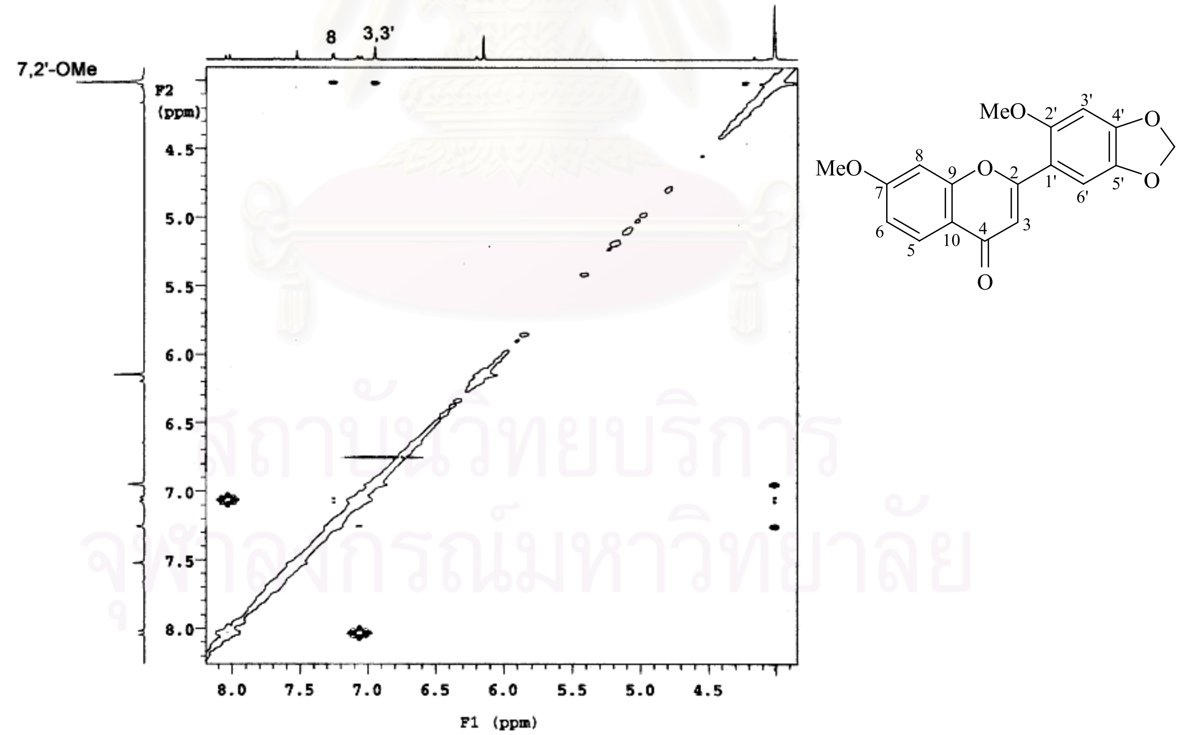

Figure 131 NOESY Spectrum of compound ME13 (acetone-d_6)

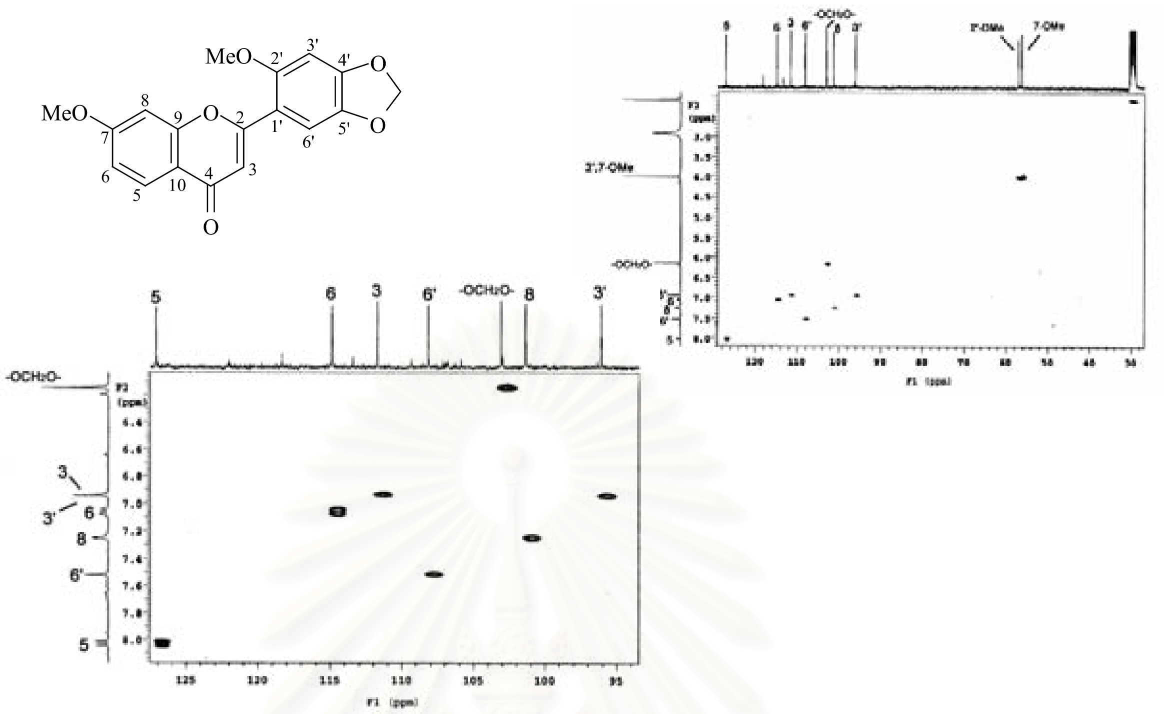

Figure 132 HSQC Spectrum of compound ME13 (acetone-d_6)

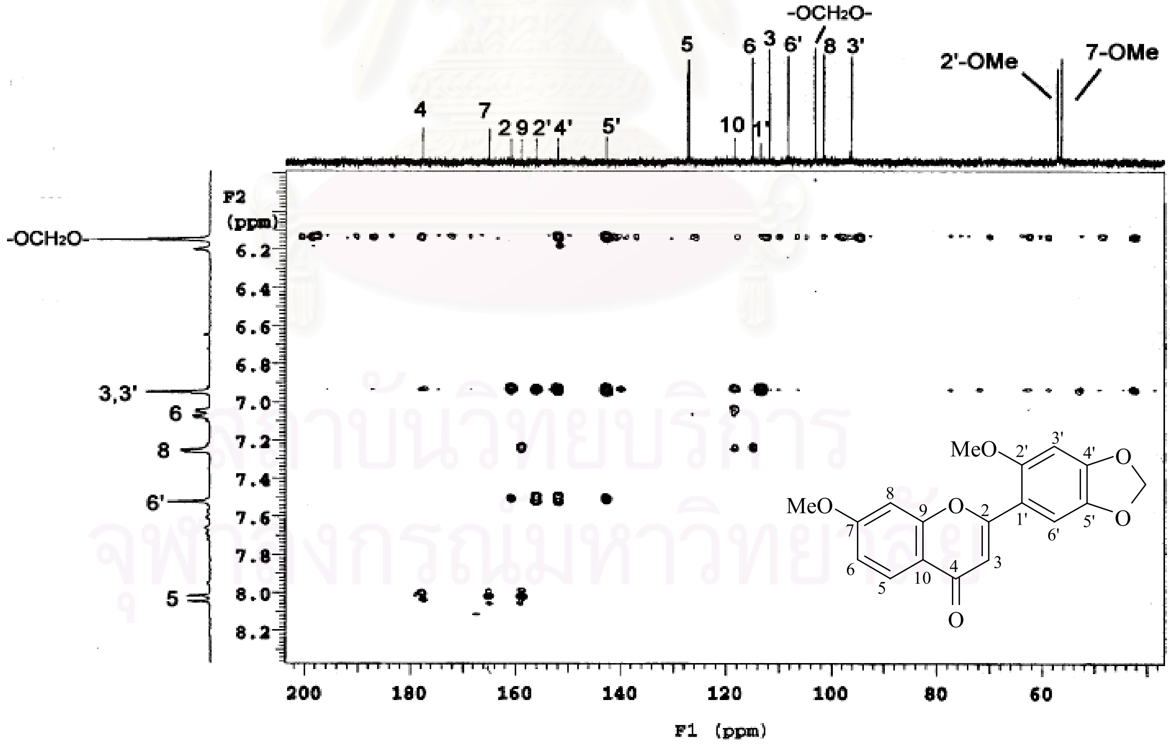

Figure 133 HMBC Spectrum of compound ME13 (acetone-d_6)

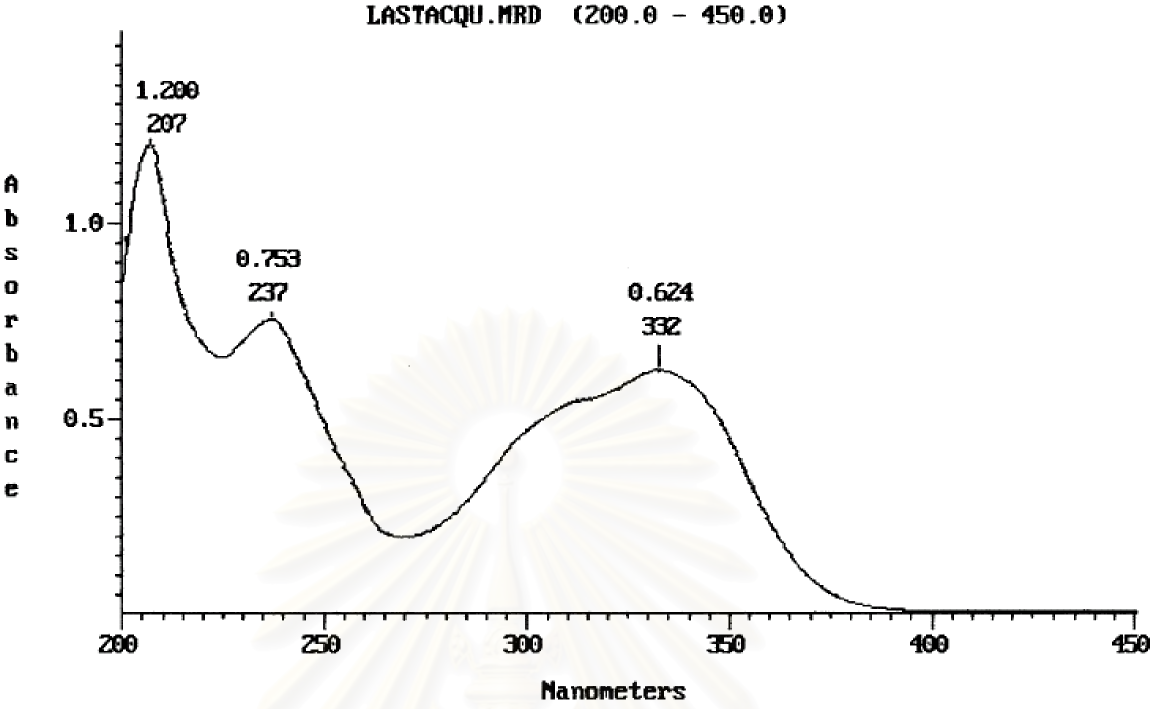

Figure 134 UV Spectrum of compound ME14 (methanol)

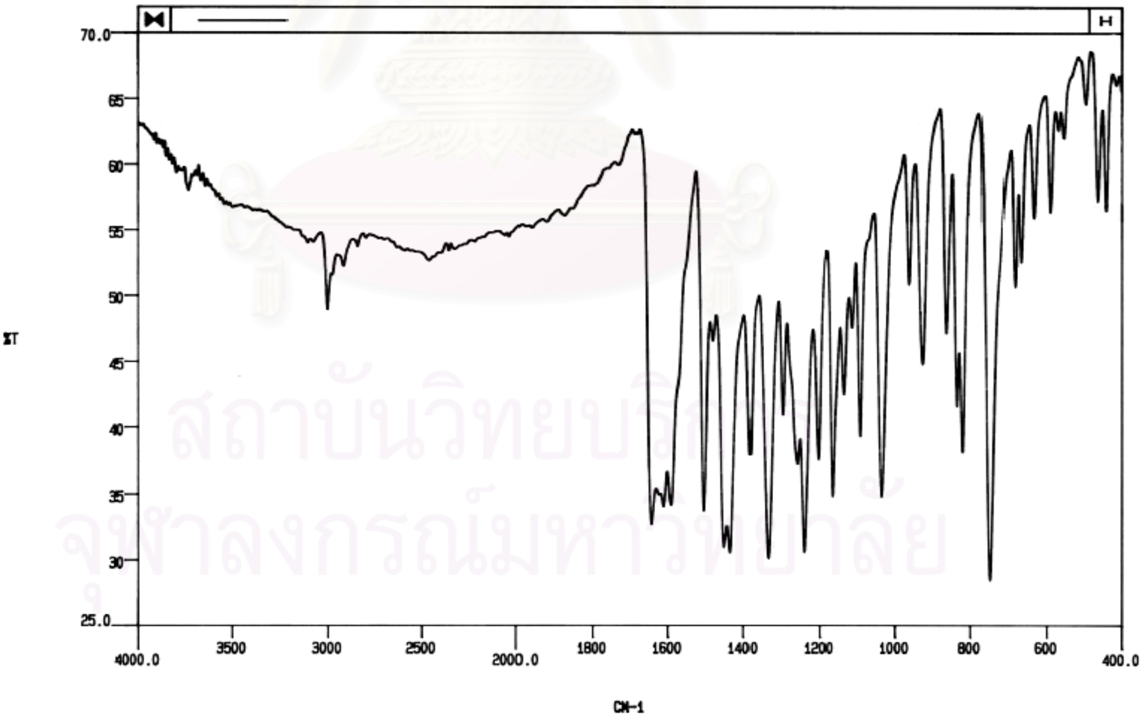

Figure 135 IR Spectrum of compound ME14 (KBr disc)

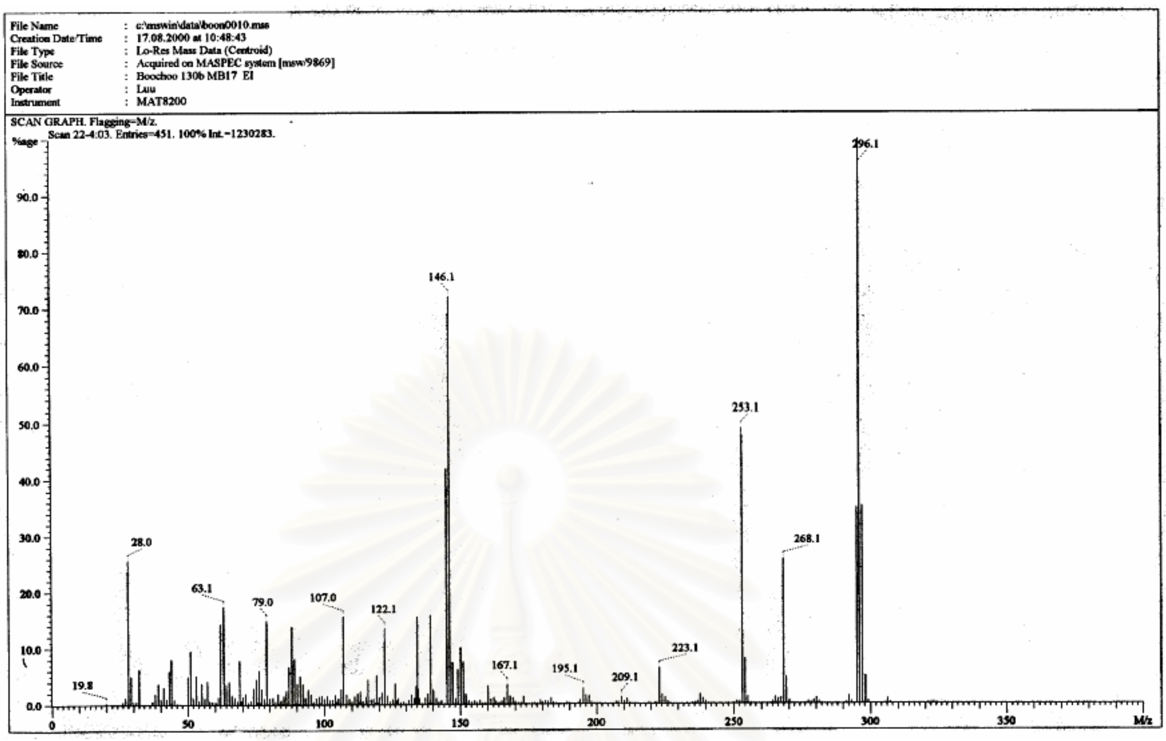

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Figure 136 EI Mass spectrum of compound ME14

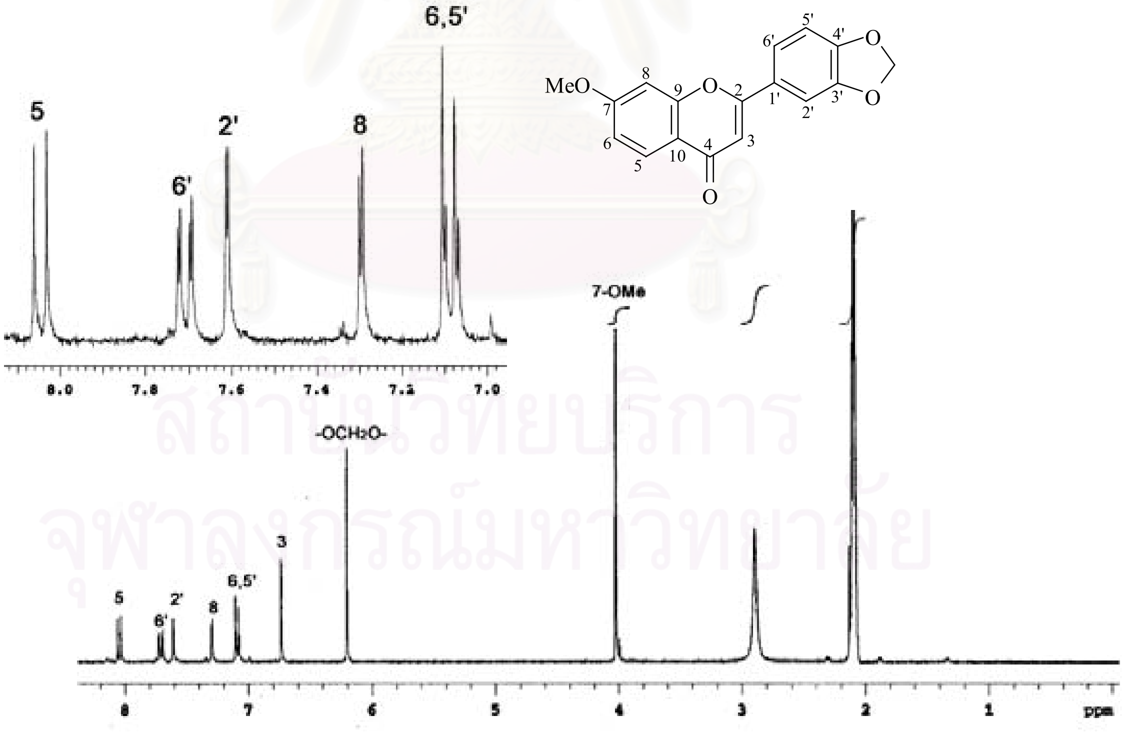
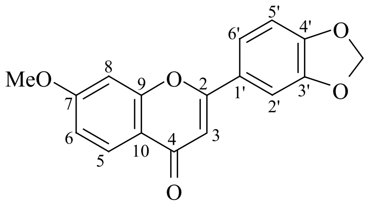


Figure 137 ^{1}H NMR (300 MHz) Spectrum of compound ME14 (acetone-d_6)



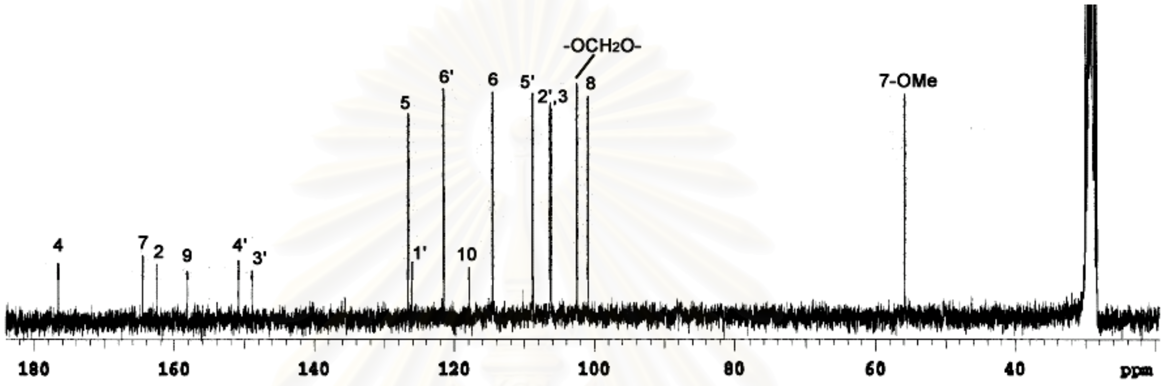


Figure 138 ^{13}C NMR (75 MHz) Spectrum of compound ME14 (acetone-d_6)

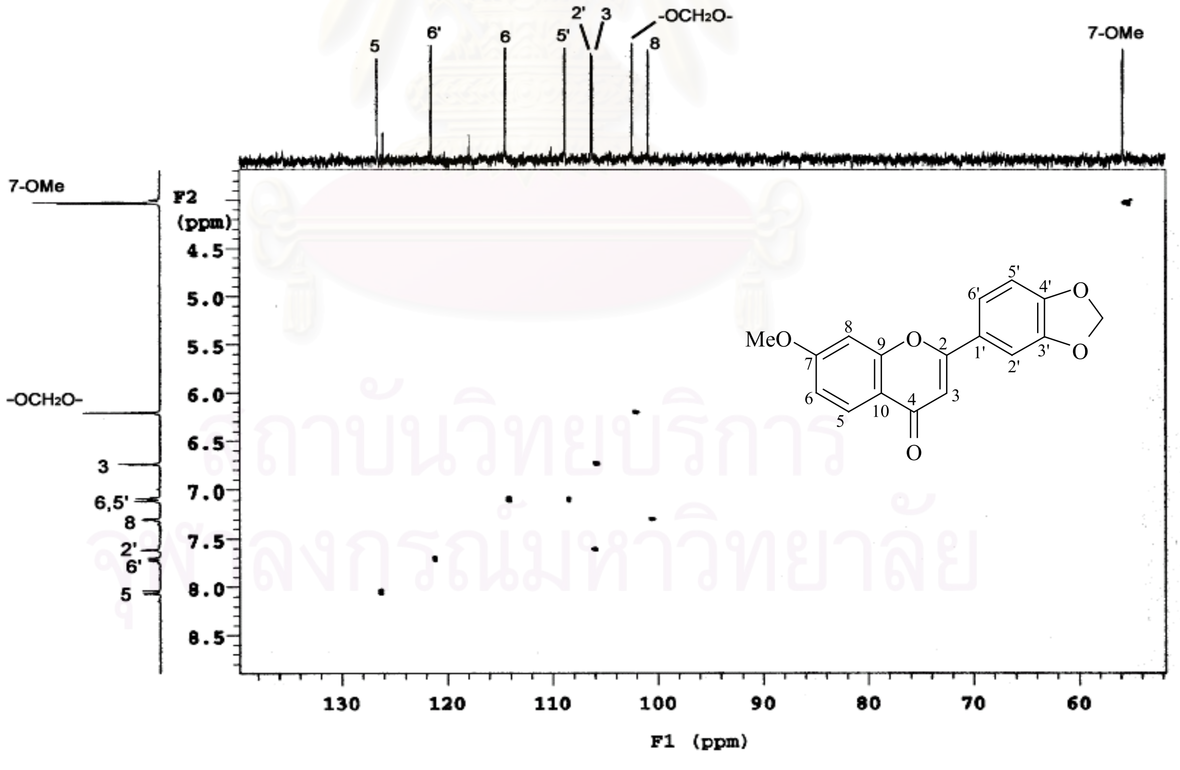


Figure 139 HSQC Spectrum of compound ME14 (acetone-d_6)

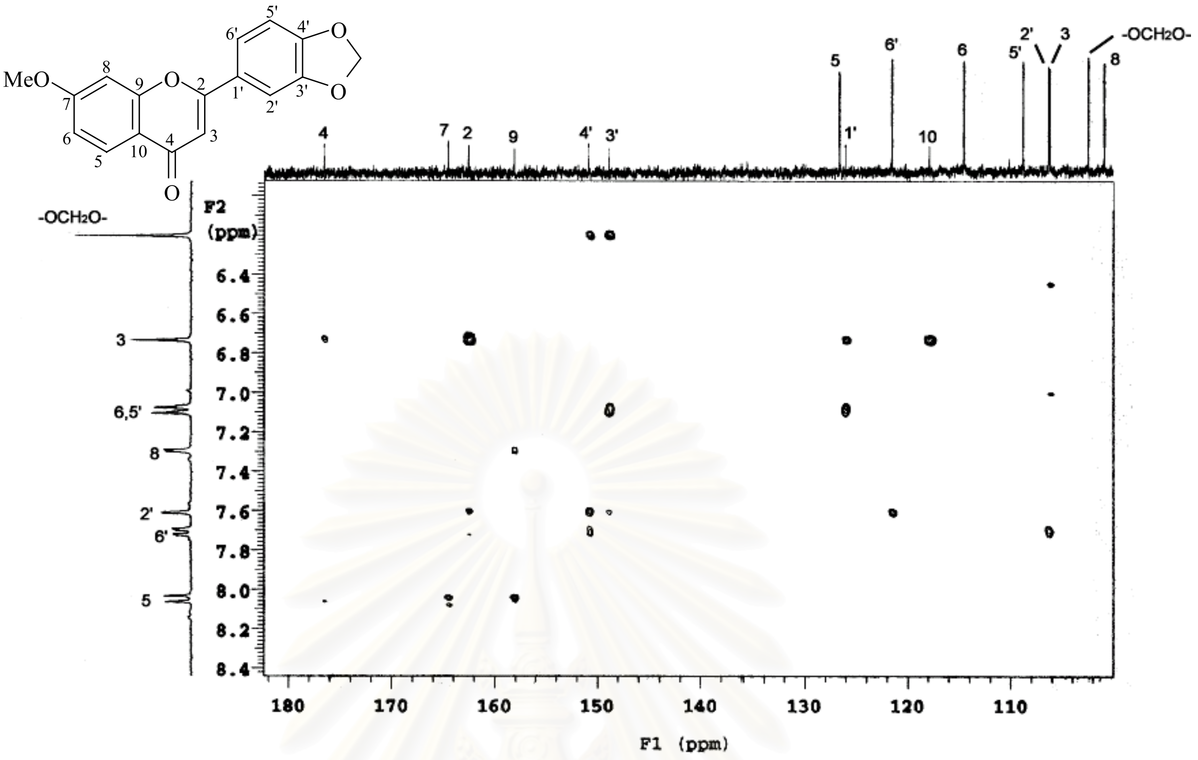


Figure 140 HMBC Spectrum of compound ME14 (acetone-d_6)

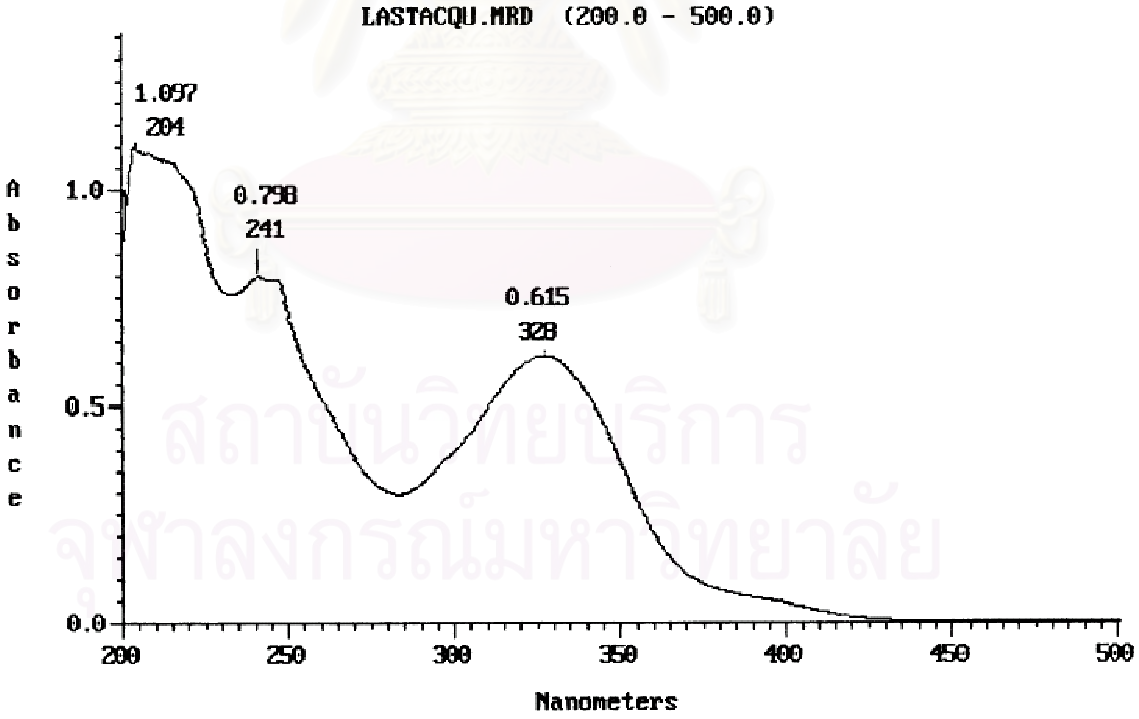


Figure 141 UV Spectrum of compound ME15 (methanol)

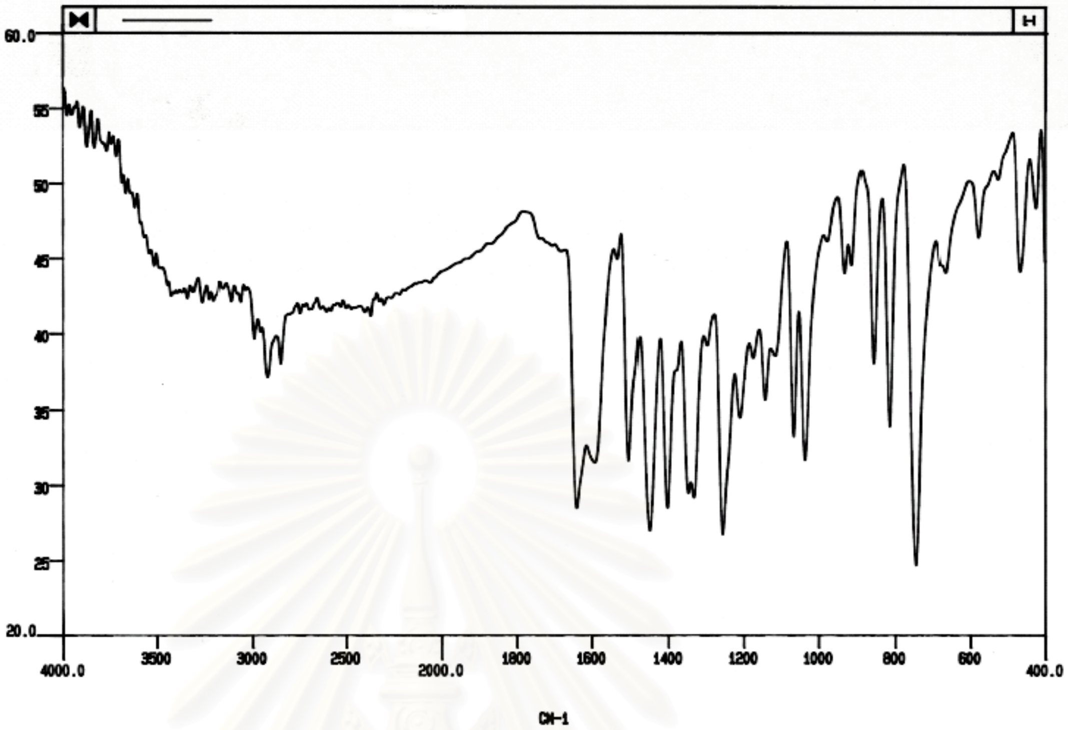


Figure 142 IR Spectrum of compound ME15 (KBr disc)

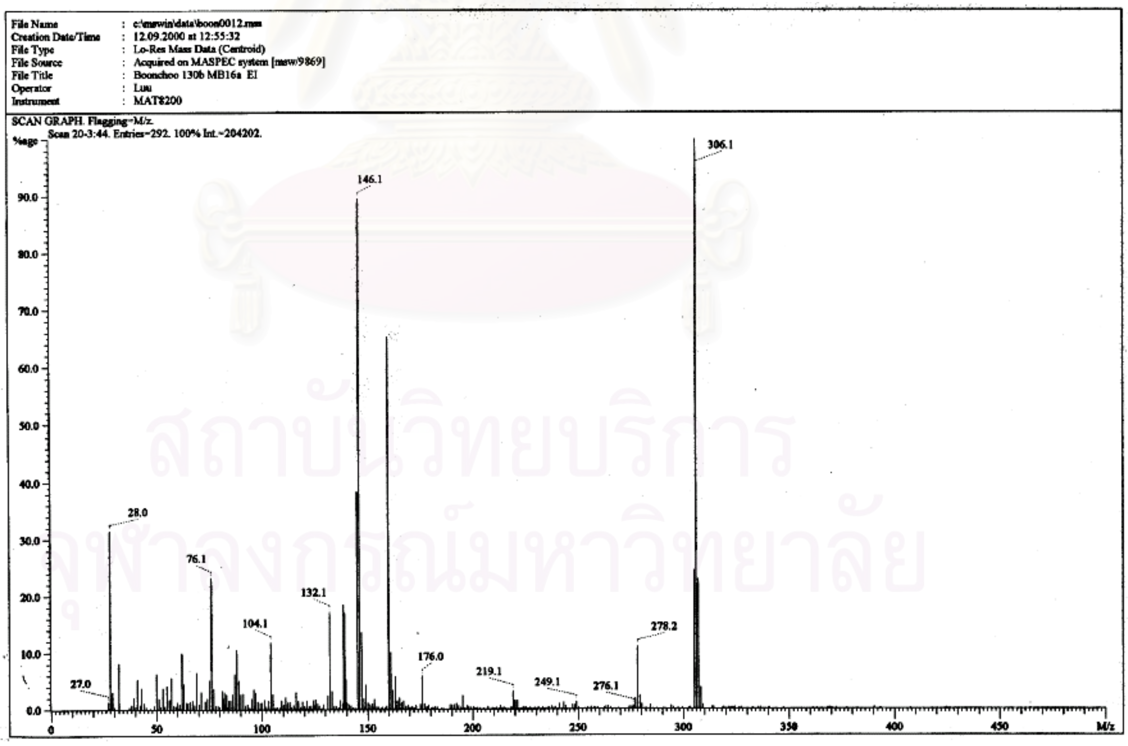

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Figure 143 EI Mass spectrum of compound ME15

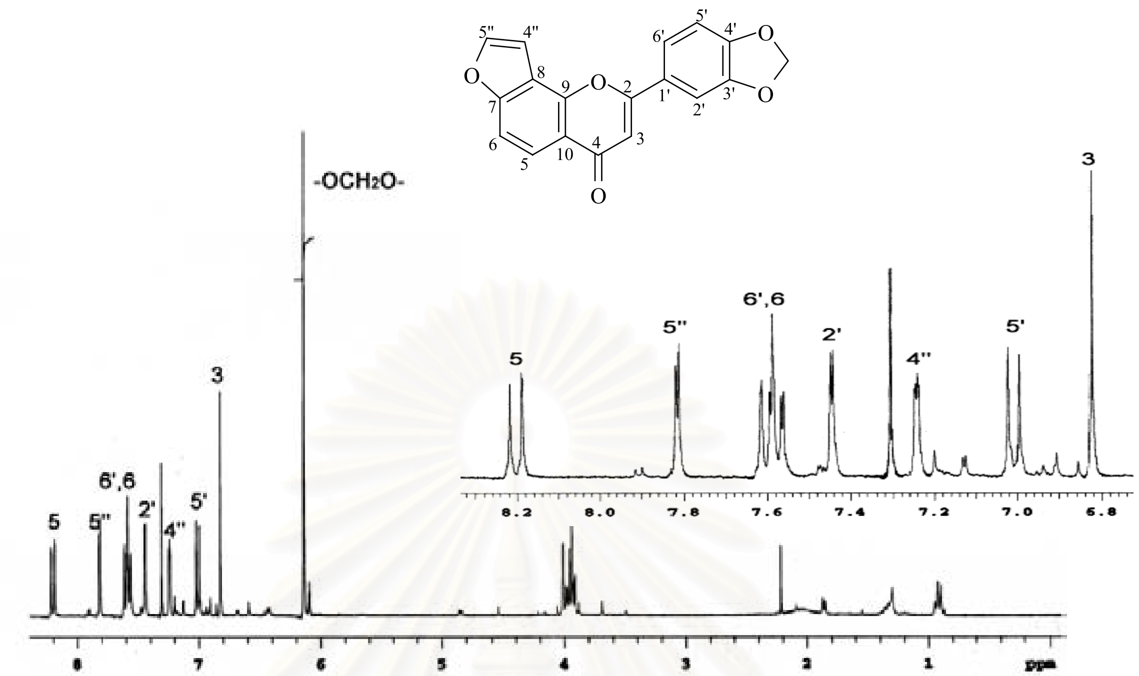

Figure 144 ^{1}H NMR (300 MHz) Spectrum of compound ME15 ($CDCl_3$)

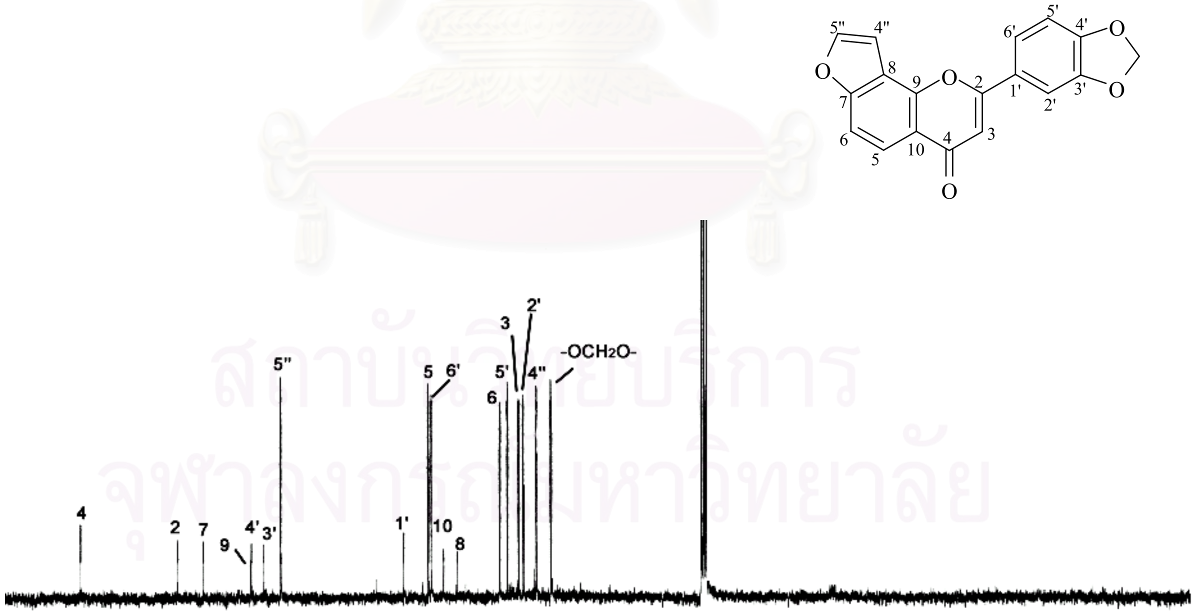

Figure 145 ^{13}C NMR (75 MHz) Spectrum of compound ME15 ($CDCl_3$)

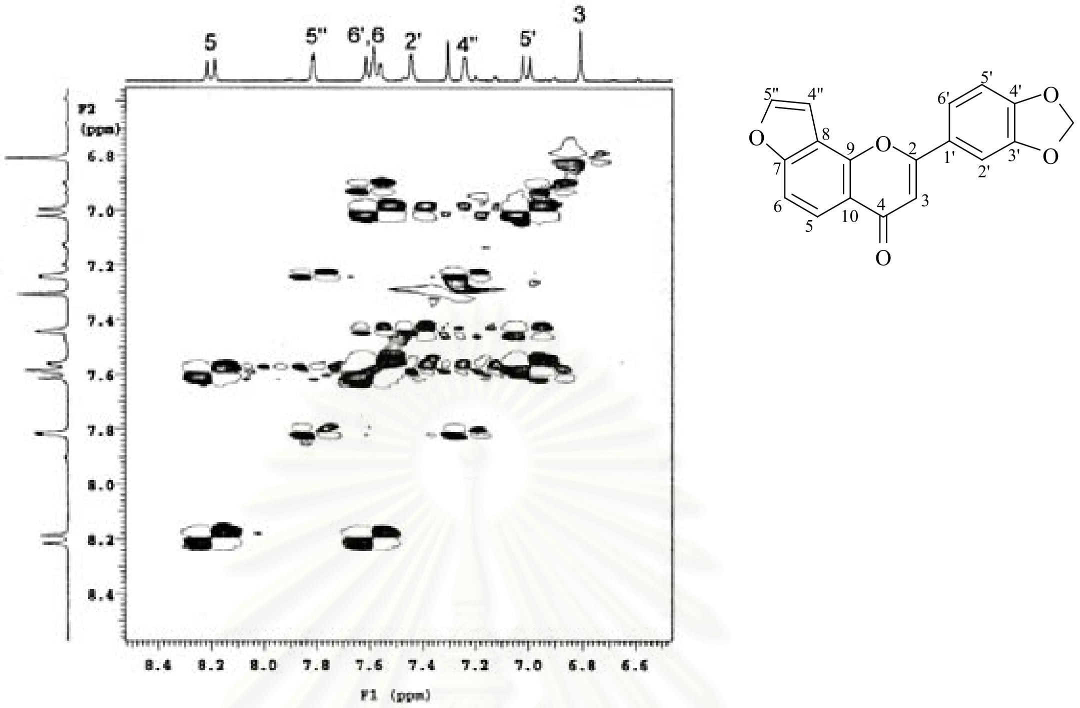

Figure 146 ^{1}H-^{1}H COSY Spectrum of compound ME15 (CDCl$_3$)

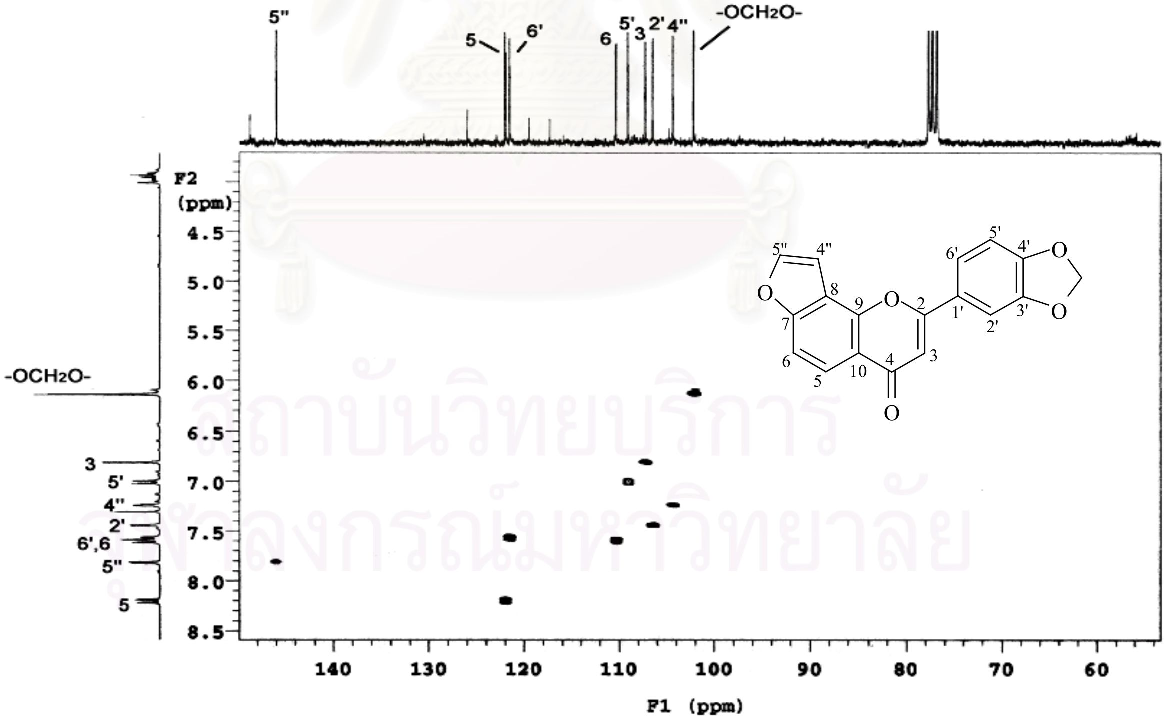

Figure 147 HSQC Spectrum of compound ME15 (CDCl$_3$)

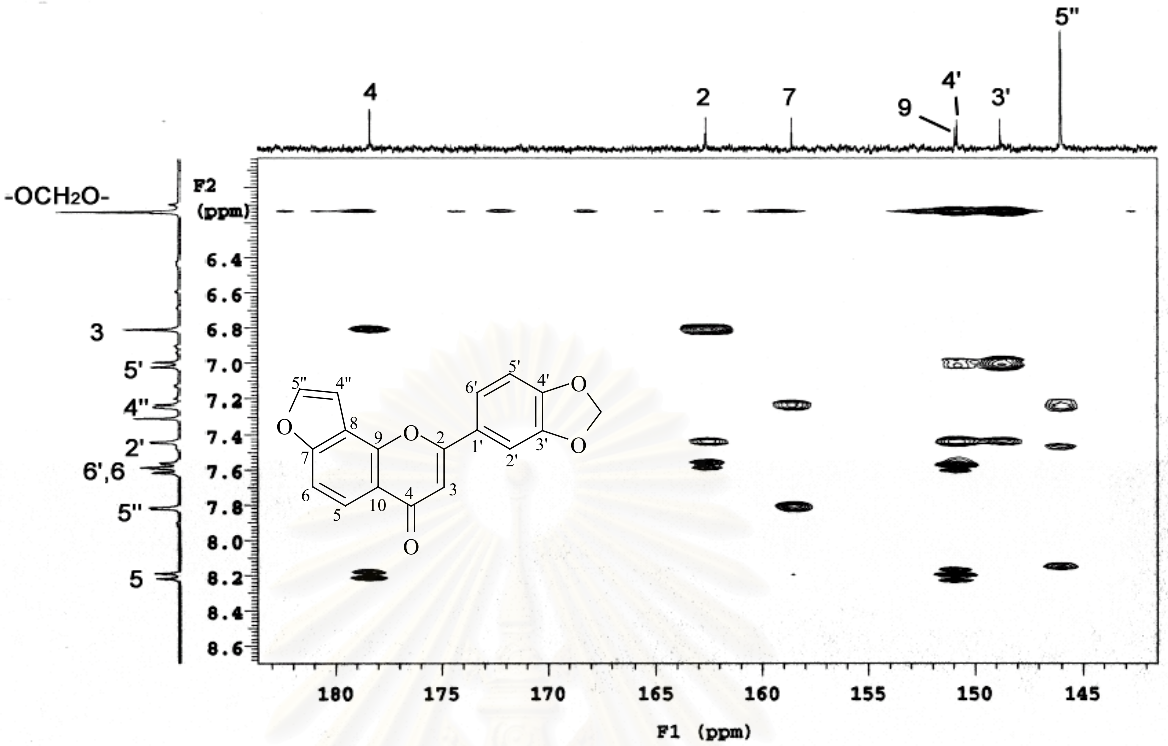

Figure 148 HMBC Spectrum of compound ME15 ($CDCl_3$) [δ_H 5.8-8.7 ppm, δ_C 141-184 ppm]

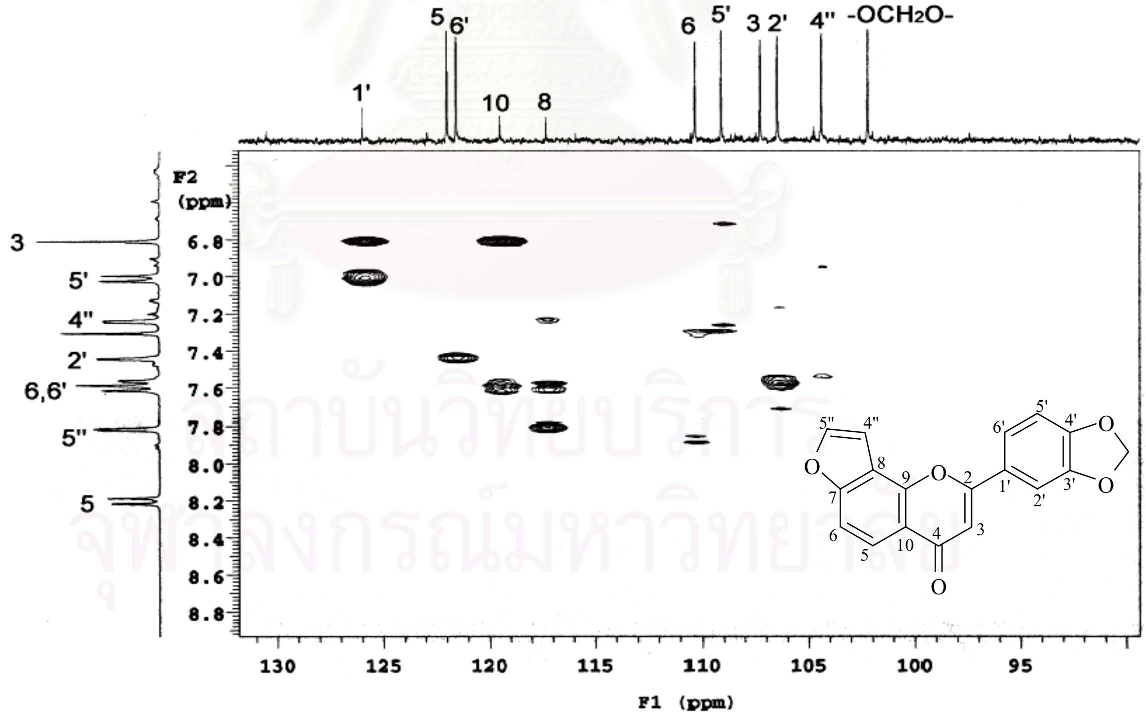

Figure 149 HMBC Spectrum of compound ME15 ($CDCl_3$) [δ_H 6.3-8.9 ppm, δ_C 89-132 ppm]

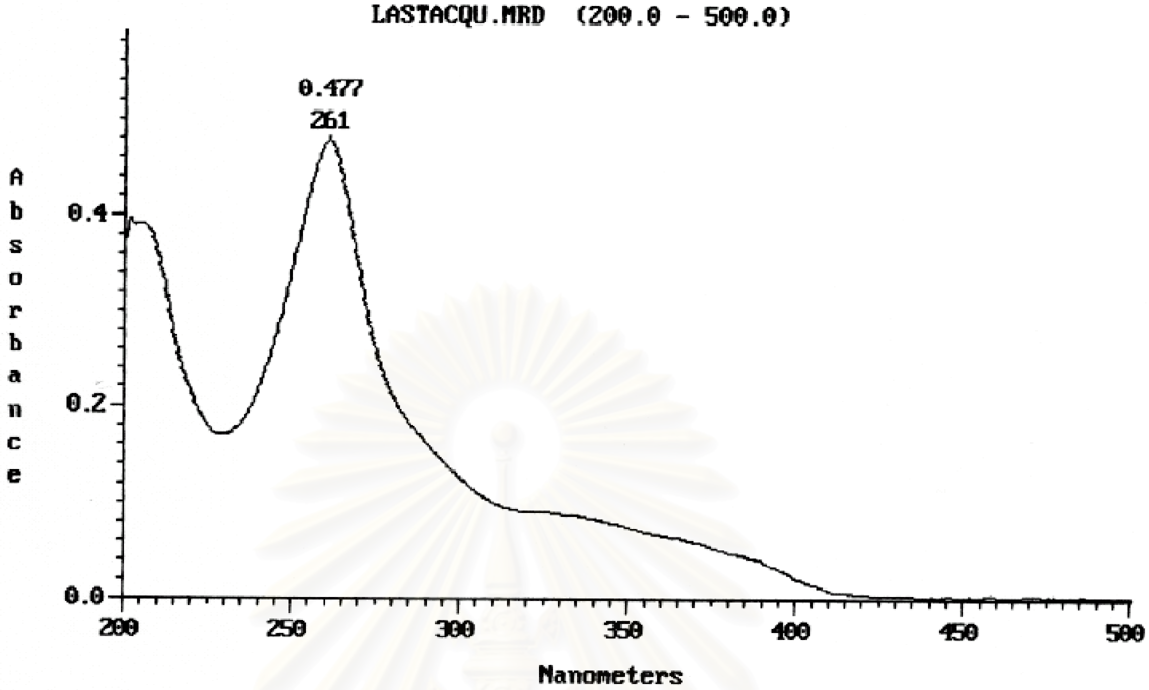

Figure 150 UV Spectrum of compound ME16 (methanol)

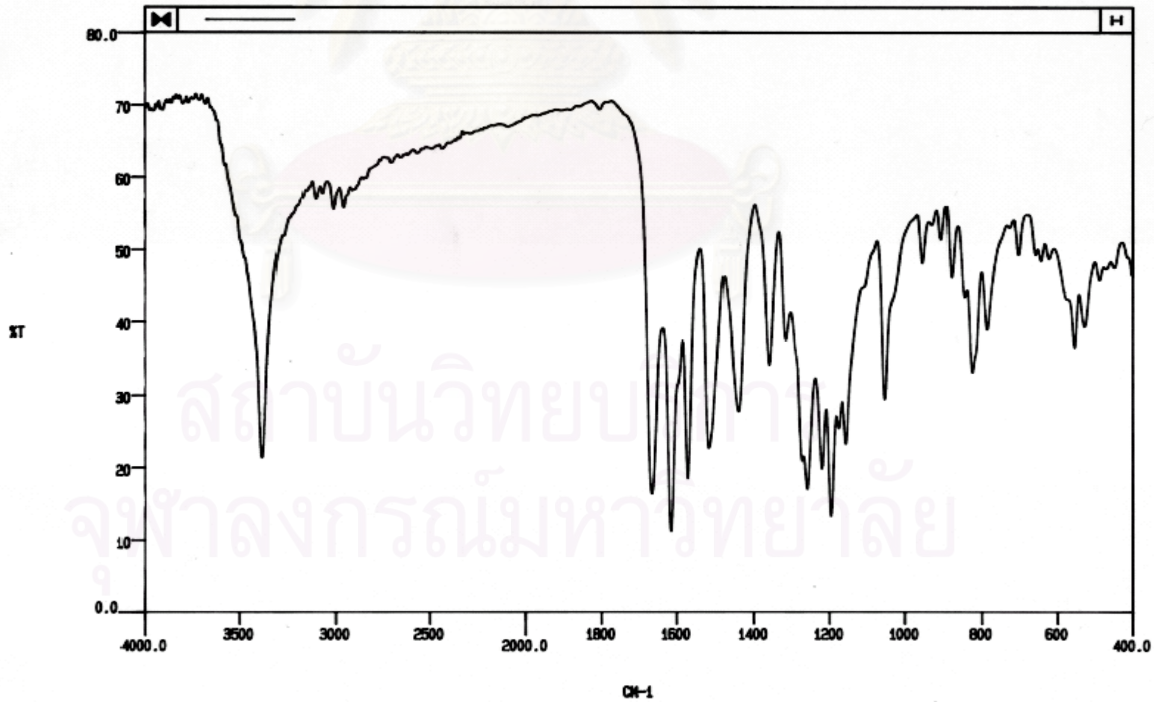

Figure 151 IR Spectrum of compound ME16 (KBr disc)

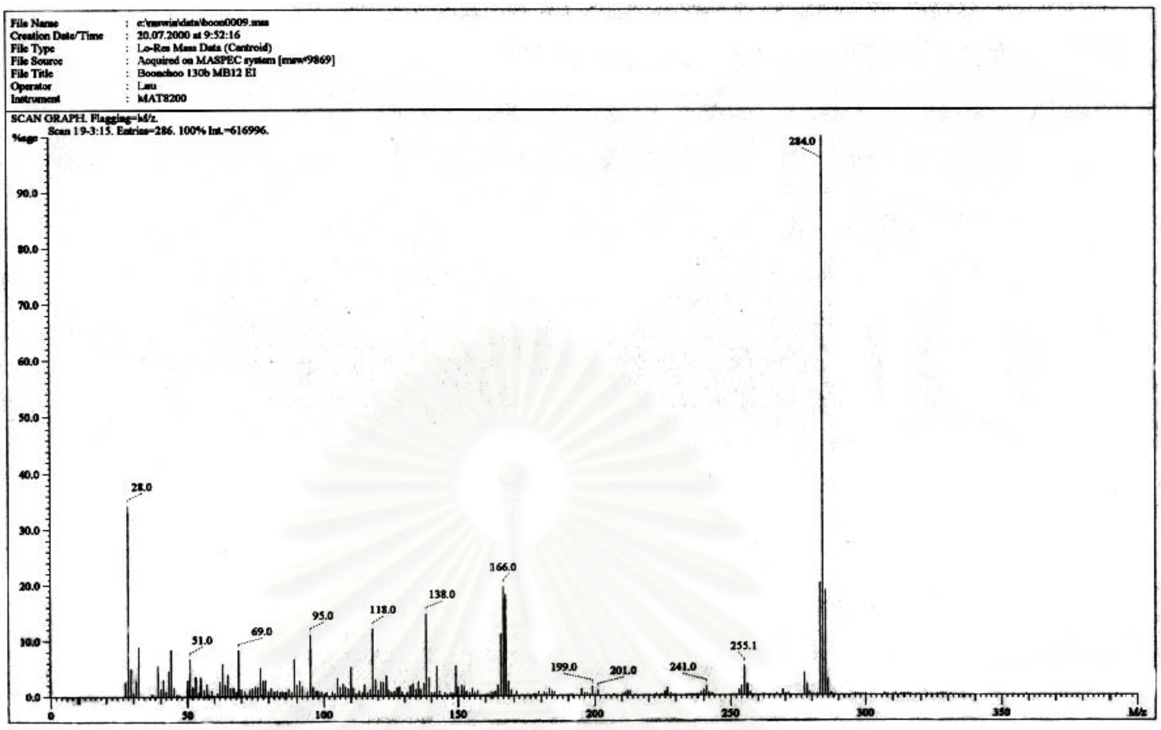

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Figure 152 EI Mass spectrum of compound ME16

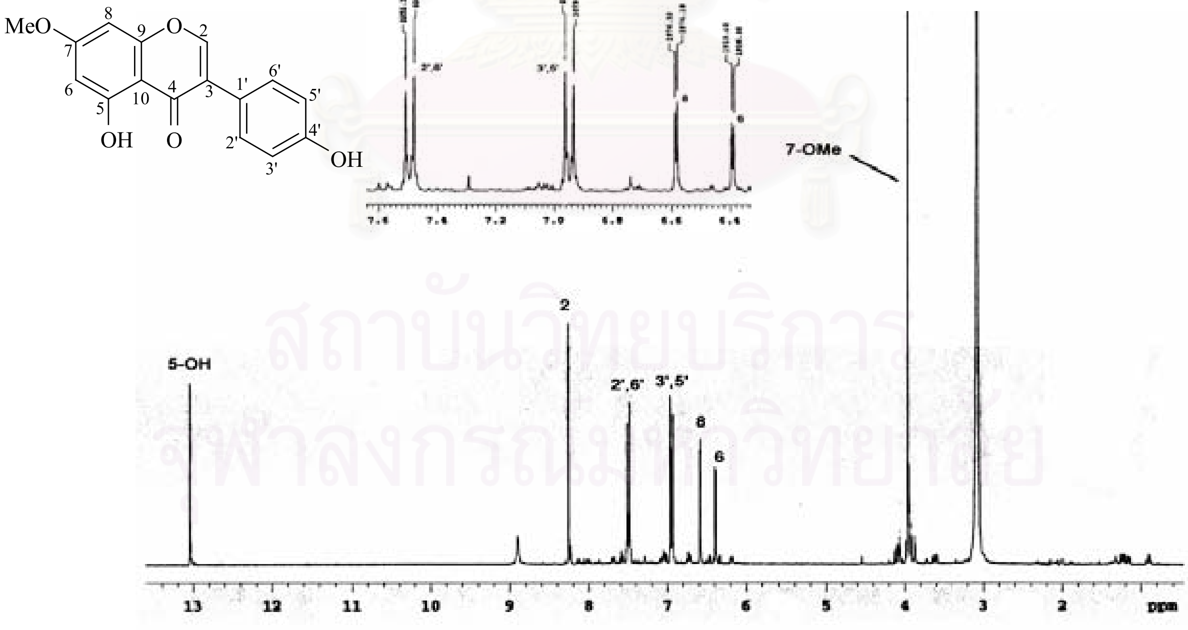
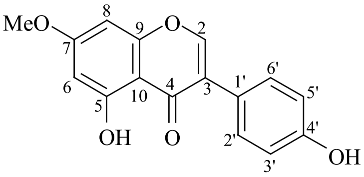


Figure 153 ^{1}H NMR (300 MHz) Spectrum of compound ME16 (acetone-d_6)



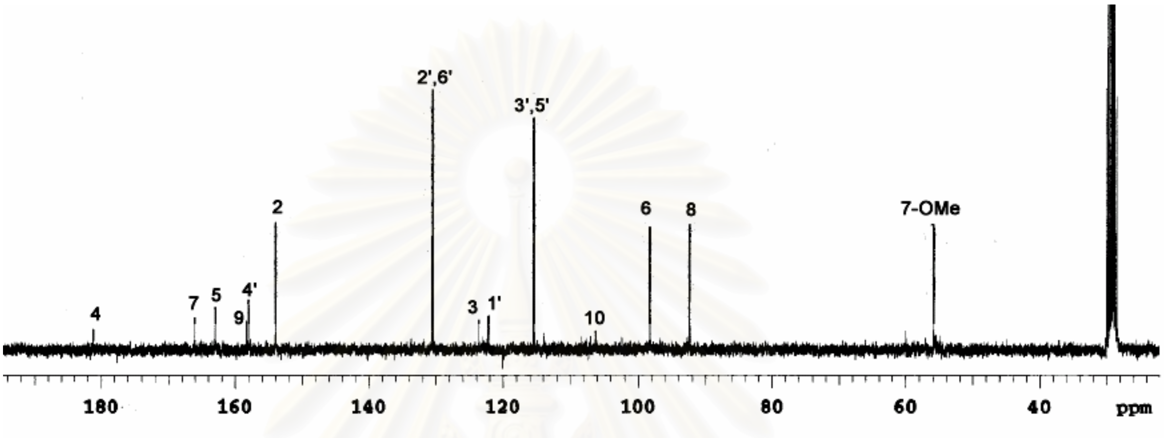


Figure 154 ^{13}C NMR (75 MHz) Spectrum of compound ME16 (acetone-d_6)

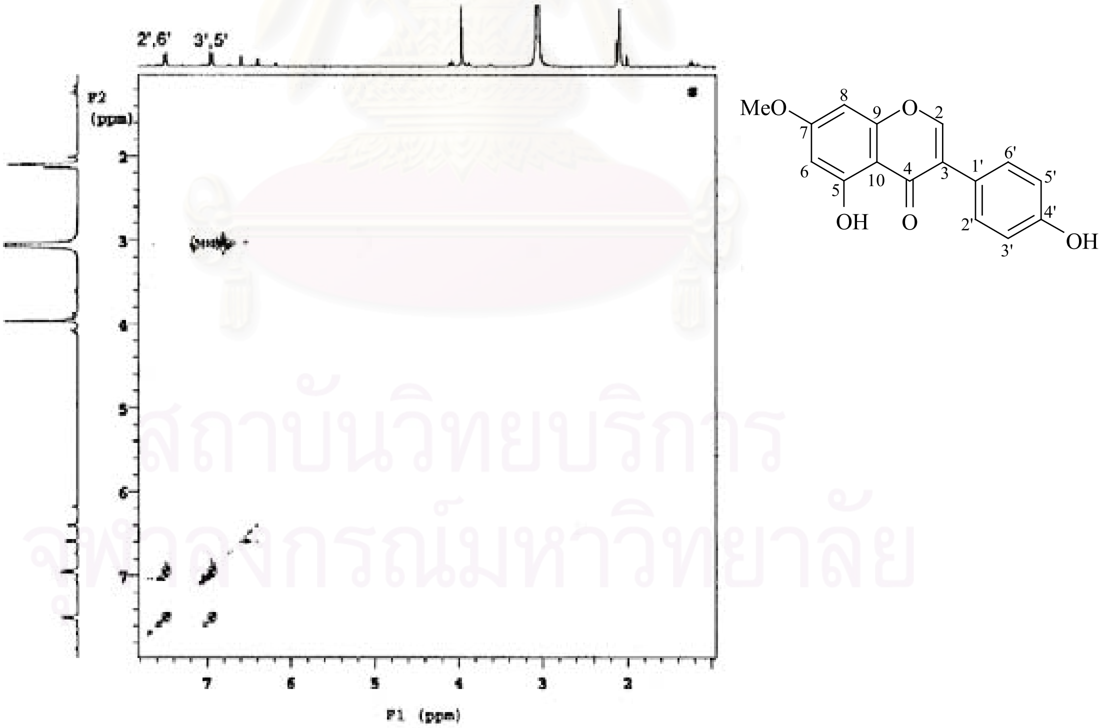


Figure 155 ^{1}H-^{1}H COSY Spectrum of compound ME16 (acetone-d_6)

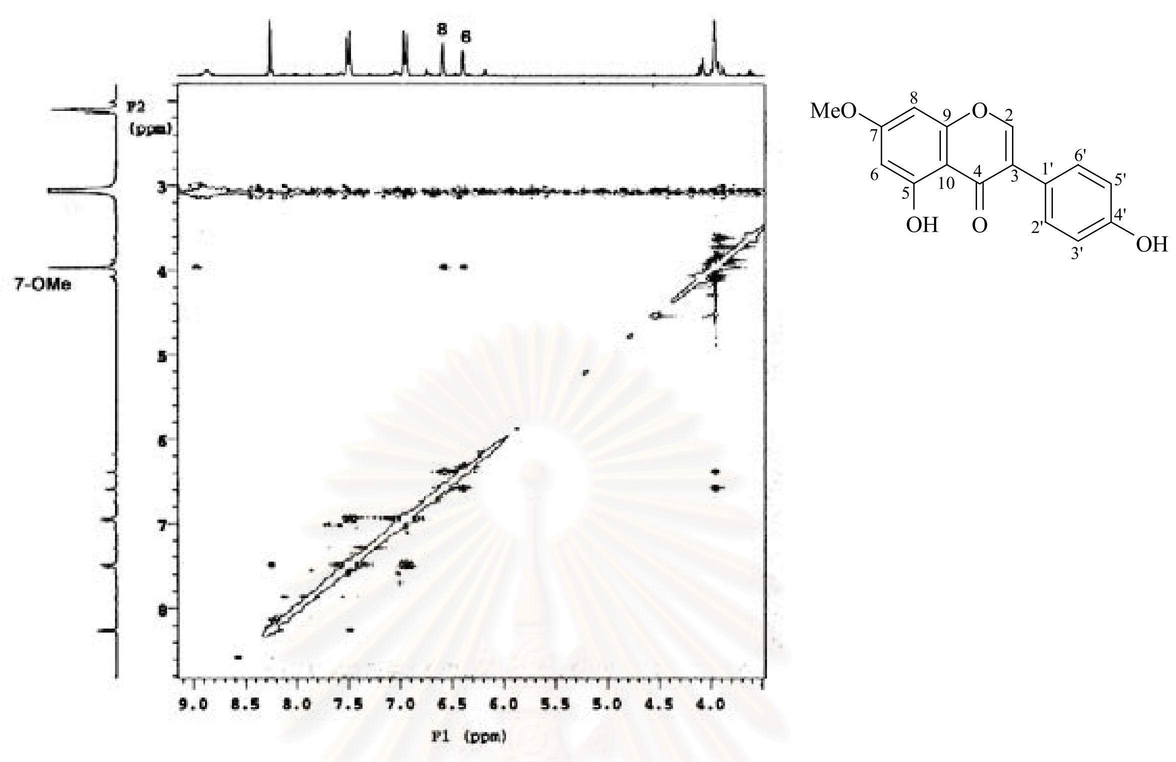


Figure 156 NOESY Spectrum of compound ME16 (acetone-d_6)

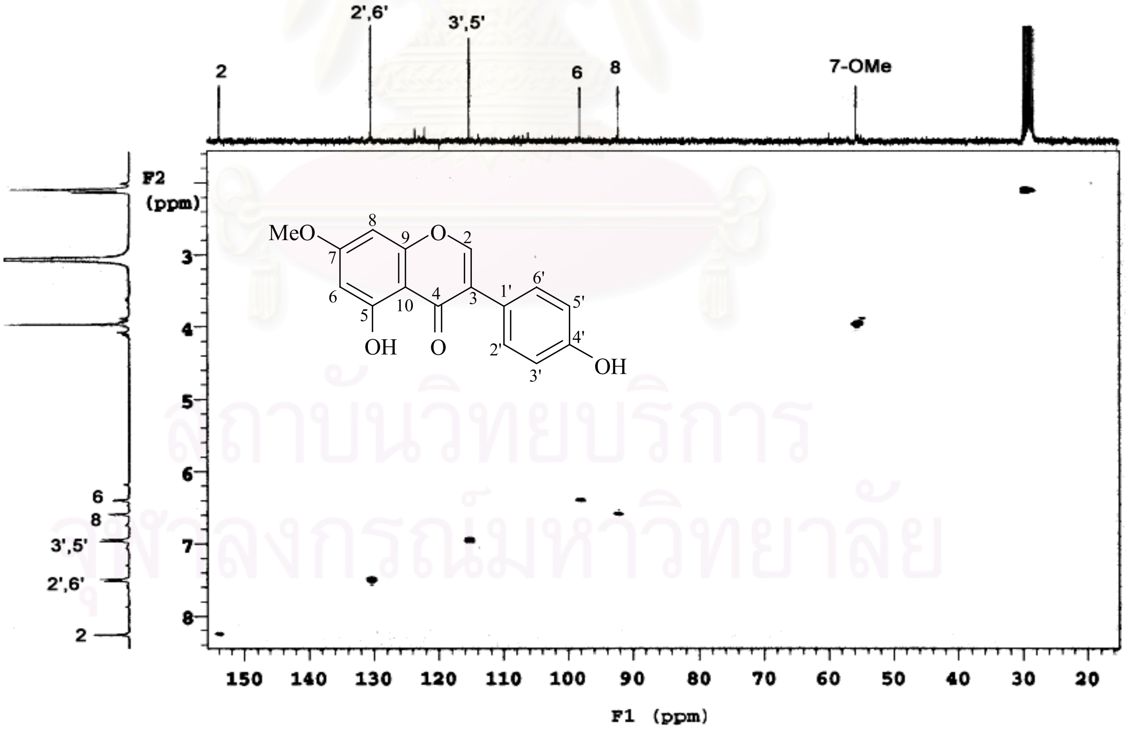


Figure 157 HSQC Spectrum of compound ME16 (acetone-d_6)

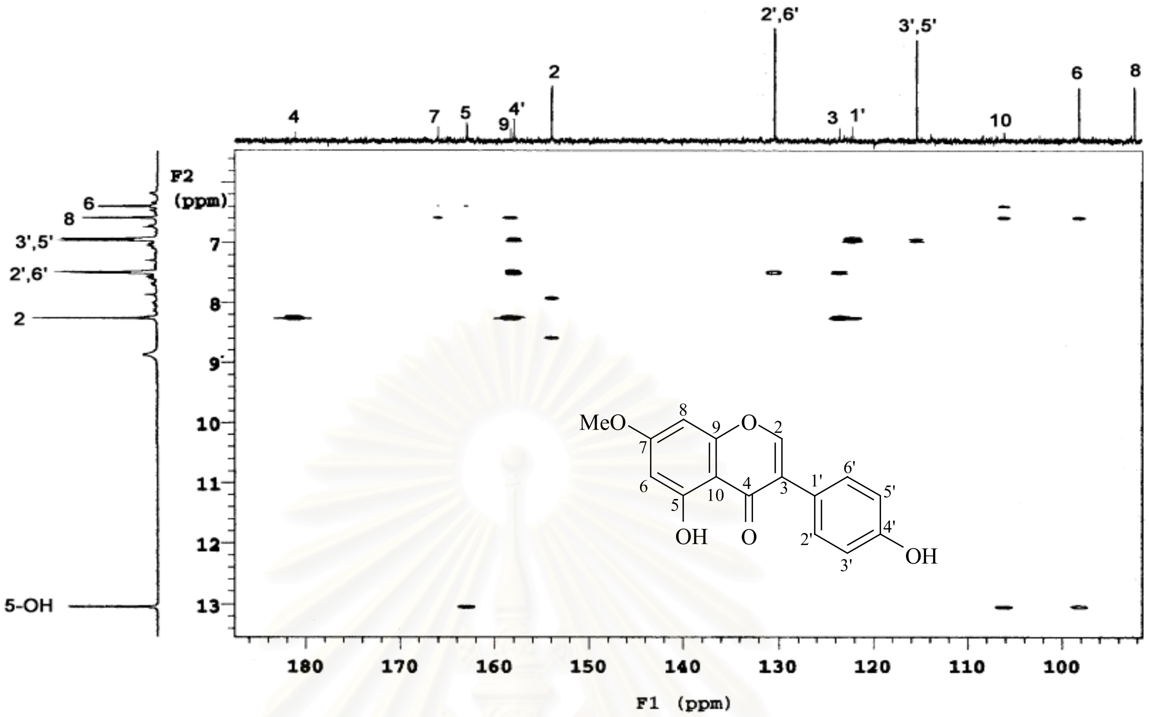


Figure 158 HMBC Spectrum of compound ME16 (acetone-d_6)

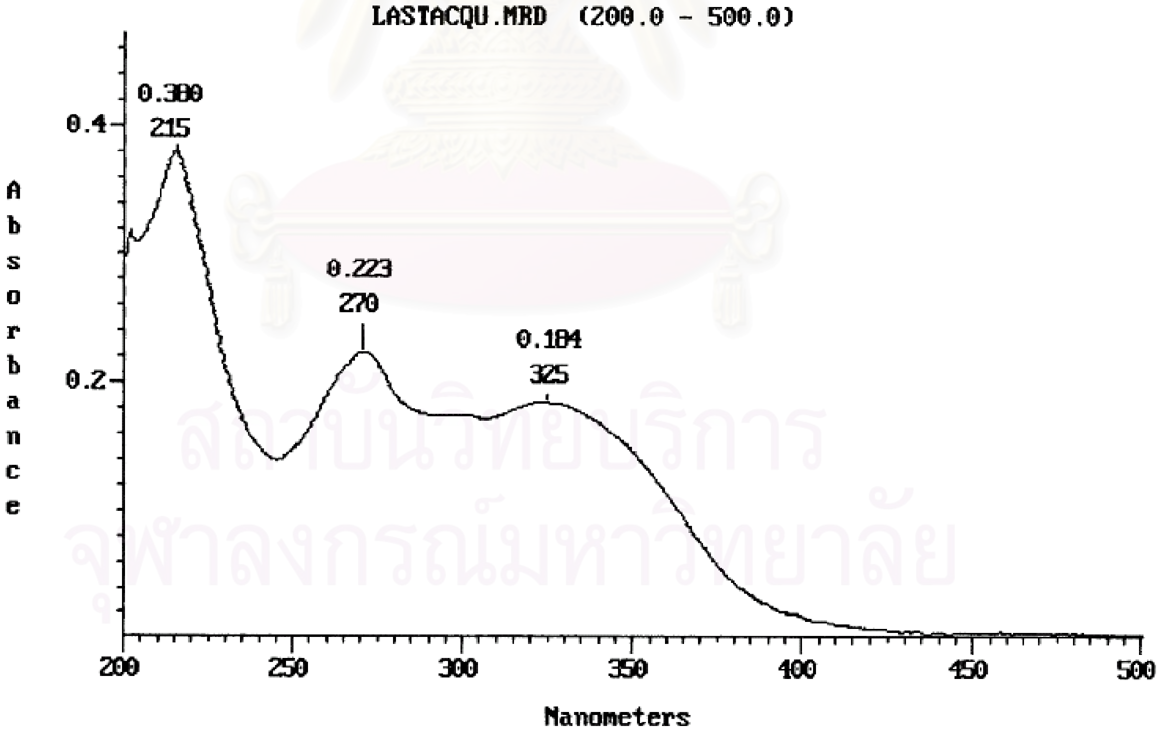


Figure 159 UV Spectrum of compound ME17 (methanol)

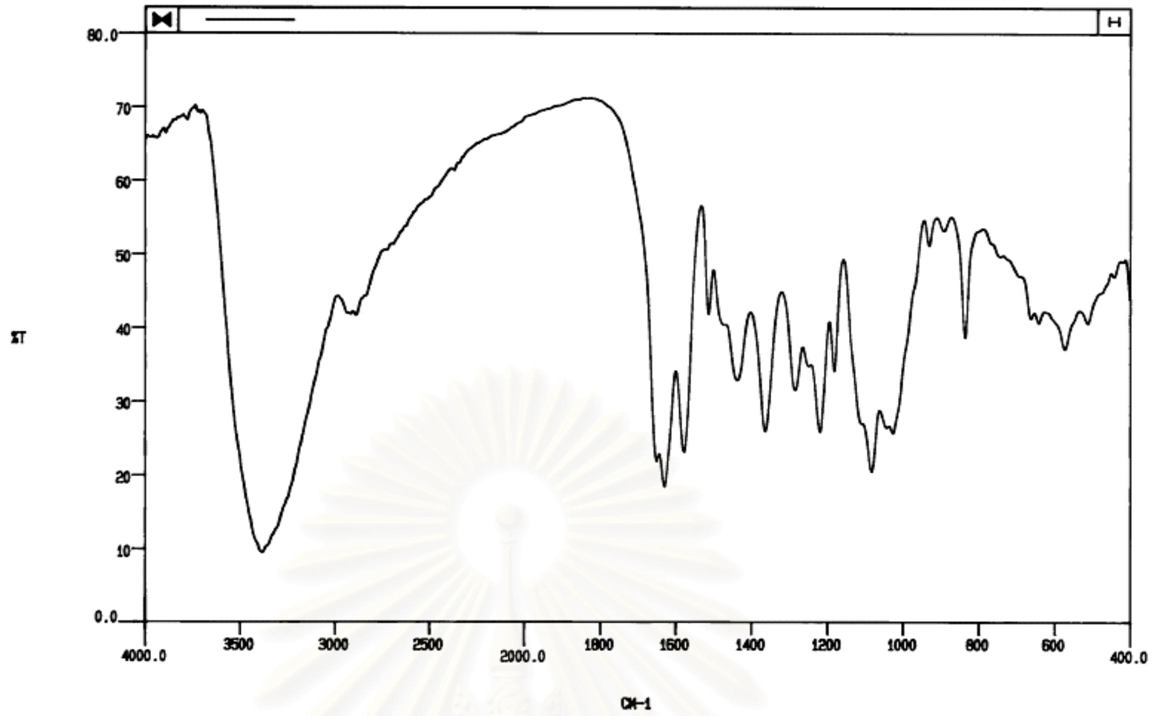


Figure 160 IR Spectrum of compound ME17 (KBr disc)

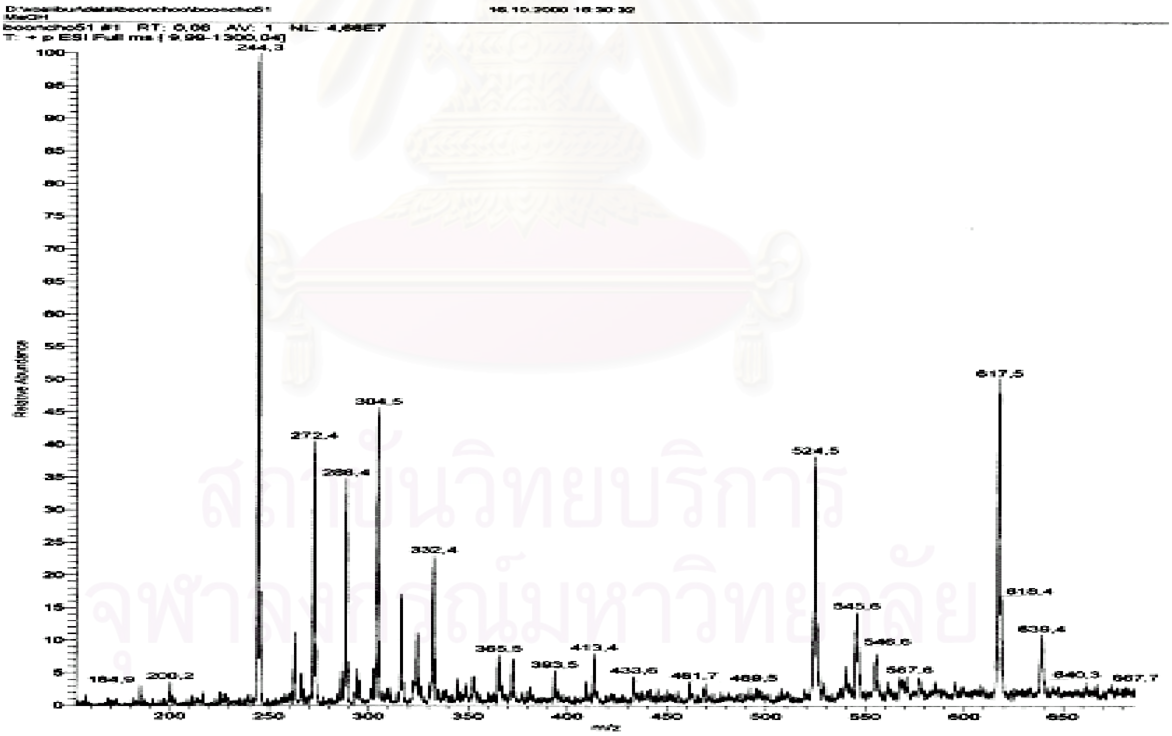


Figure 161 ESI Mass spectrum (positive ion mode) of compound ME17

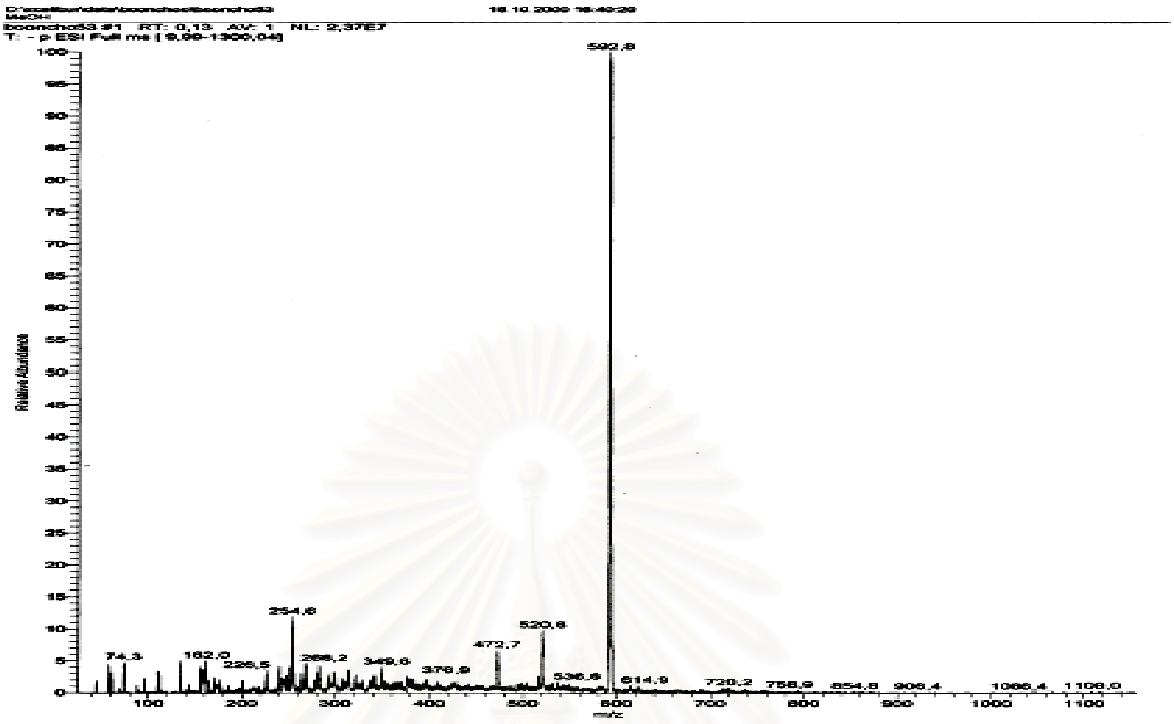


Figure 162 ESI Mass spectrum (negative ion mode) of compound ME17

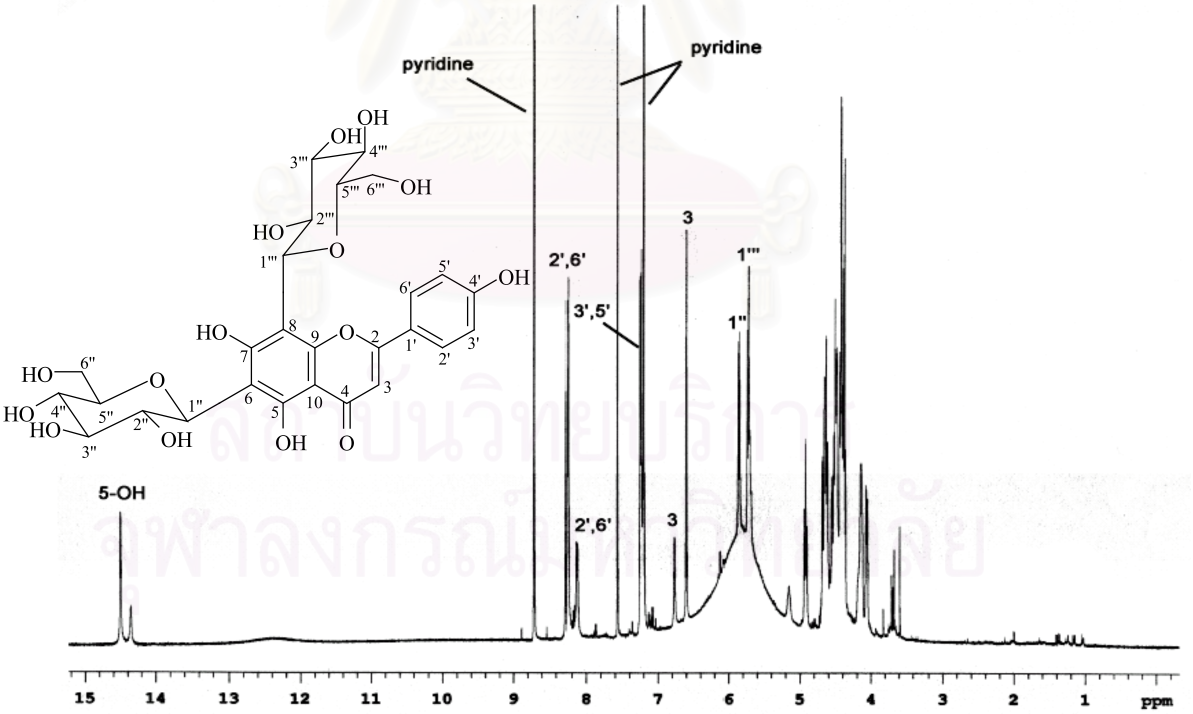


Figure 163 ^{1}H NMR (500 MHz) Spectrum of compound ME17 (pyridine-d_5)

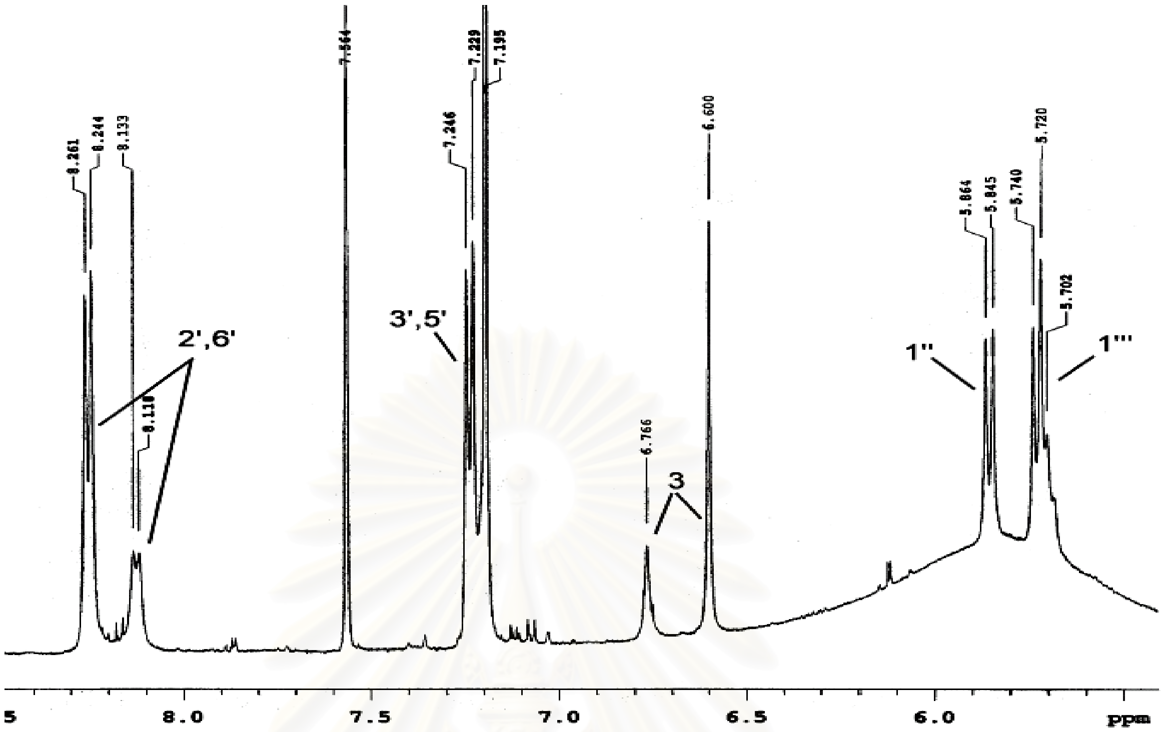


Figure 164 ^{1}H NMR (500 MHz) Spectrum of compound ME17 (pyridine-d_5, 5.5-8.5 ppm)

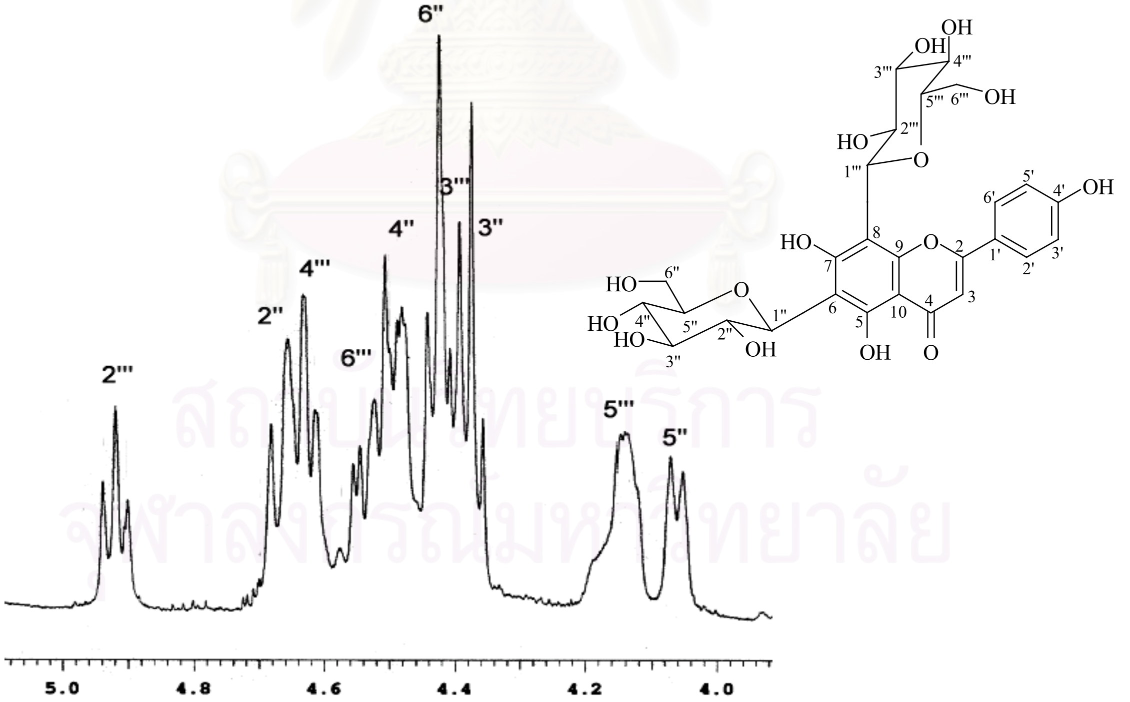


Figure 165 ^{1}H NMR (500 MHz) Spectrum of compound ME17 (pyridine-d_5, 3.9-5.1 ppm)

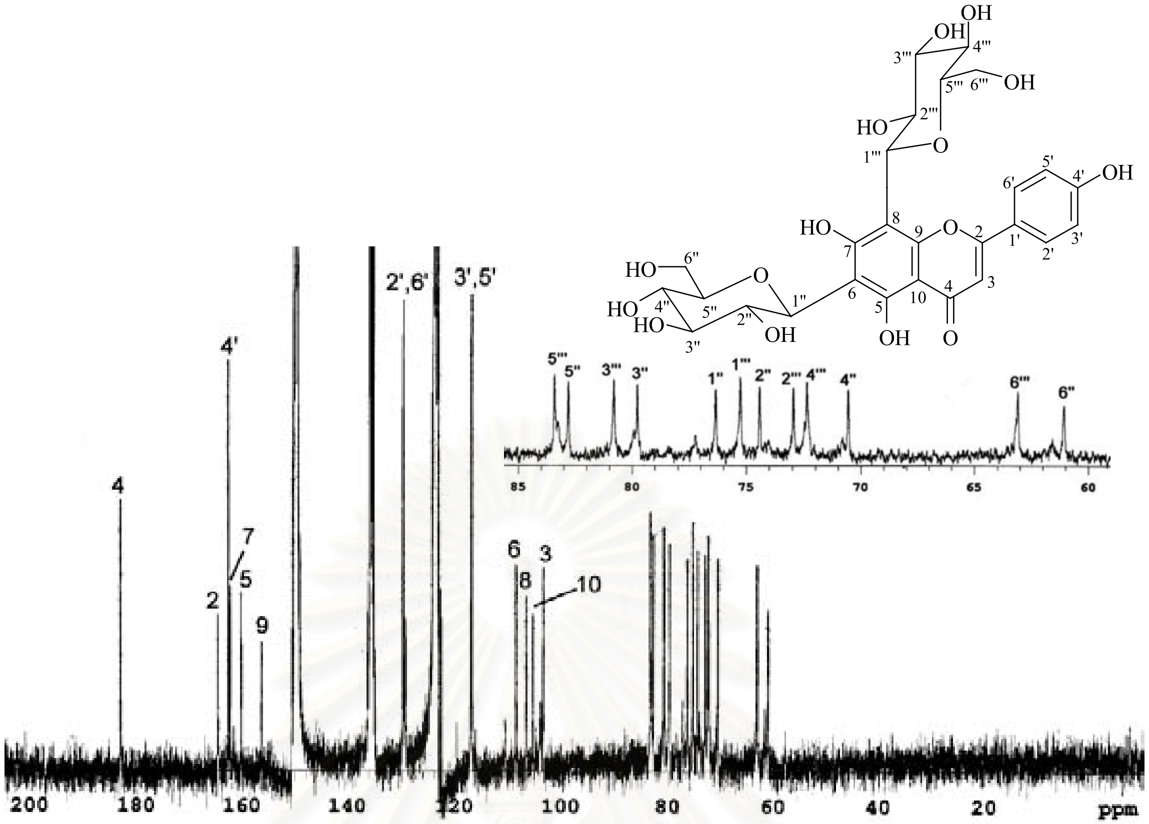


Figure 166 ^{13}C NMR (75 MHz) Spectrum of compound ME17 (pyridine-d_5)

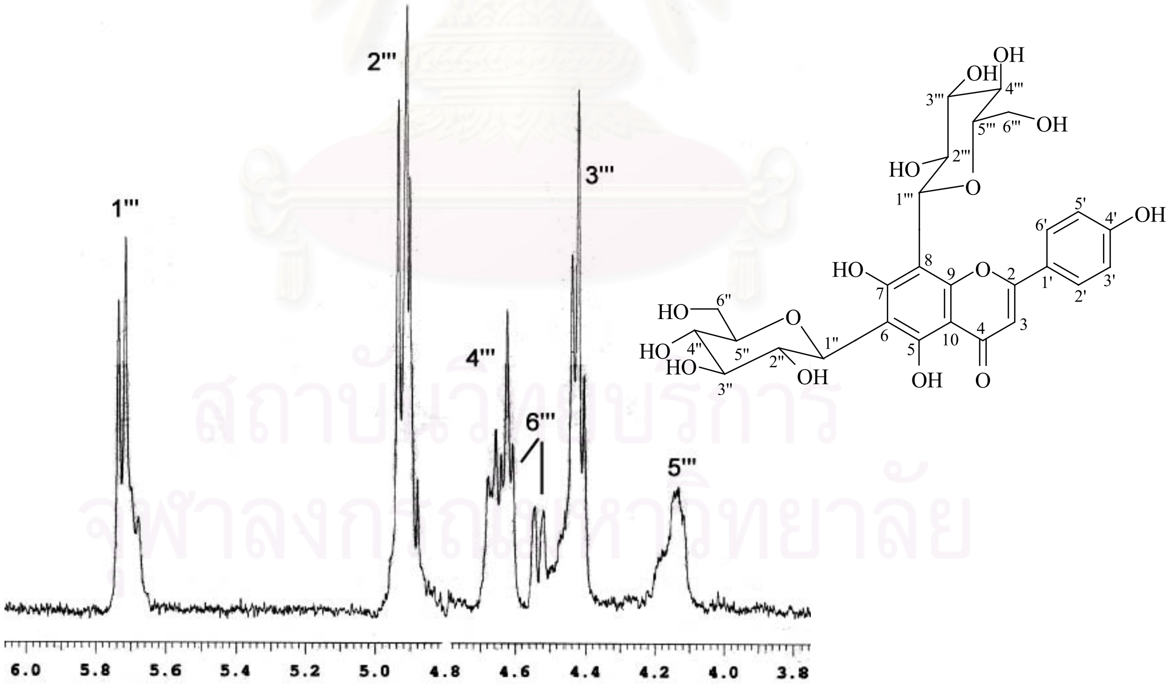


Figure 167 TOCSY Spectrum of compound ME17 (1st glucose moiety, pyridine-d_5)

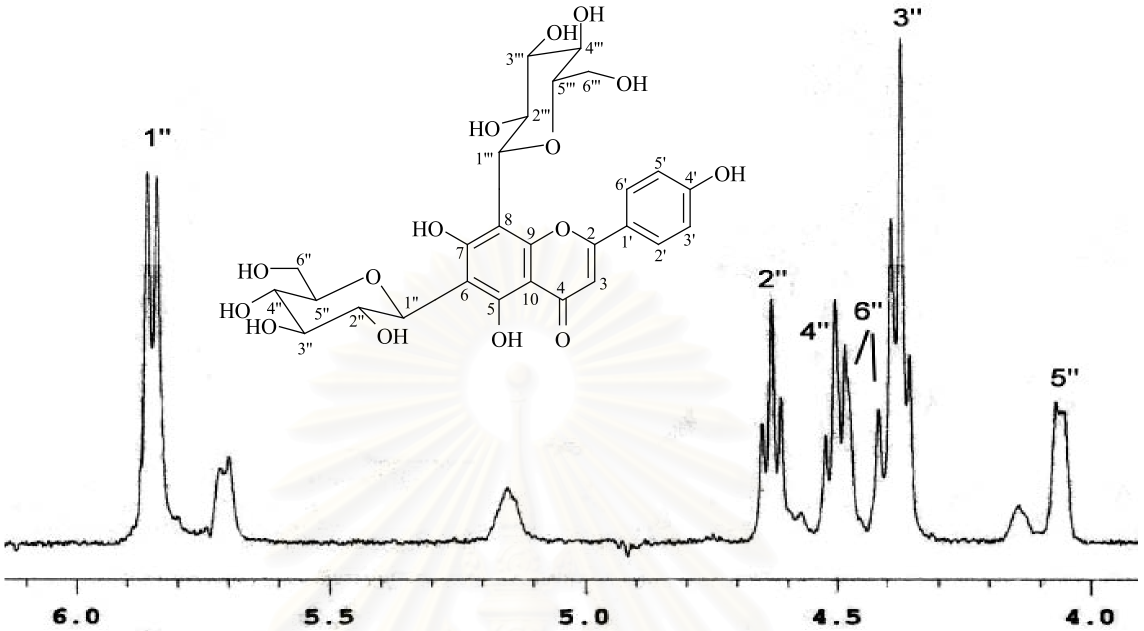


Figure 168 TOCSY Spectrum of compound ME17 (2^{nd} glucose moiety, pyridine-d_5)

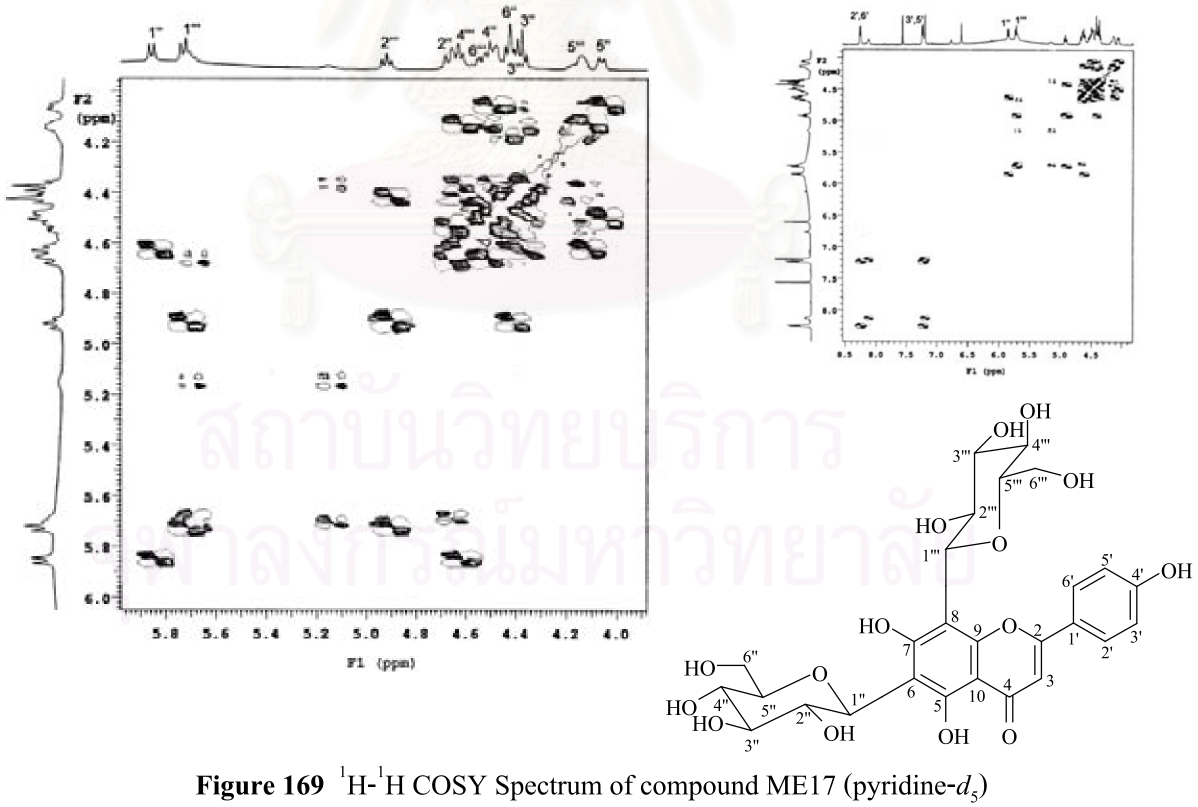


Figure 169 ^1H-^1H COSY Spectrum of compound ME17 (pyridine-d_5)

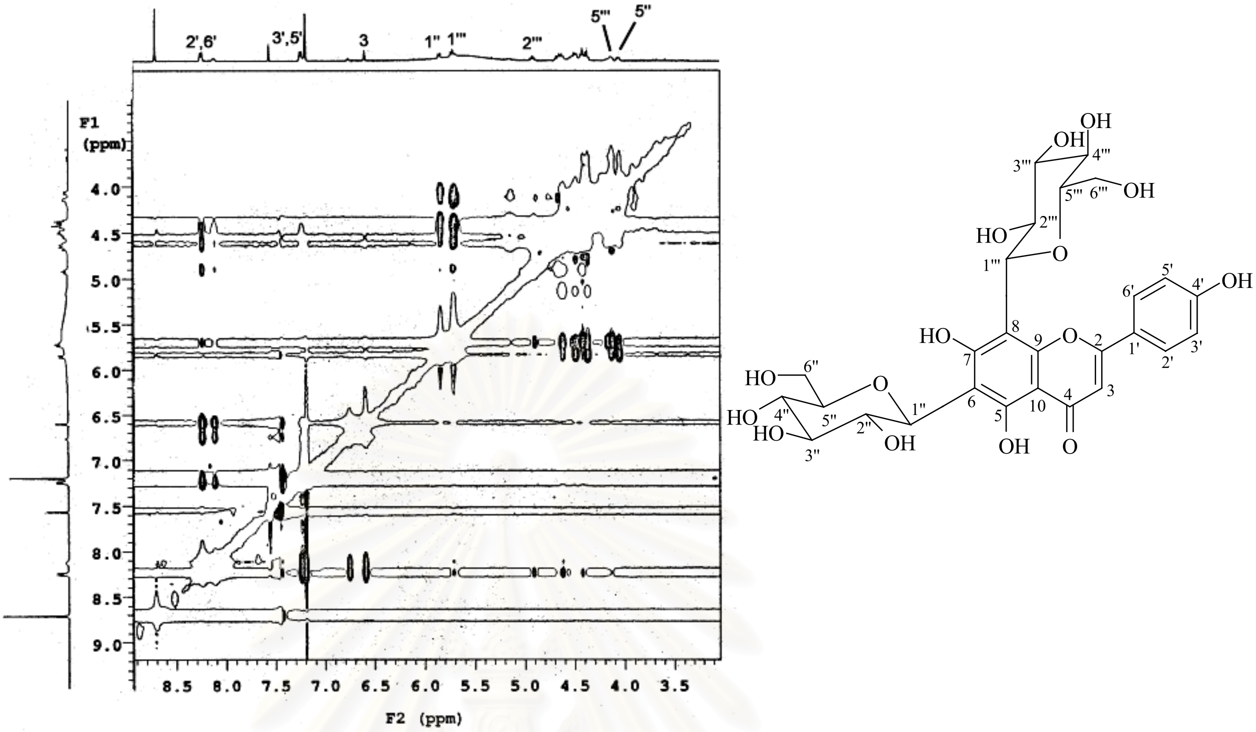


Figure 170 ROESY Spectrum of compound ME17 (pyridine-d_5)

Figure 171 HSQC-TOCSY Spectrum of compound ME17 (pyridine-d_5)

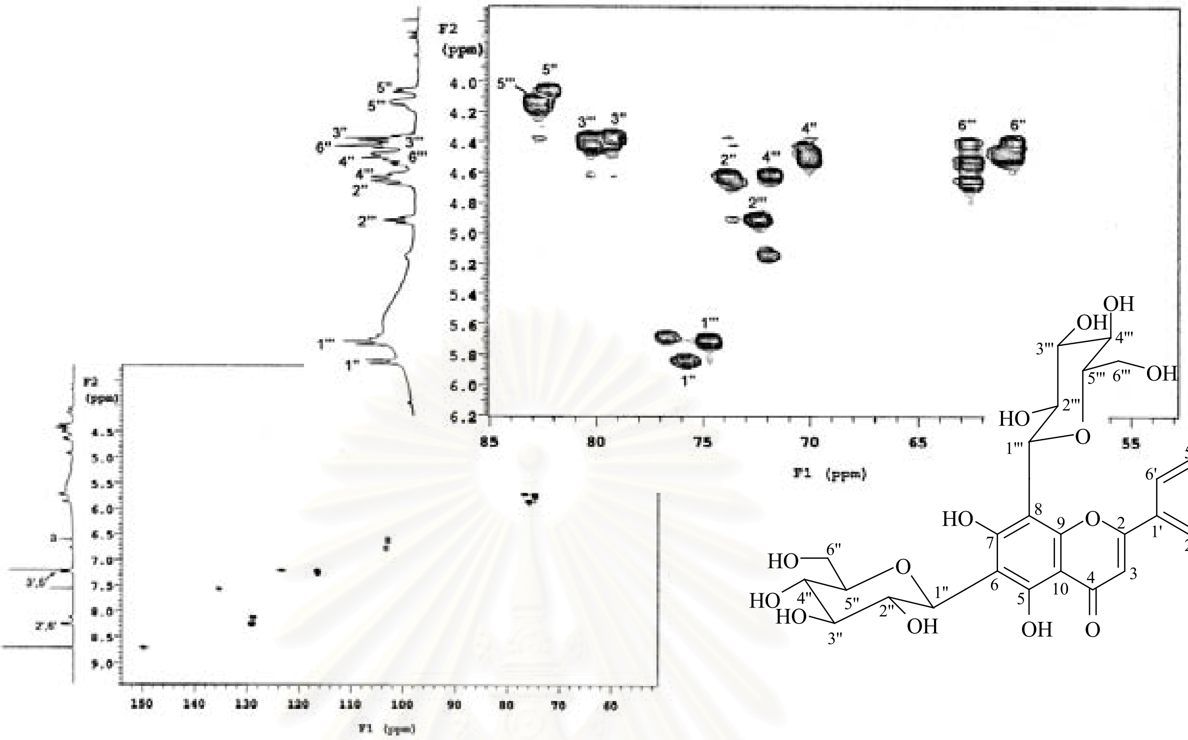


Figure 172 HSQC Spectrum of compound ME17 (pyridine-d_5)

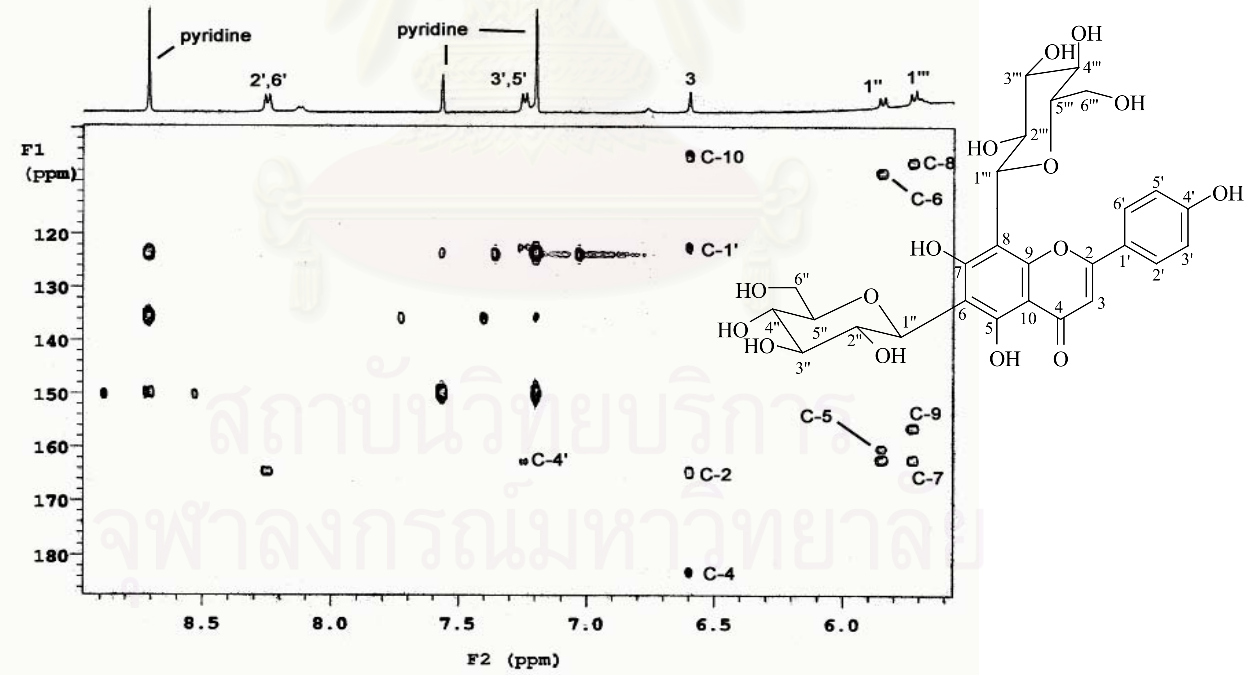


Figure 173 HMBC Spectrum of compound ME17 (pyridine-d_5) [δ_H 5.5-9.0 ppm, δ_C 100-190 ppm]

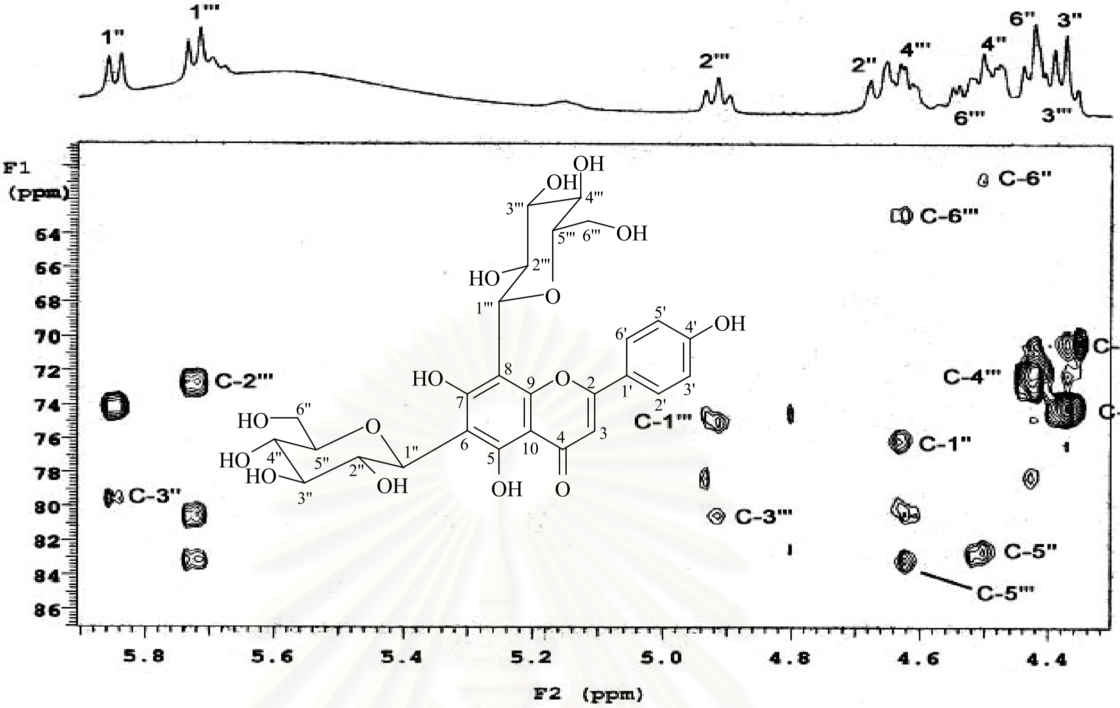


Figure 174 HMBC Spectrum of compound ME17 (pyridine-*d*$_5$) [δ_H 4.3-5.9 ppm, δ_C 60-87 ppm]

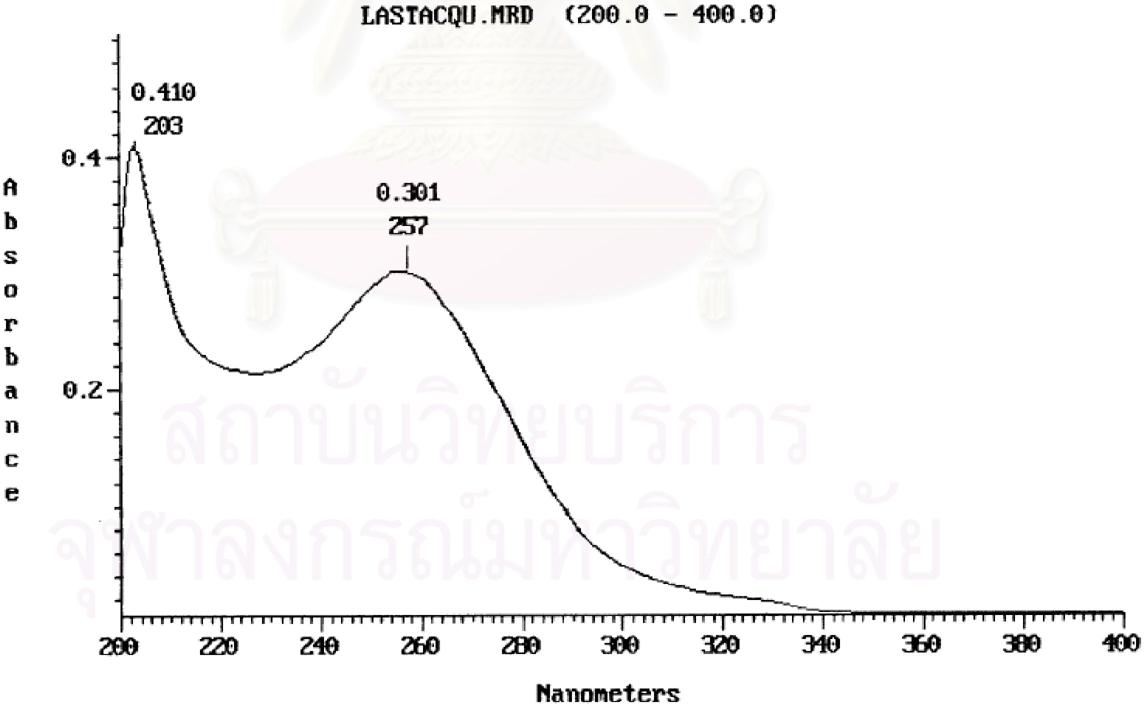


Figure 175 UV Spectrum of compound ME18 (methanol)

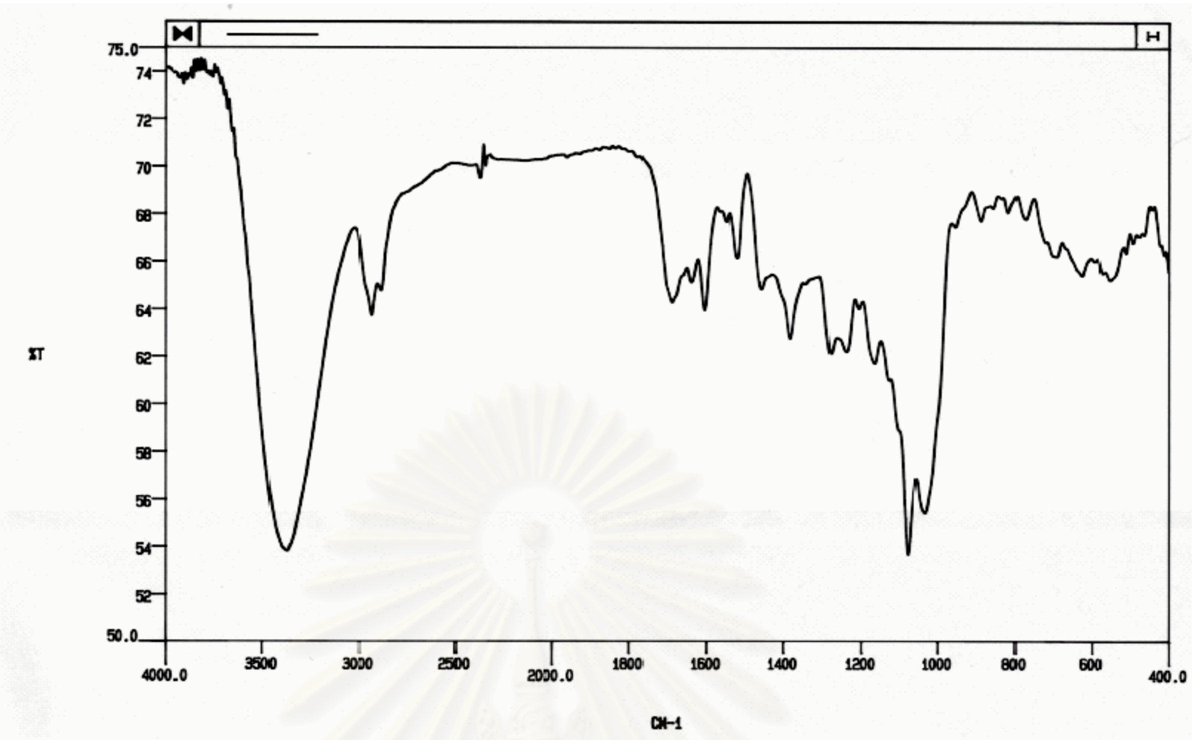


Figure 176 IR Spectrum of compound ME18 (KBr disc)

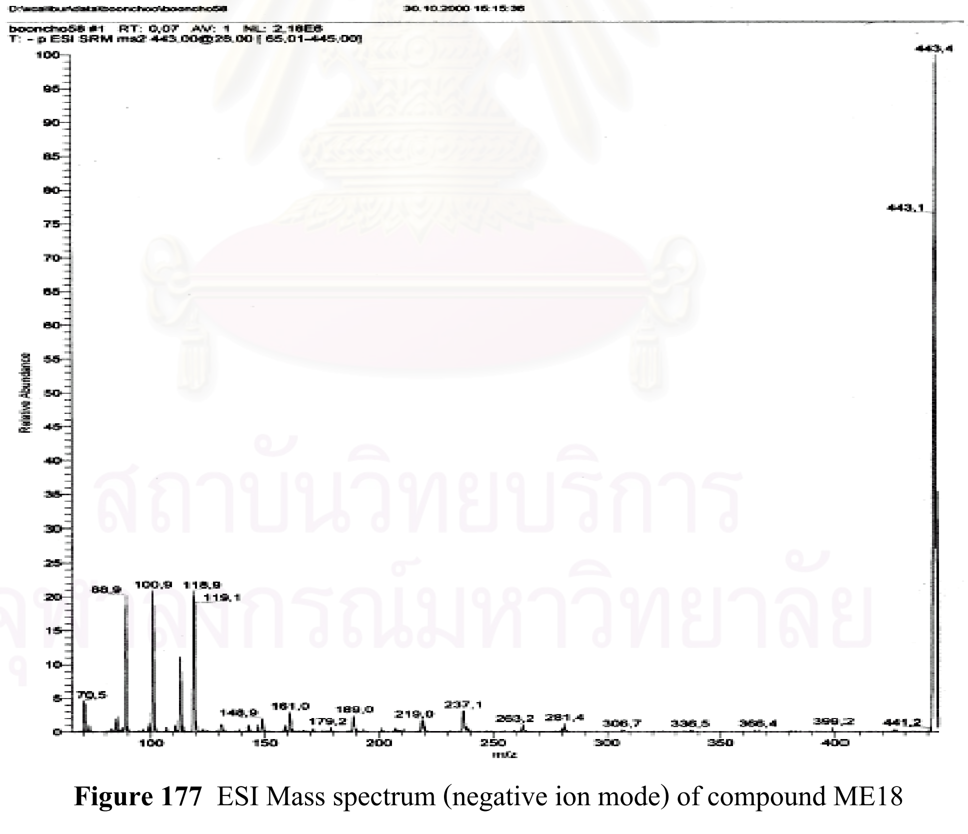


Figure 177 ESI Mass spectrum (negative ion mode) of compound ME18

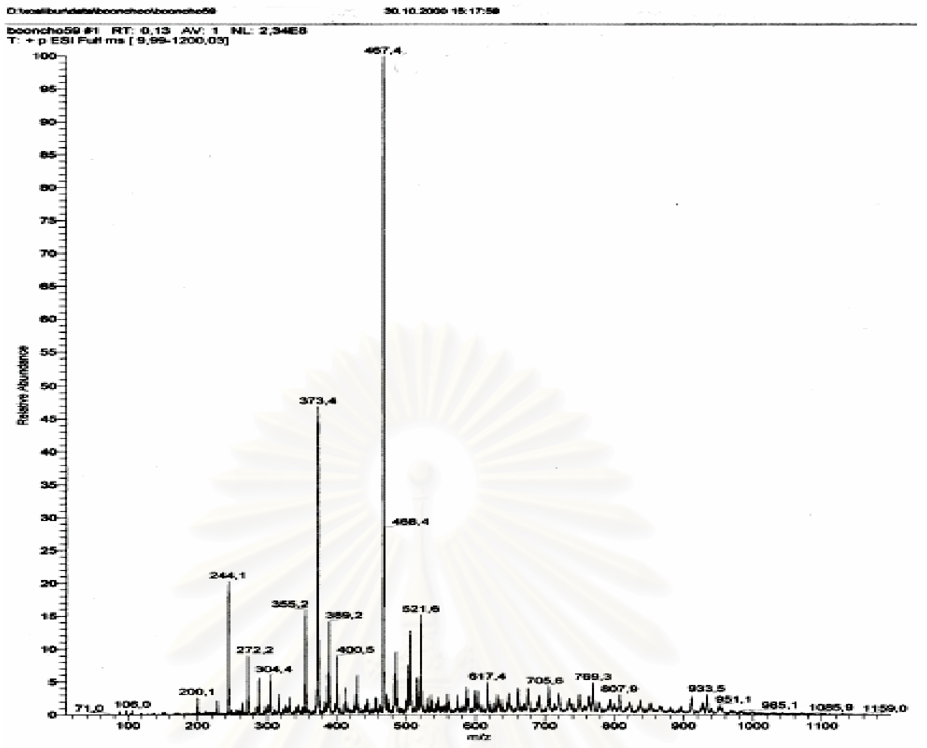


Figure 178 ESI Mass spectrum (positive ion mode) of compound ME18

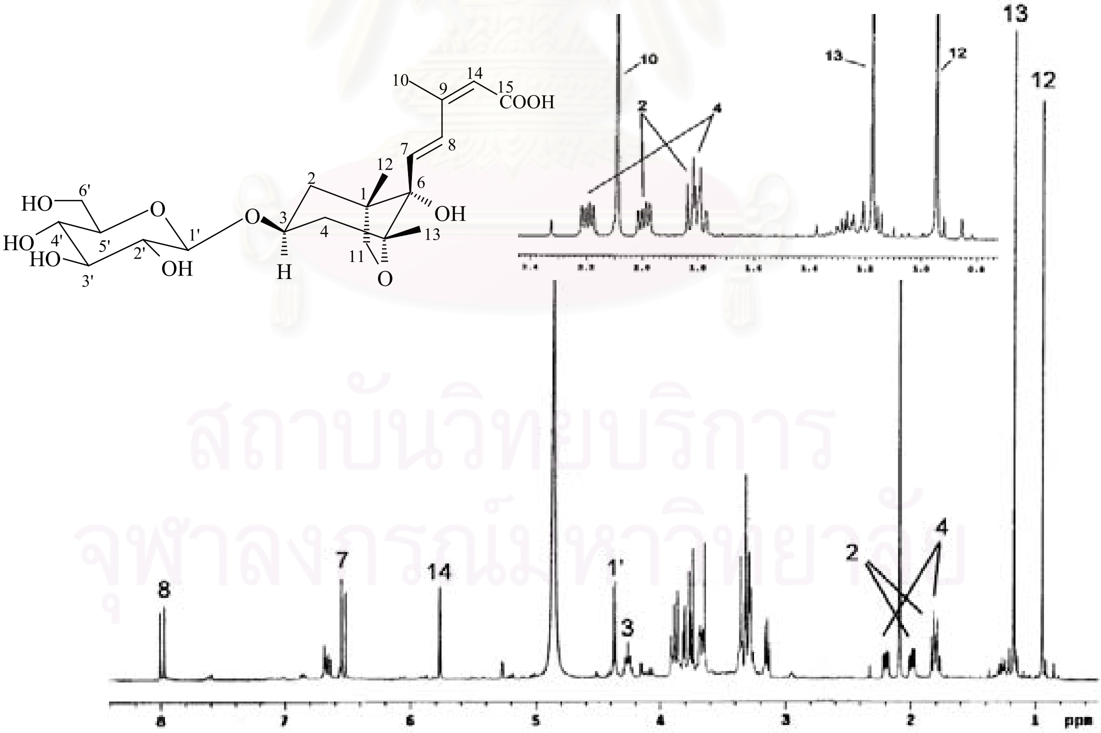


Figure 179 ^{1}H NMR (500 MHz) Spectrum of compound ME18 (methanol-d_4)

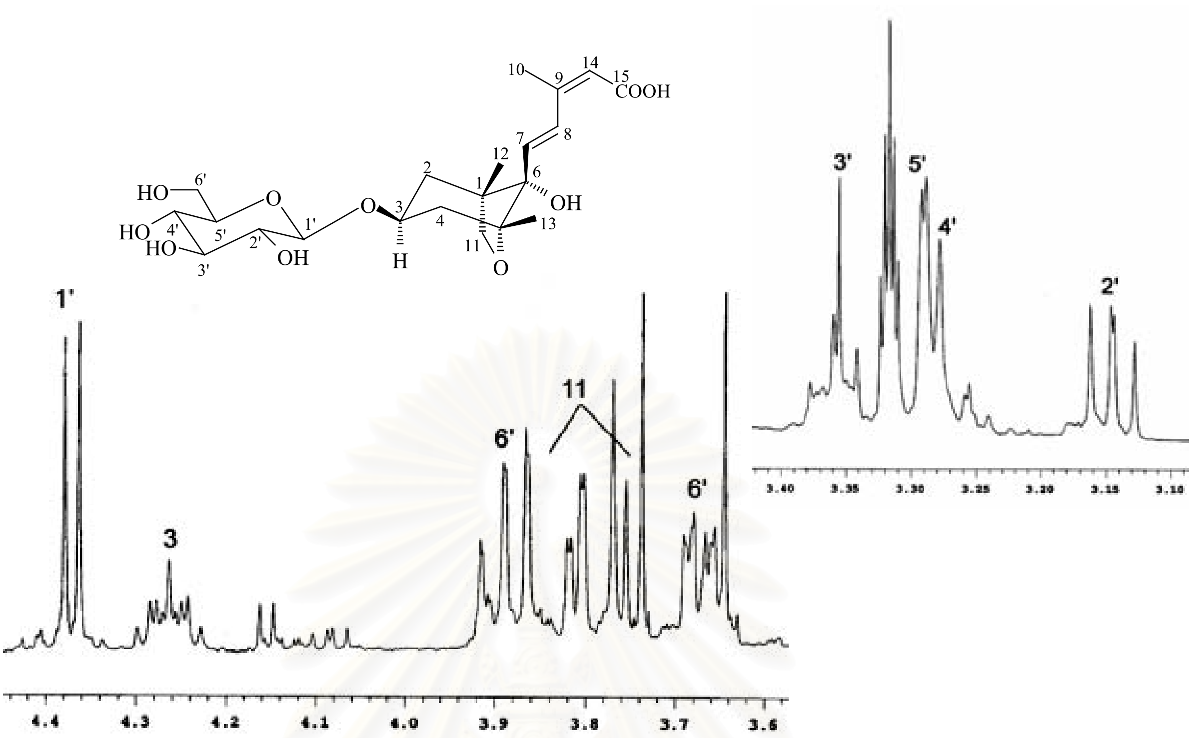


Figure 180 ^{1}H NMR (500 MHz) Spectrum of compound ME18 (methanol-d_4, 3.1-3.4 and 3.6-4.4 ppm)

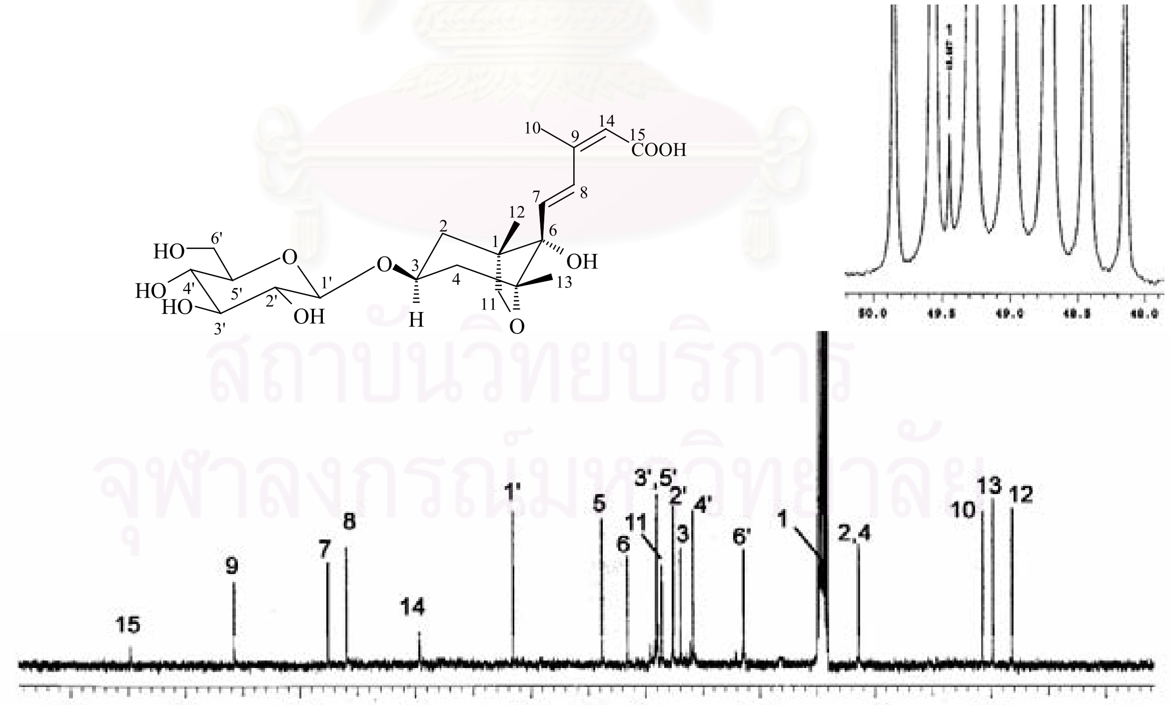


Figure 181 ^{13}C NMR (75 MHz) Spectrum of compound ME18 (methanol-d_4)

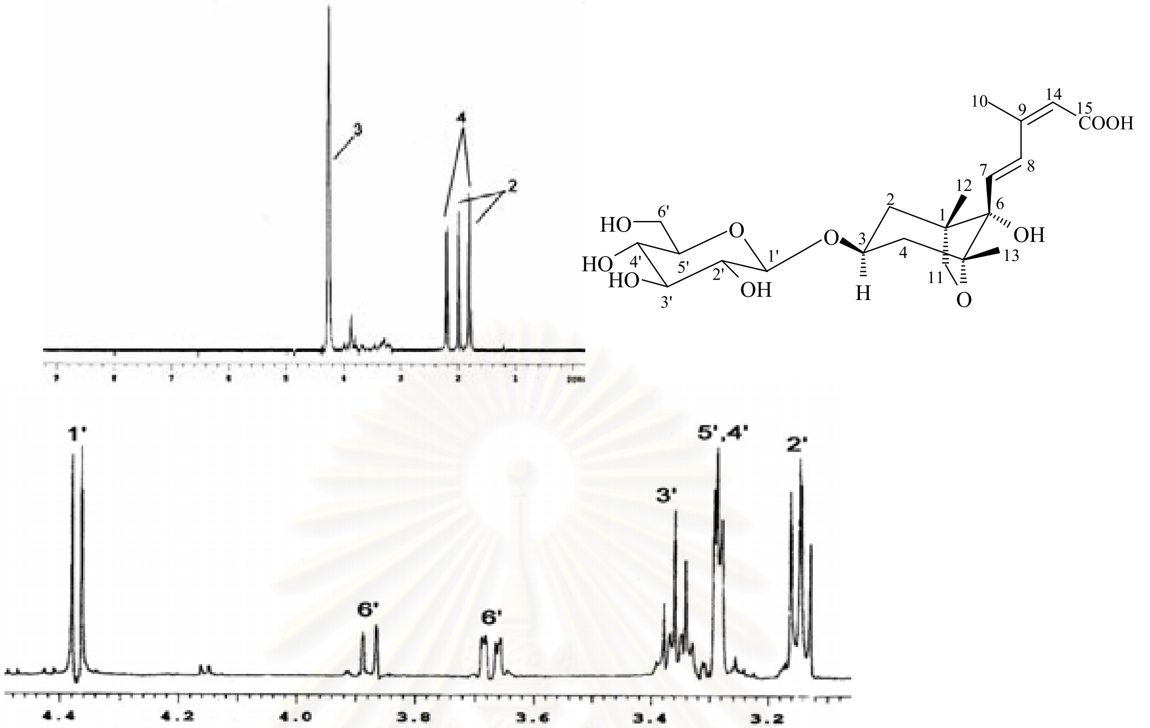


Figure 182 TOCSY Spectrum of compound ME18 (methanol-d_4)

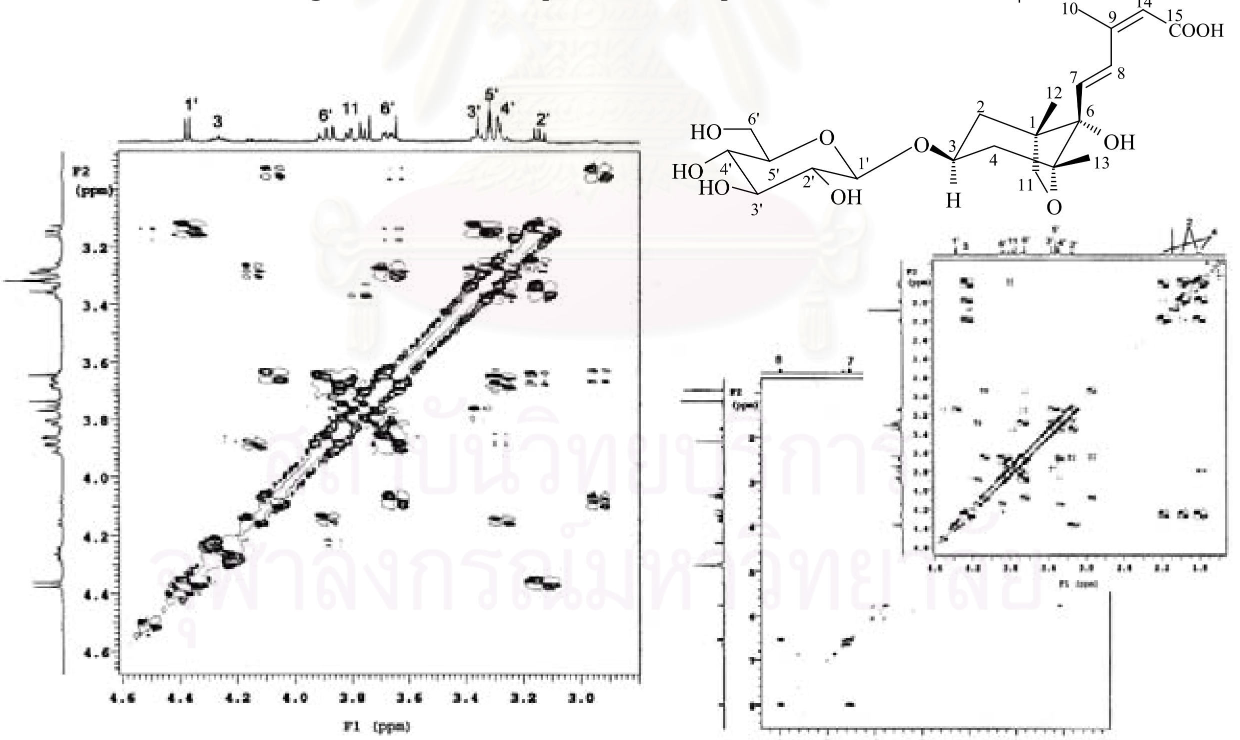


Figure 183 ^{1}H-^{1}H COSY Spectrum of compound ME18 (methanol-d_4)

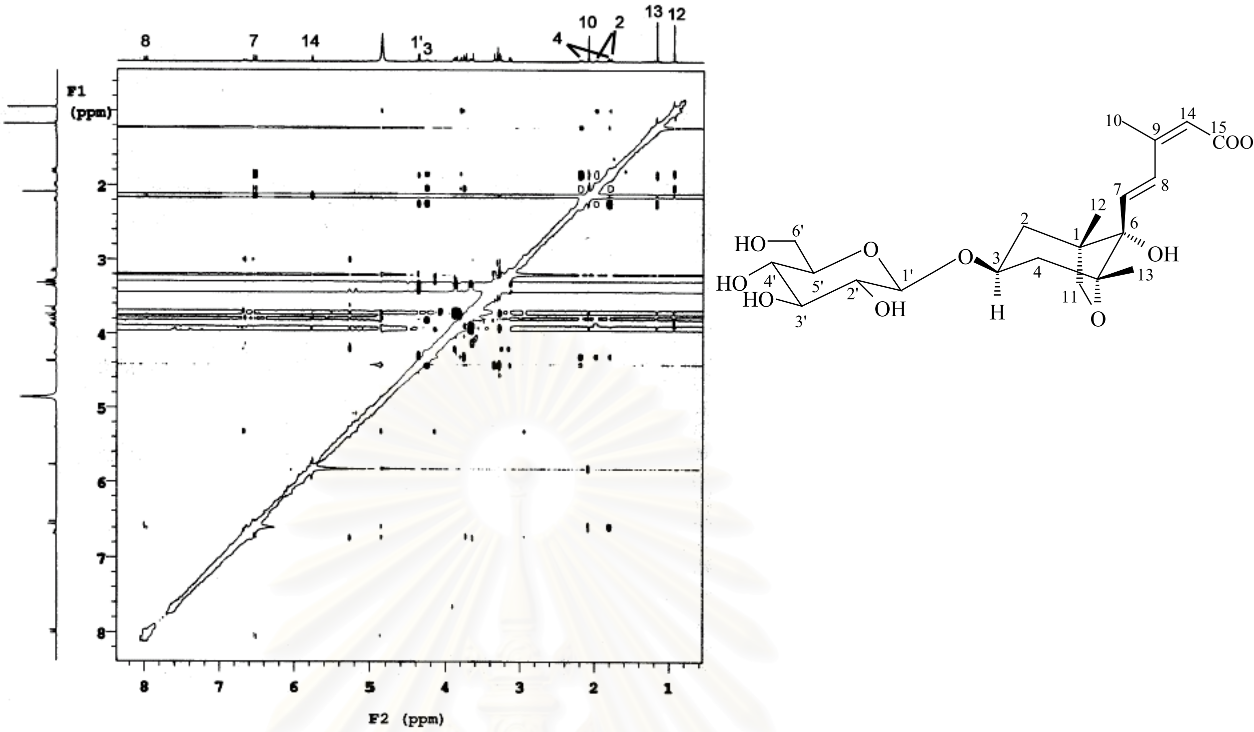


Figure 184 NOESY Spectrum of compound ME18 (methanol-d_4)

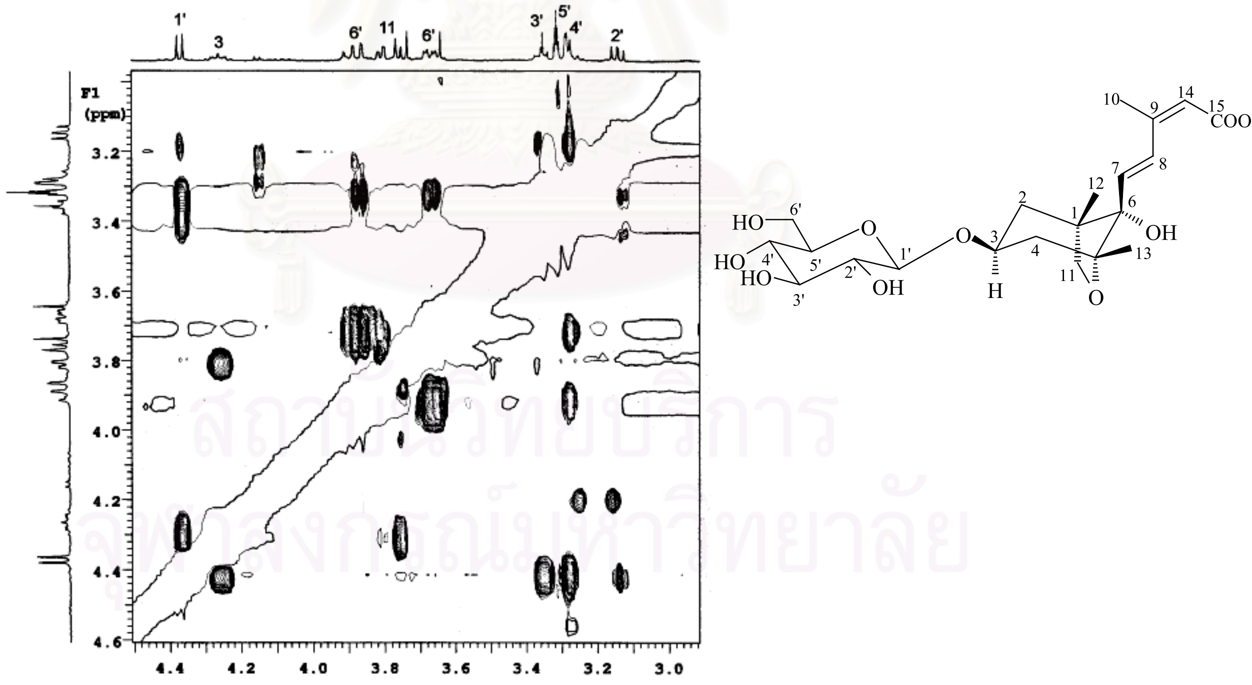


Figure 185 NOESY Spectrum of compound ME18 (methanol-d_4, 2.9-4.5 ppm)

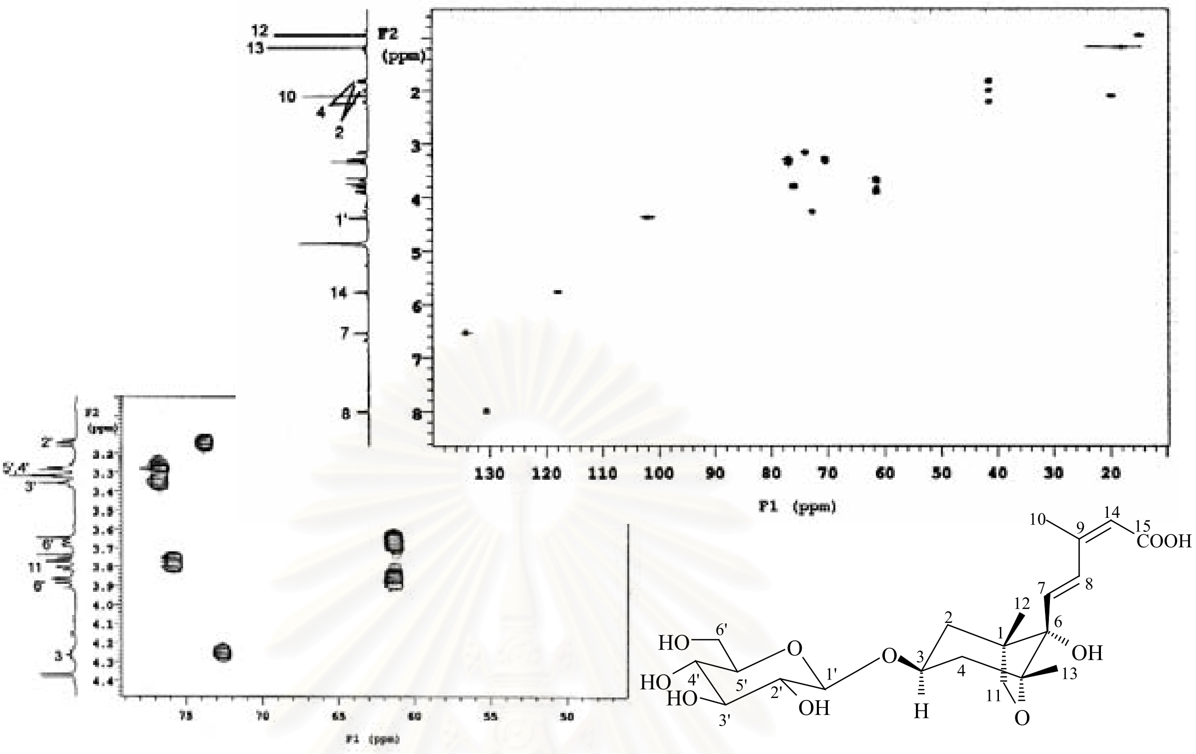


Figure 186 HSQC Spectrum of compound ME18 (methanol-d_4)

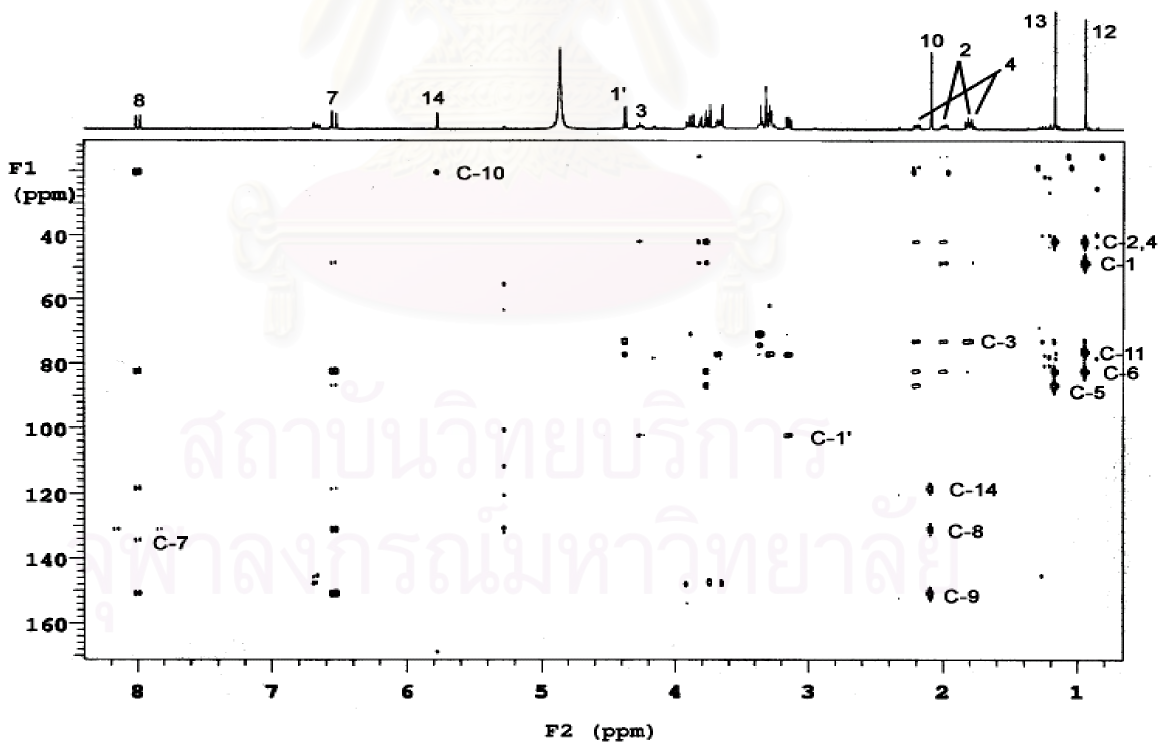


Figure 187 HMBC Spectrum of compound ME18 (methanol-d_4) [δ_H 0.6-8.4 ppm, δ_C 10-170 ppm]

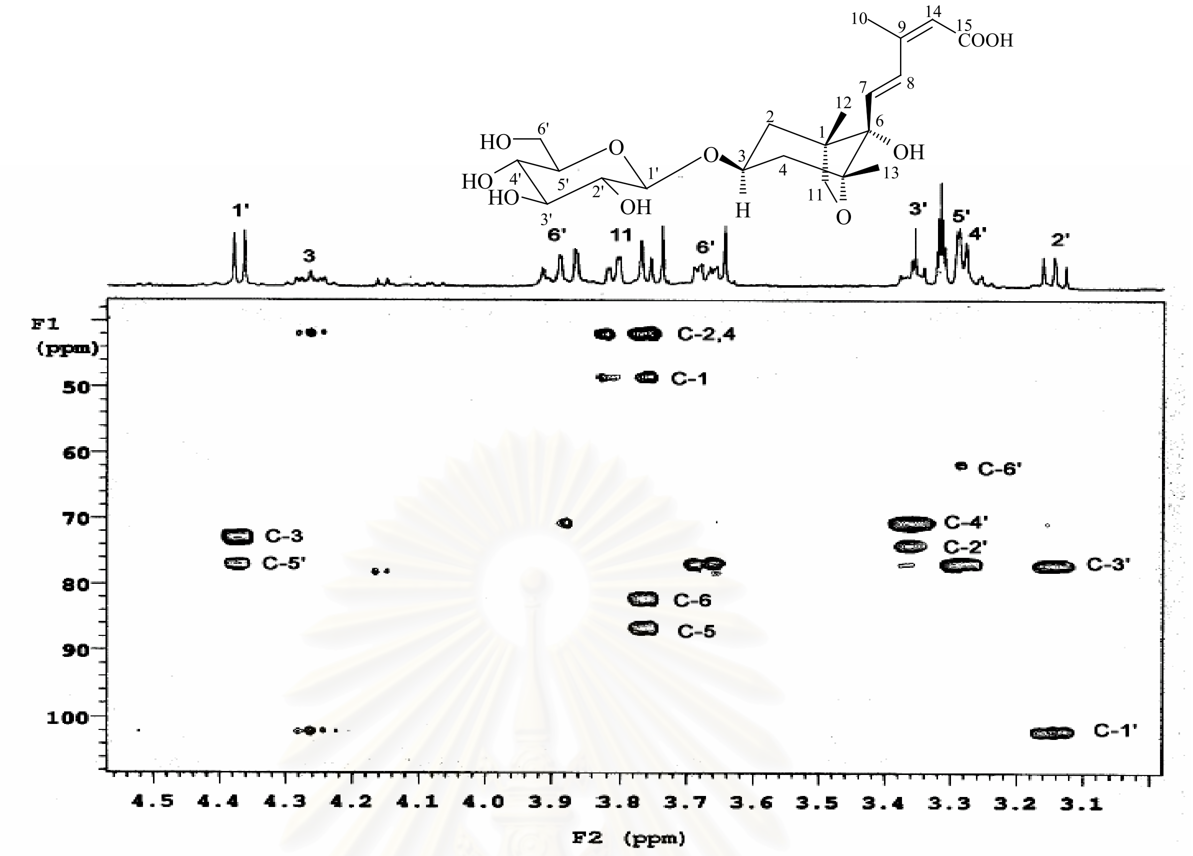


Figure 188 HMBC Spectrum of compound ME18 (methanol-d_4) [δ_H 3.0-4.6 ppm, δ_C 38-110 ppm]

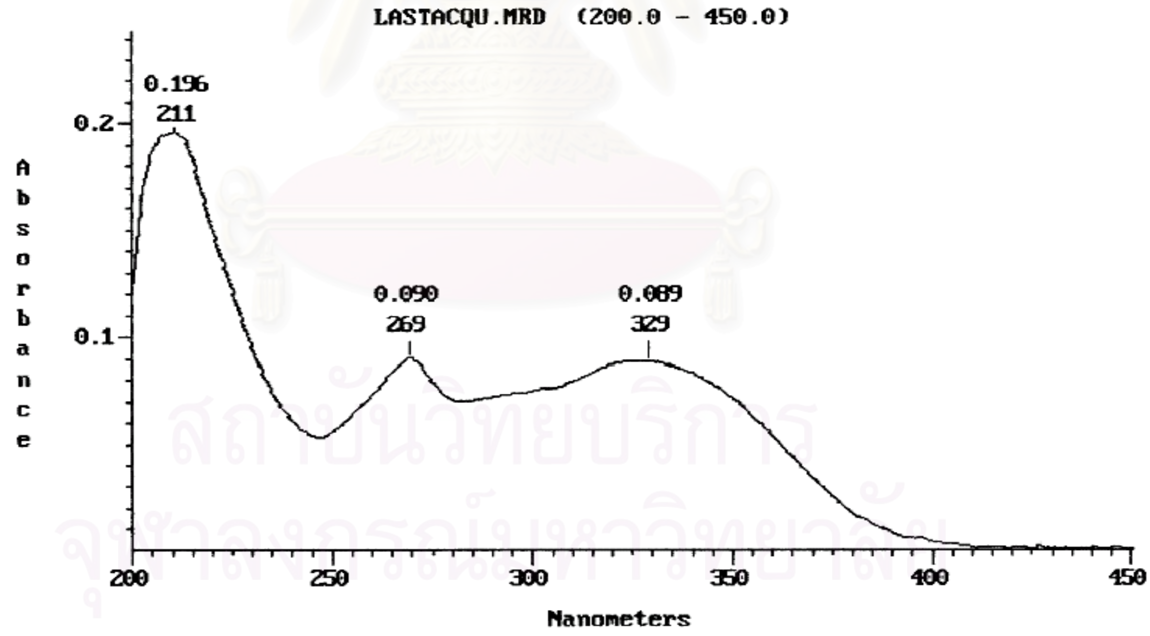


Figure 189 UV Spectrum of compound ME19 (methanol)

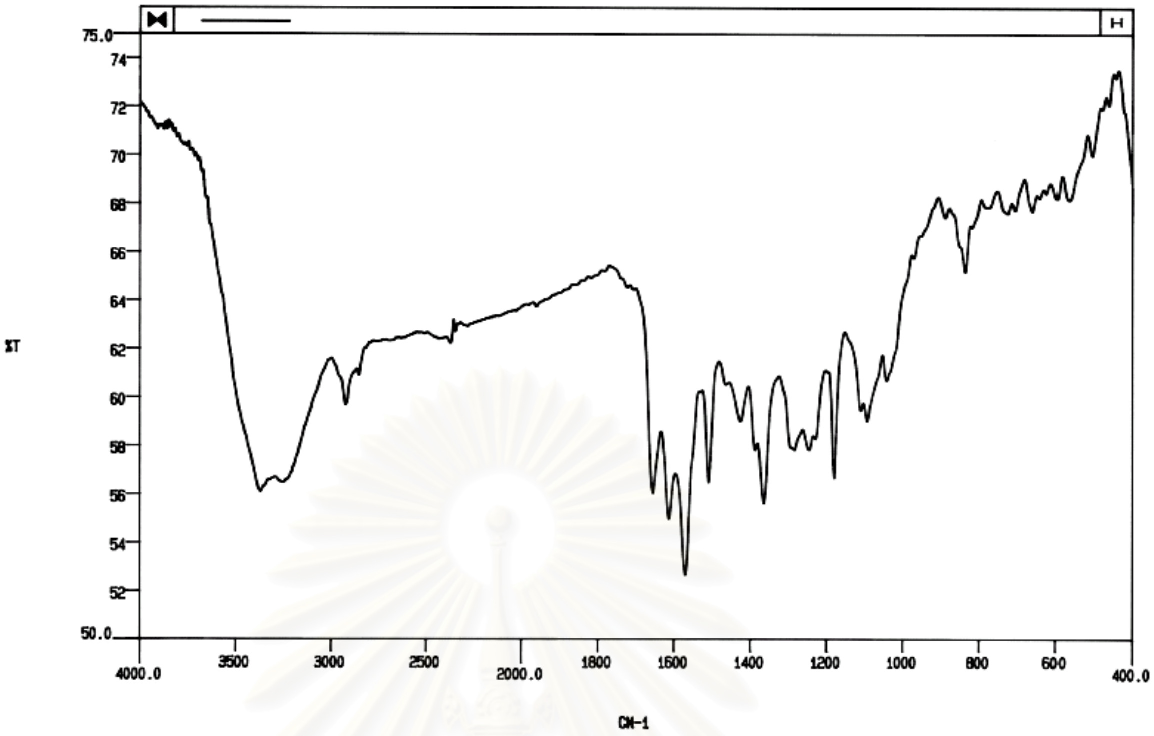


Figure 190 IR Spectrum of compound ME19 (KBr disc)

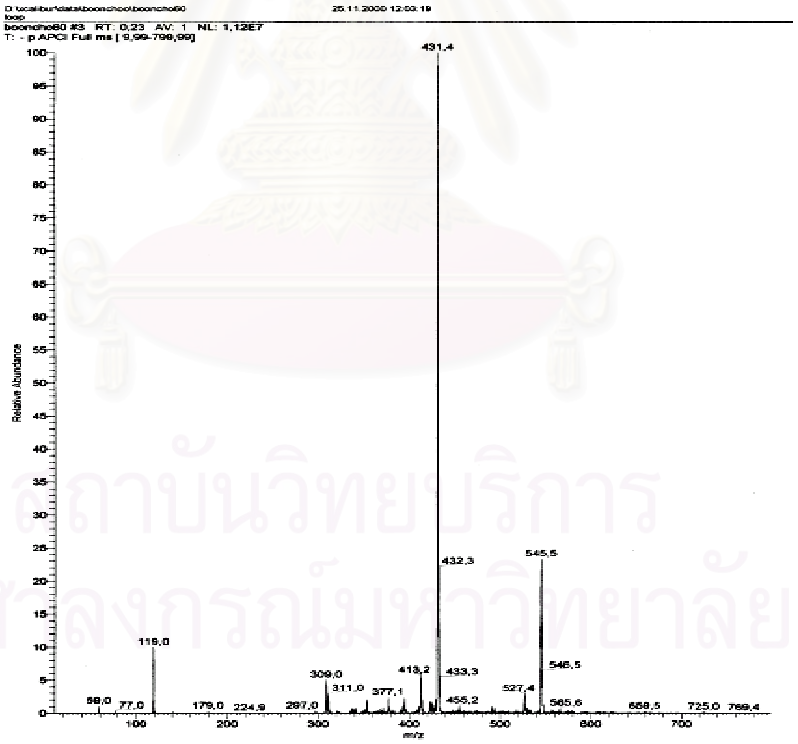


Figure 191 ESI Mass spectrum (negative ion mode) of compound ME19

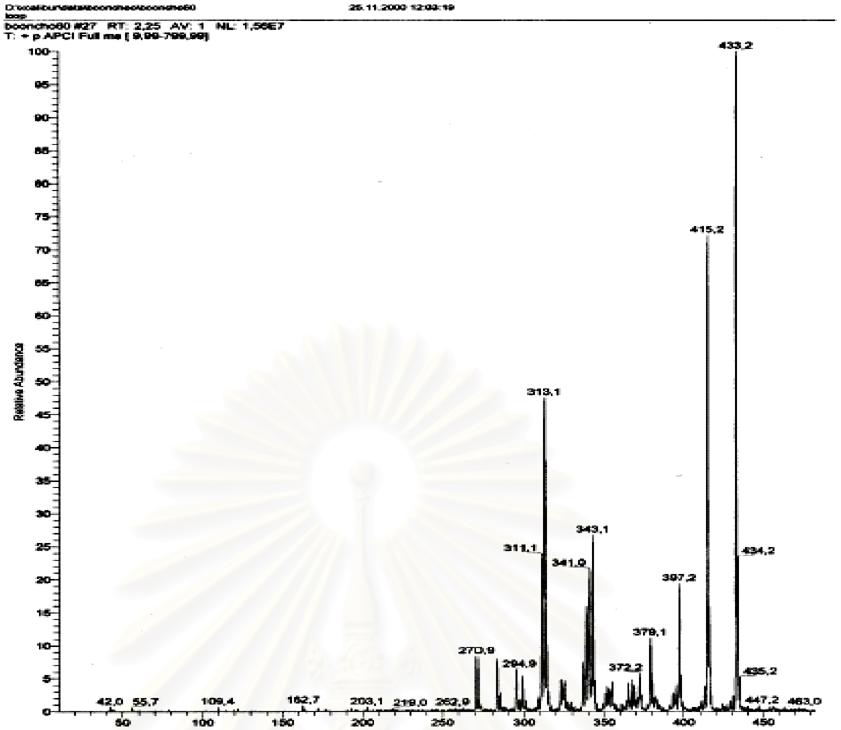


Figure 192 ESI Mass spectrum (positive ion mode) of compound ME19

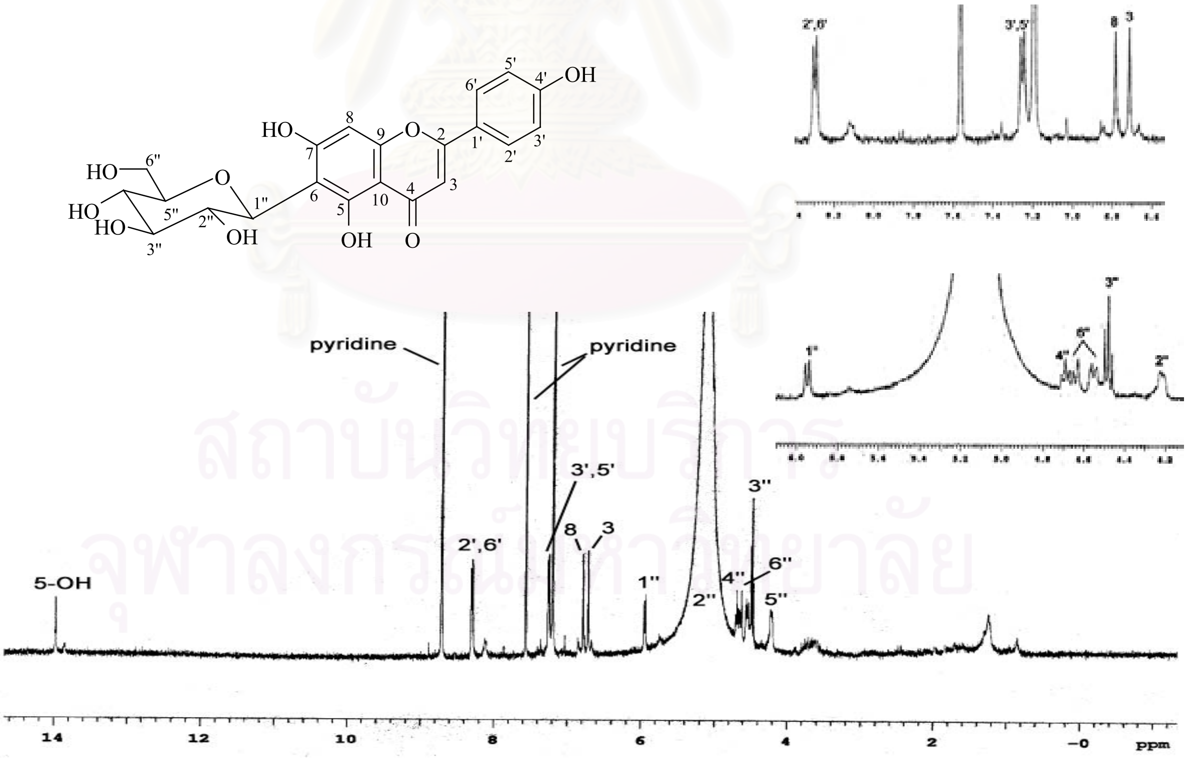


Figure 193 ^{1}H NMR (500 MHz) Spectrum of compound ME19 (pyridine-d_5)

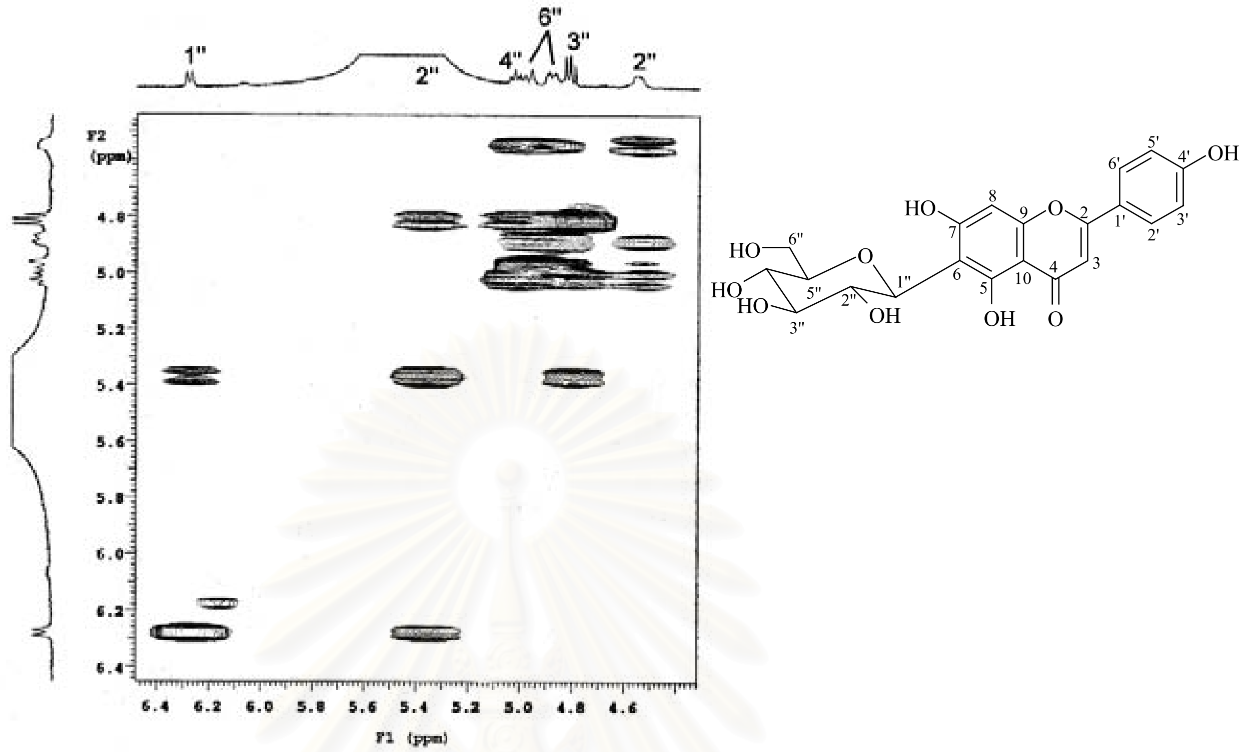


Figure 194 ^{1}H-^{1}H COSY Spectrum of compound ME19 (pyridine-d_5)

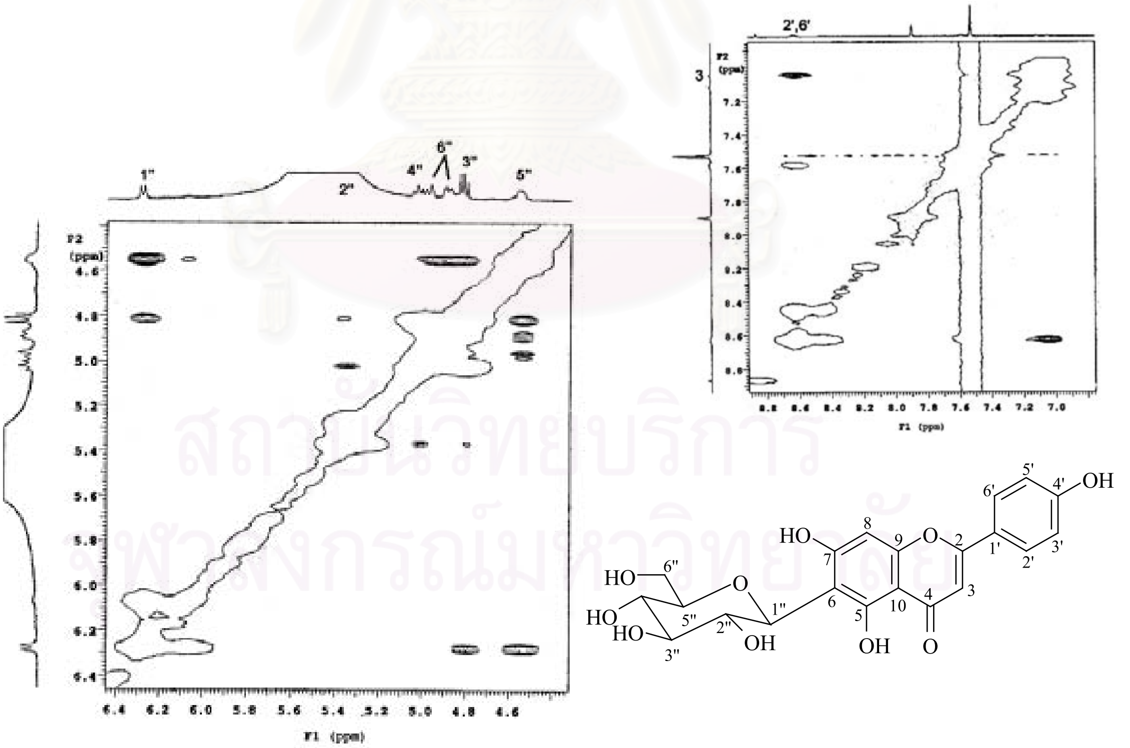


Figure 195 ROESY Spectrum of compound ME19 (pyridine-d_5)

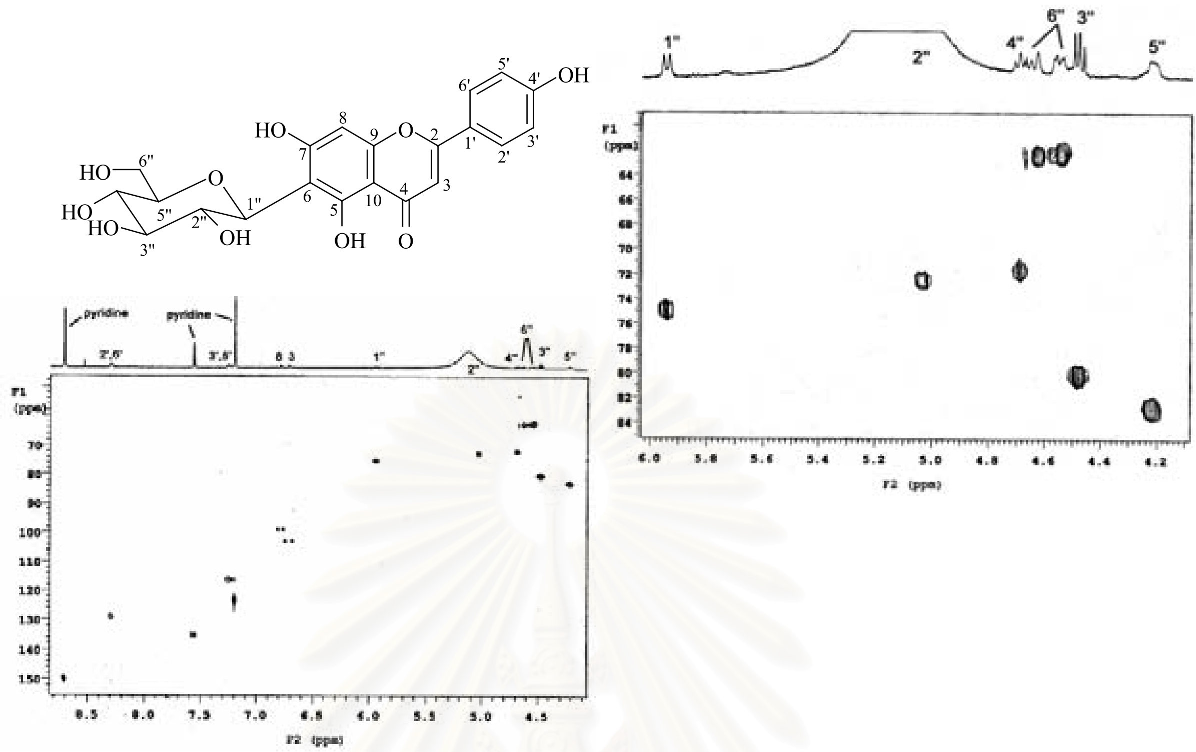


Figure 196 HSQC Spectrum of compound ME19 (pyridine-d_5)

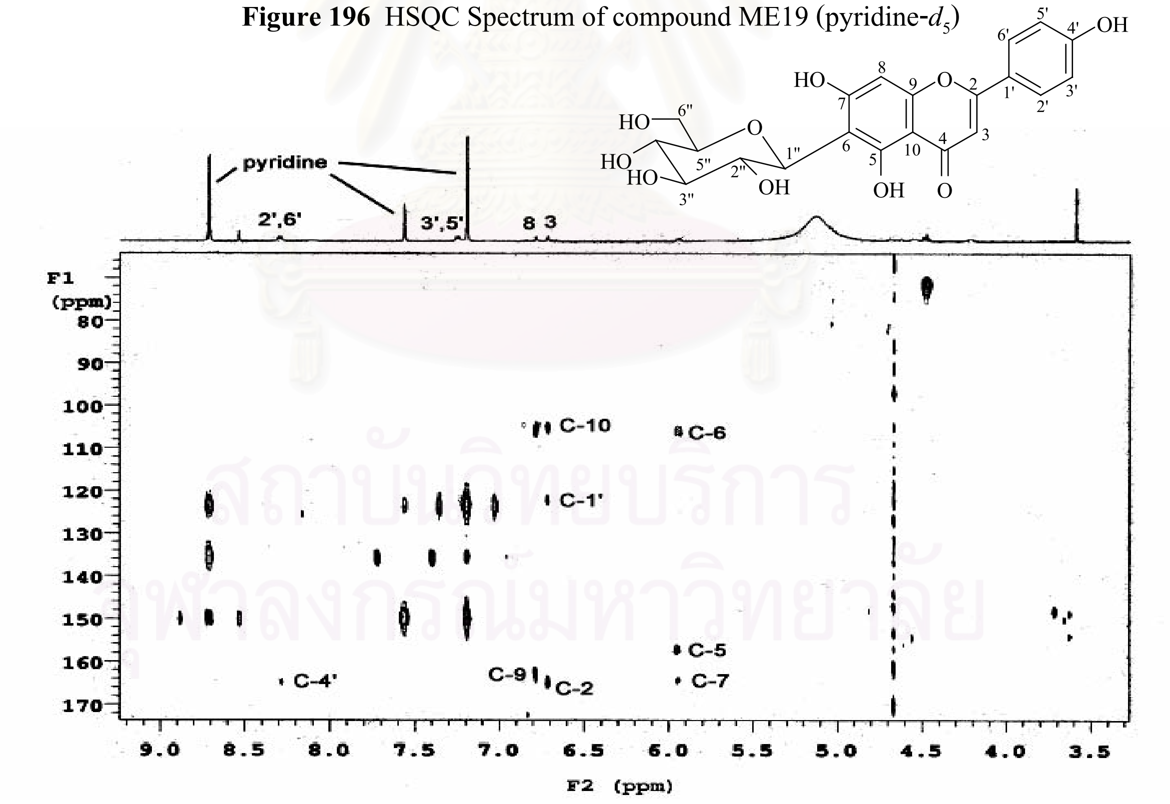


Figure 197 HMBC Spectrum of compound ME19 (pyridine-d_5)

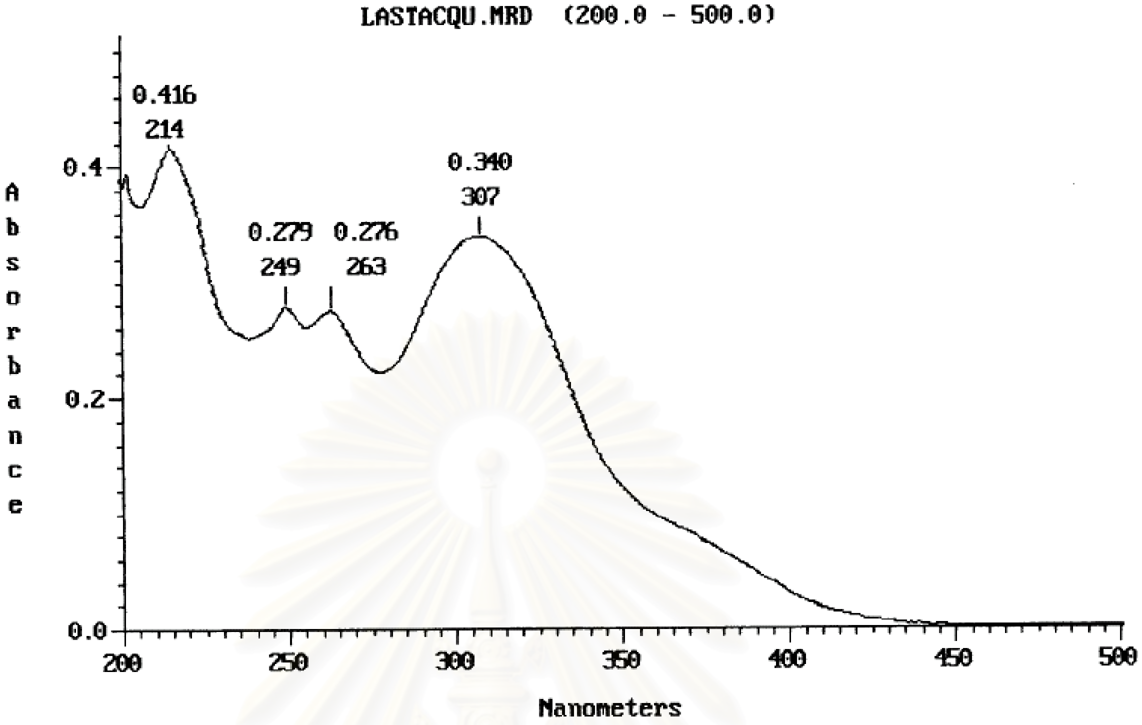


Figure 198 UV Spectrum of compound ME20 (methanol)

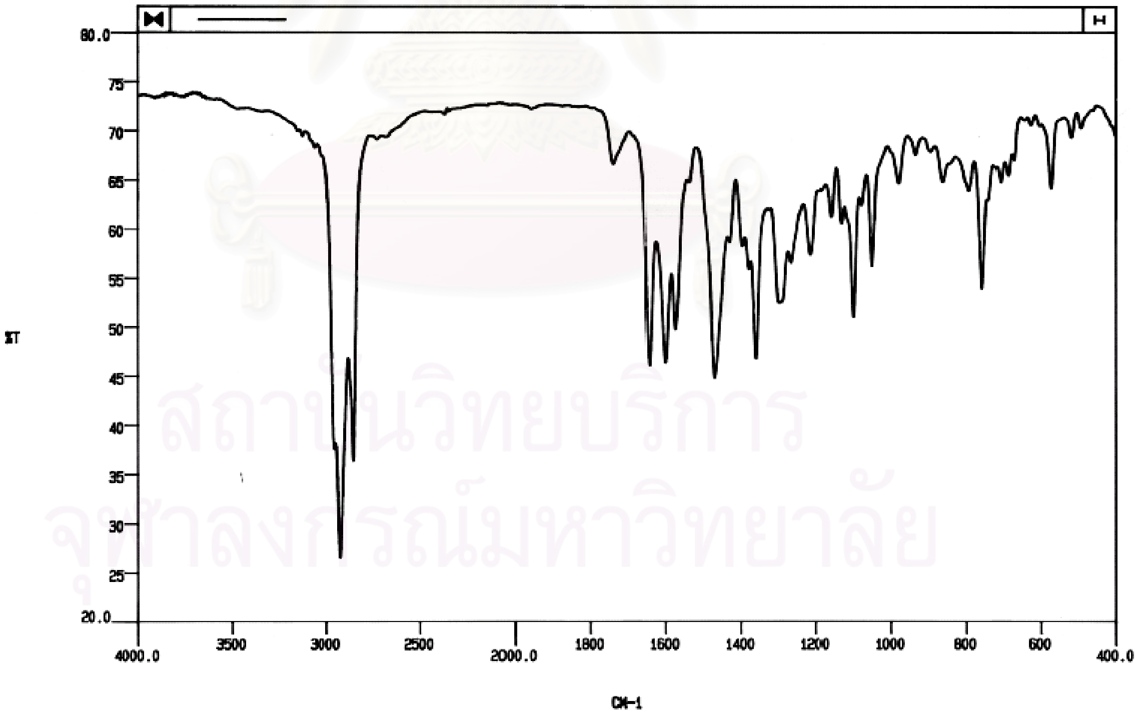


Figure 199 IR Spectrum of compound ME20 (KBr disc)

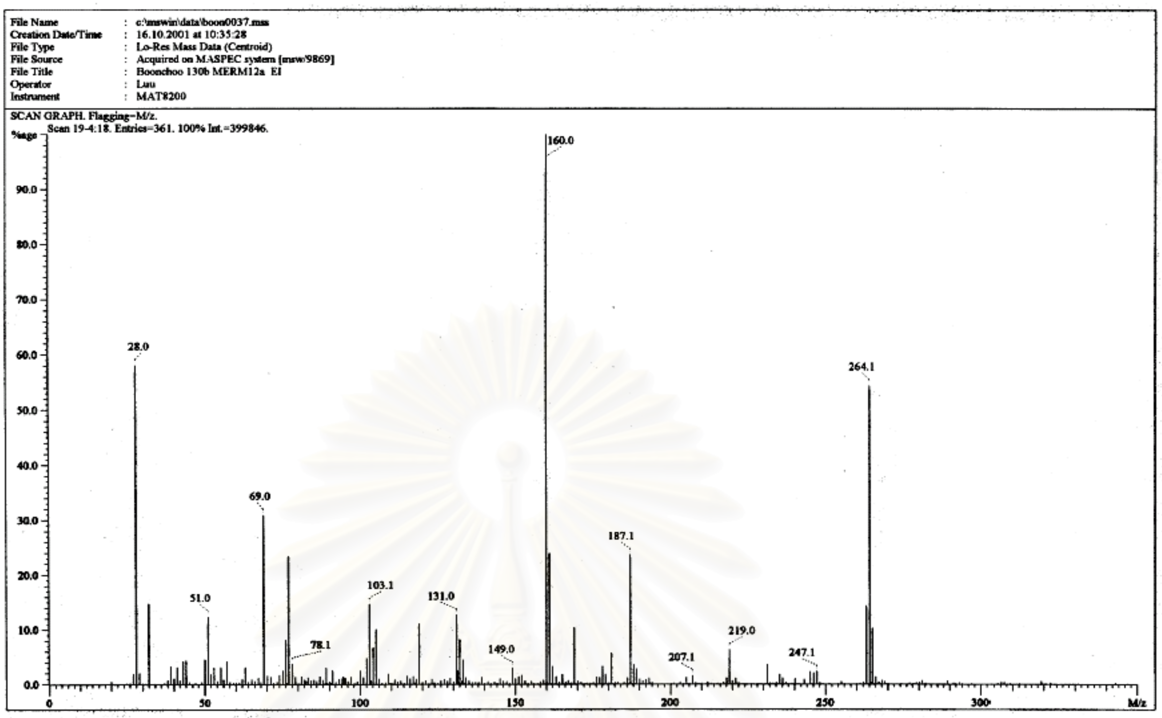


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Figure 200 EI Mass spectrum of compound ME20

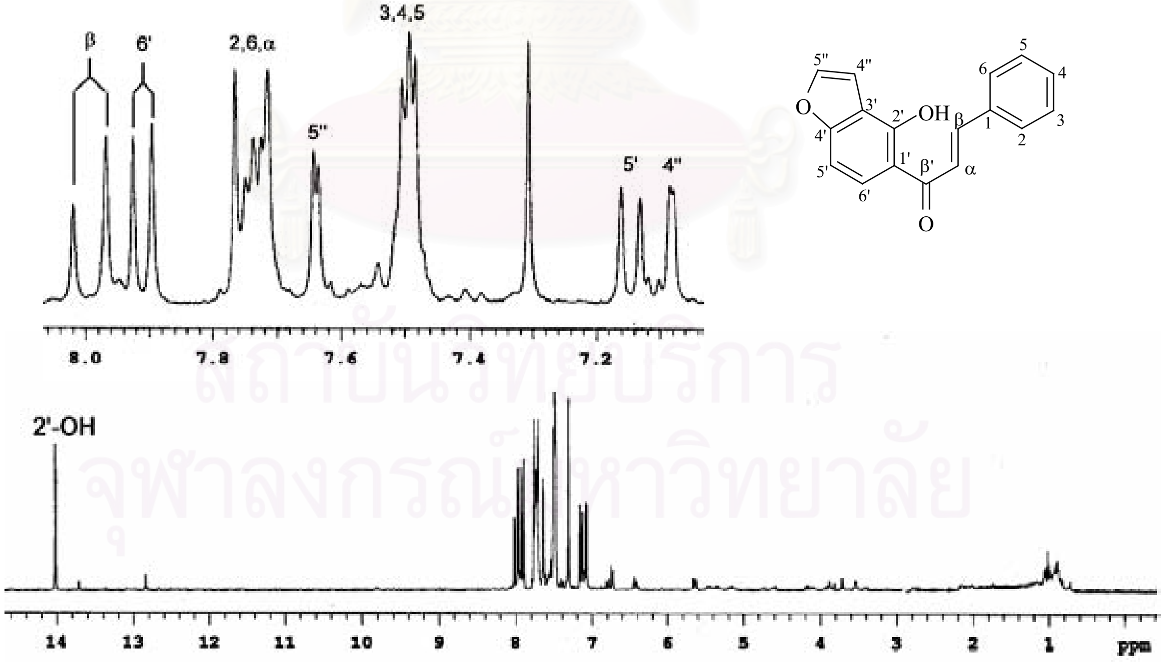

Figure 201 ^{1}H NMR (300 MHz) Spectrum of compound ME20 (CDCl$_3$)

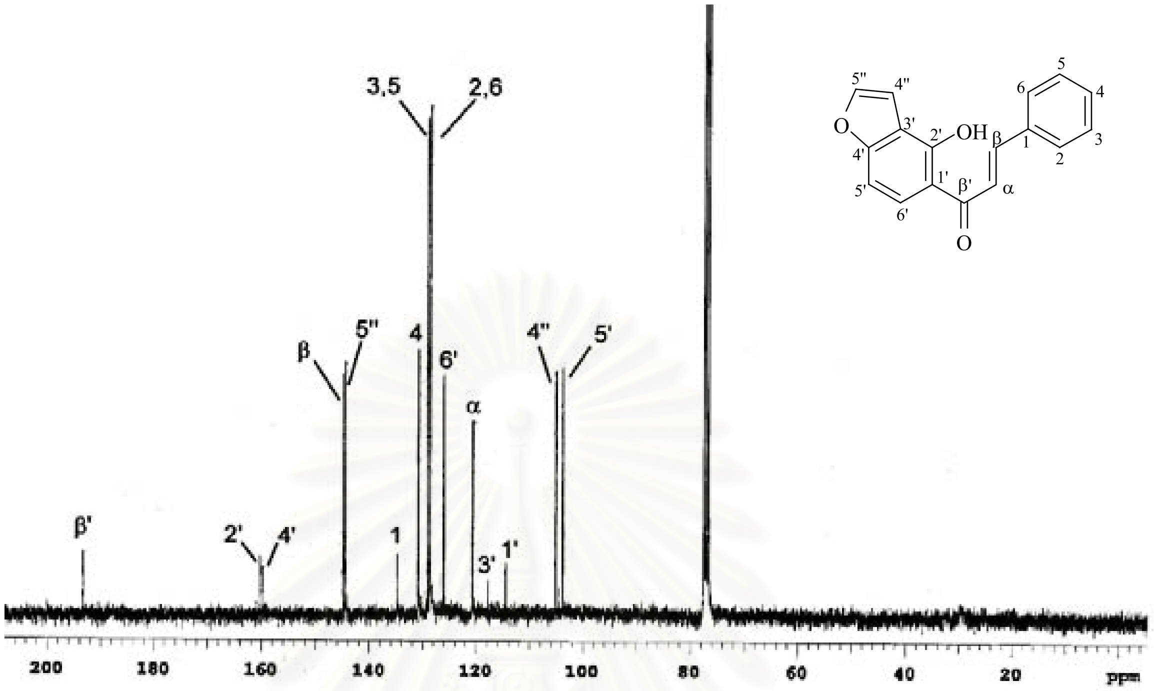

Figure 202 ^{13}C NMR (75 MHz) Spectrum of compound ME20 (CDCl$_3$)

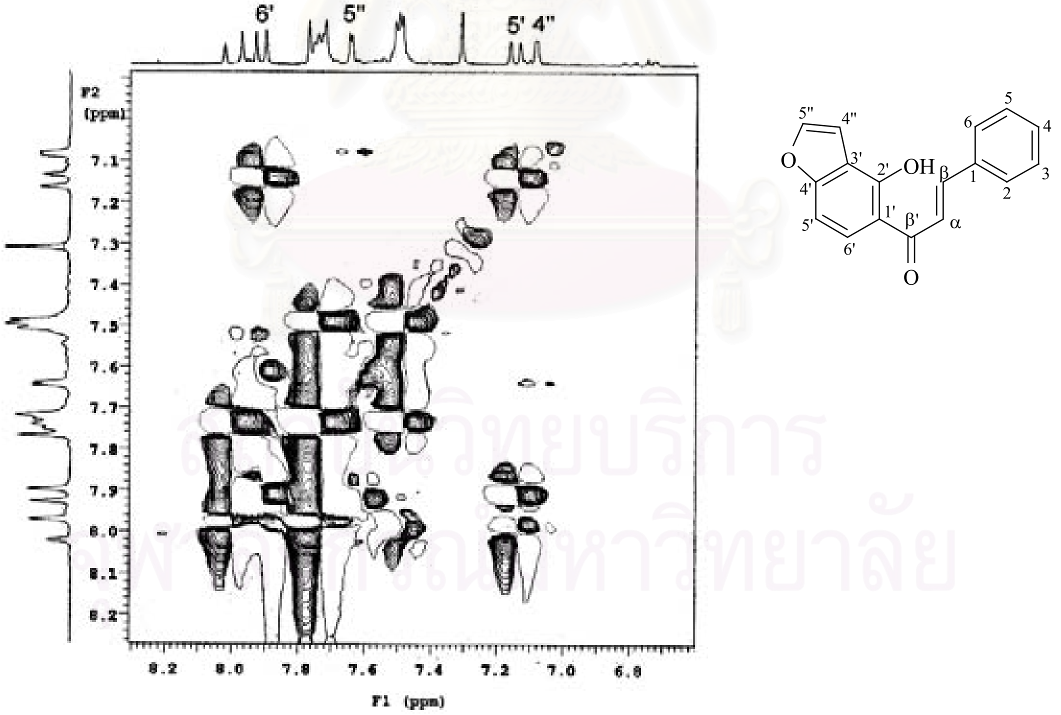

Figure 203 ^{1}H-^{1}H COSY Spectrum of compound ME20 (CDCl$_3$)

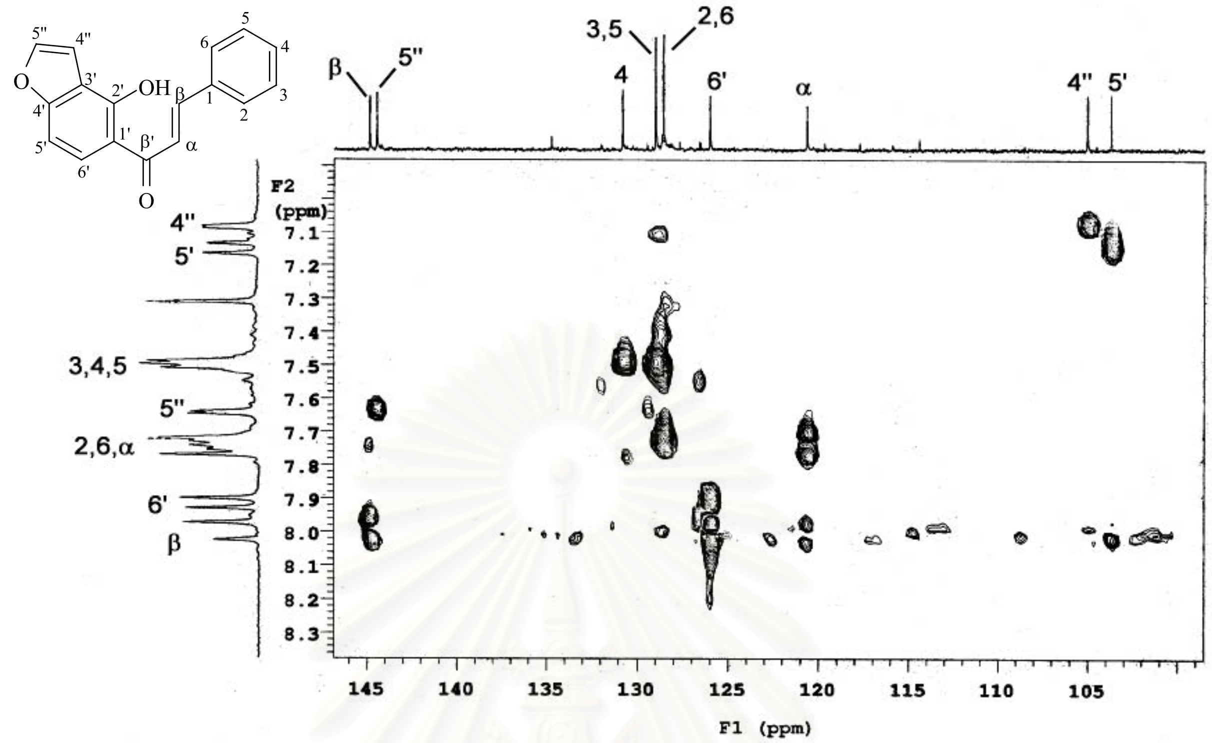

Figure 204 HSQC Spectrum of compound ME20 ($CDCl_3$)

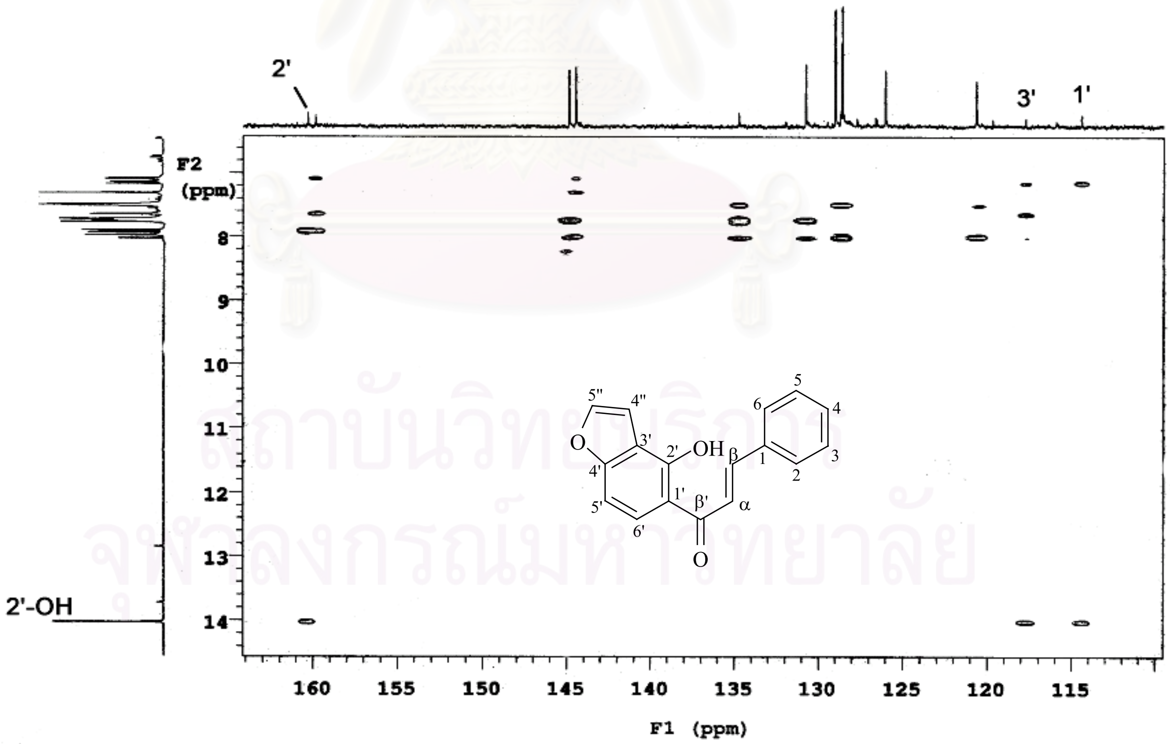

Figure 205 HMBC Spectrum of compound ME20 ($CDCl_3$) [δ_H 6.4-14.6 ppm, δ_C 110-165 ppm]

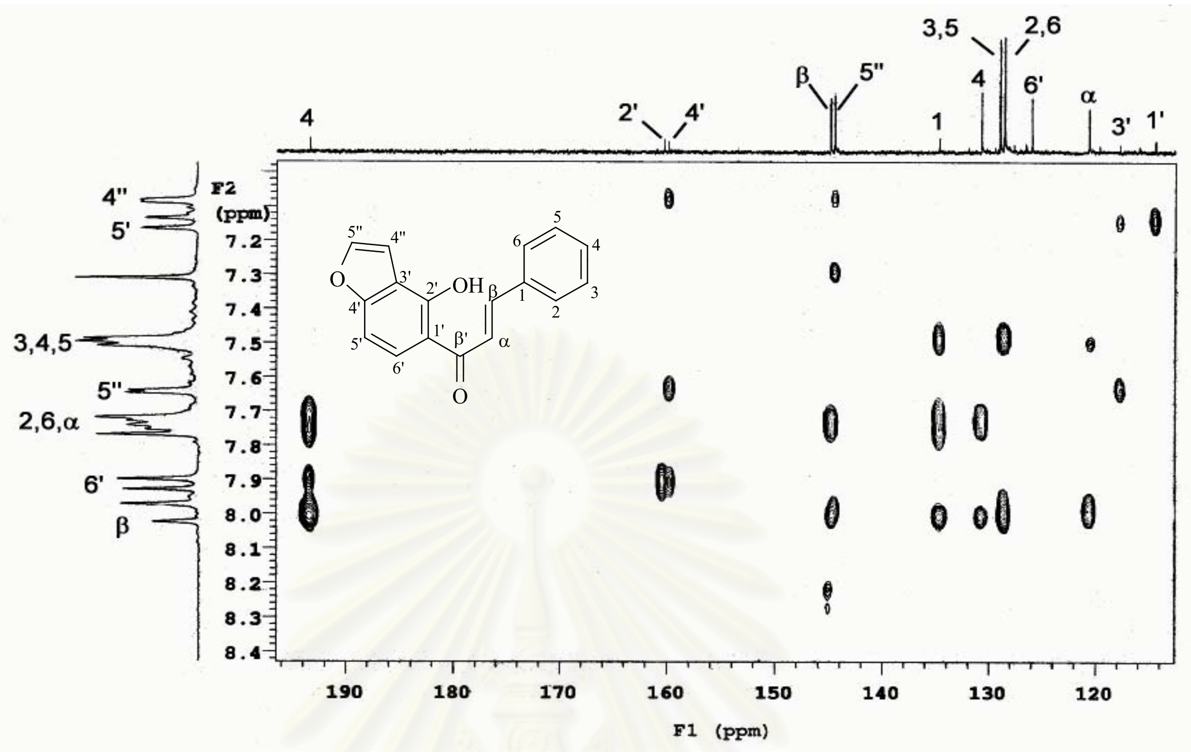

Figure 206 HMBC Spectrum of compound ME20 ($CDCl_3$) [δ_H 6.9-8.4 ppm, δ_C 112-196 ppm]

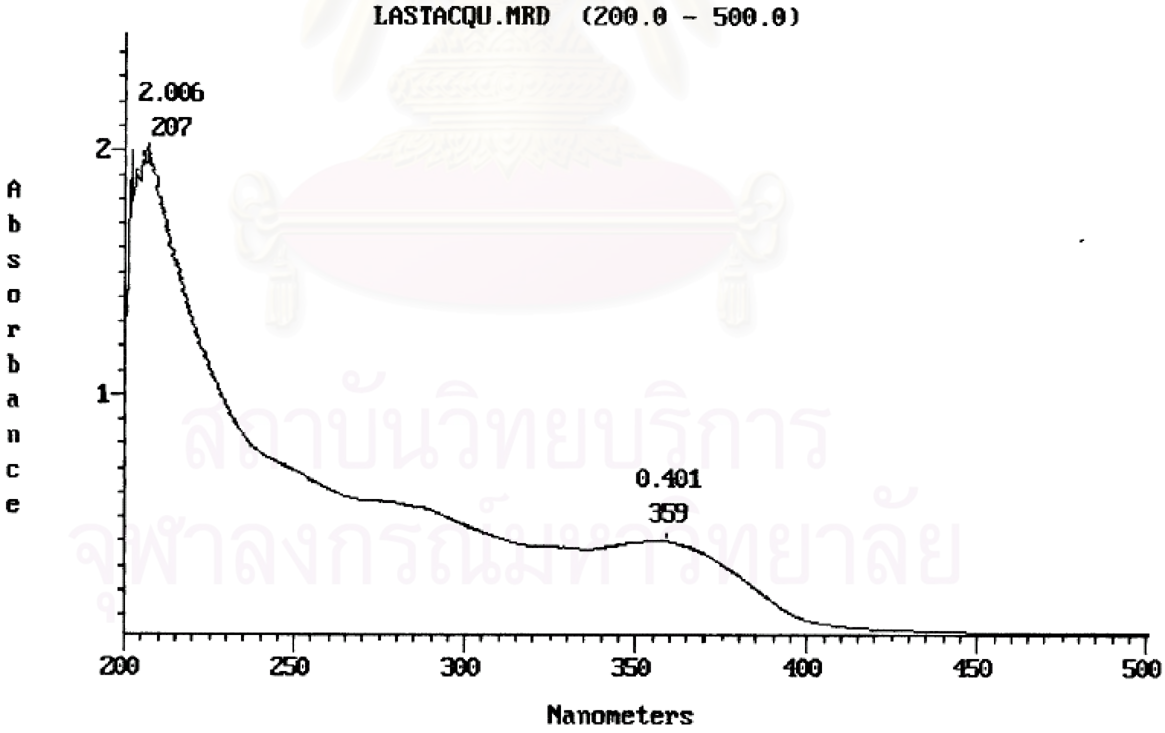

Figure 207 UV Spectrum of compound ME22 (methanol)

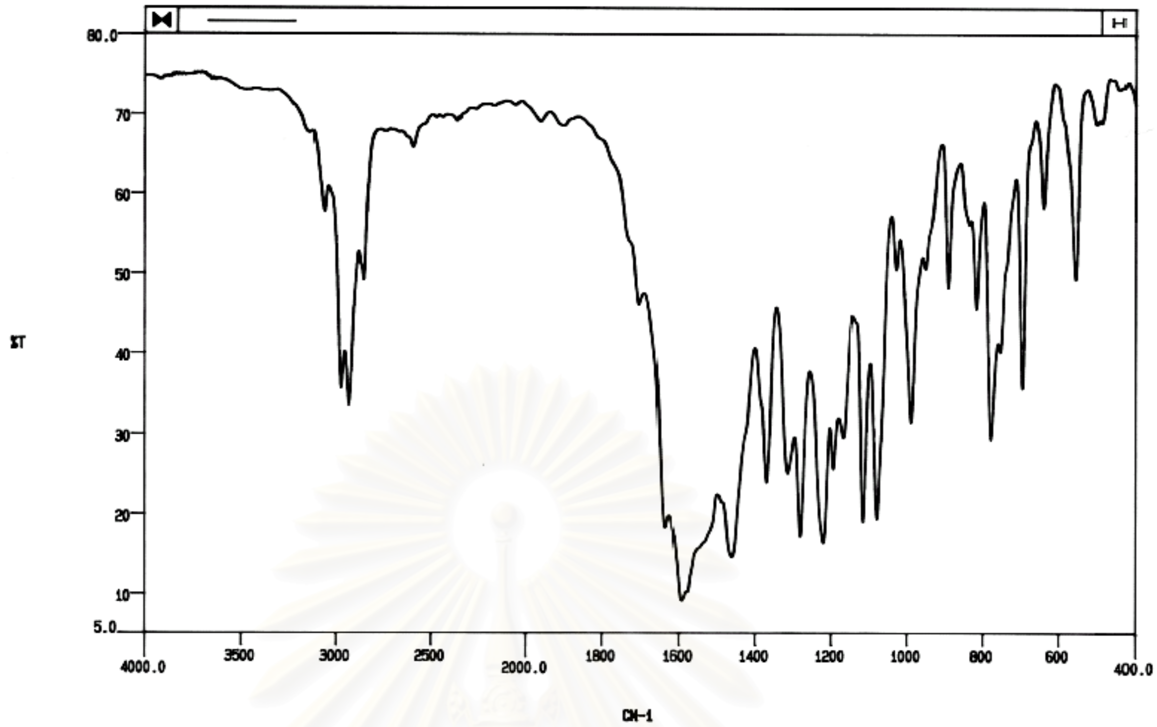

Figure 208 IR Spectrum of compound ME21 (KBr disc)

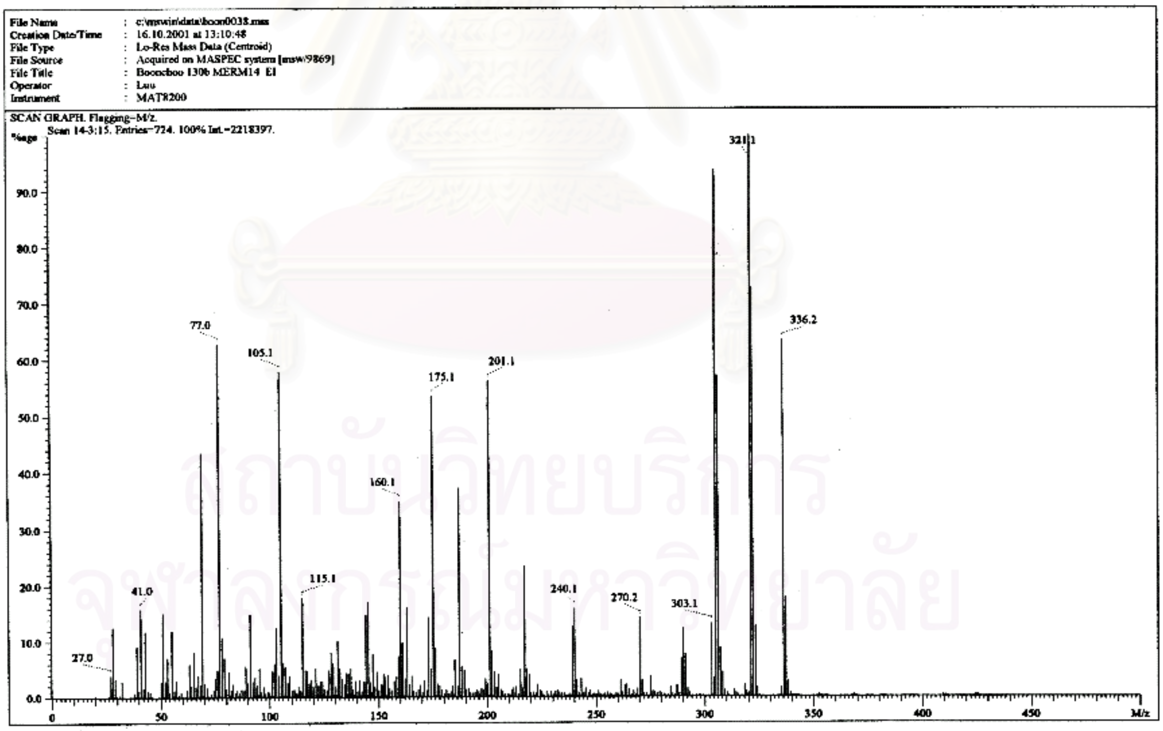
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Figure 209 EI Mass spectrum of compound ME21

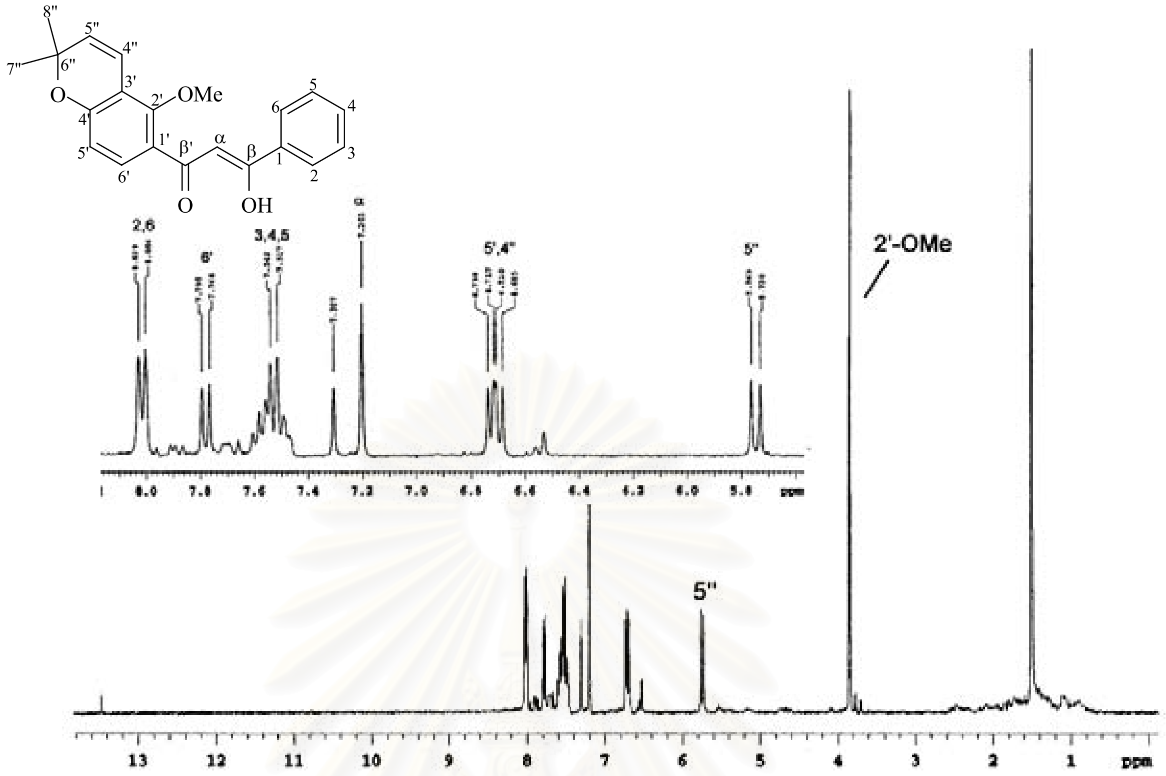


Figure 210 ^{1}H NMR (300 MHz) Spectrum of compound ME21 (CDCl$_3$)

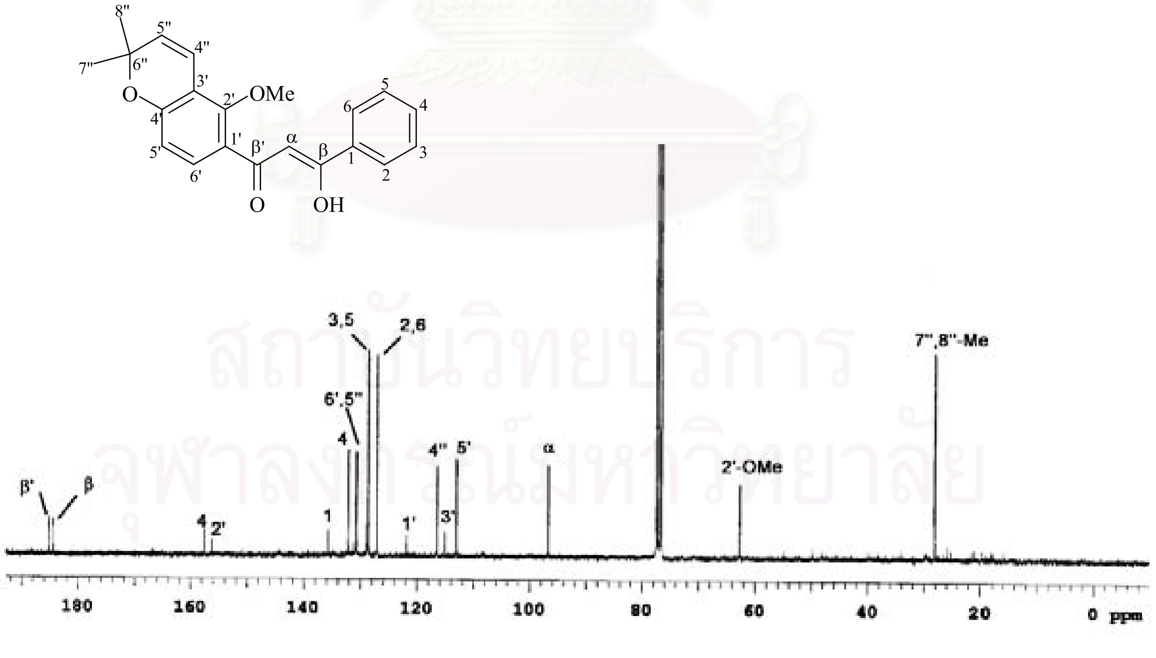


Figure 211 ^{13}C NMR (75 MHz) Spectrum of compound ME21 (CDCl$_3$)

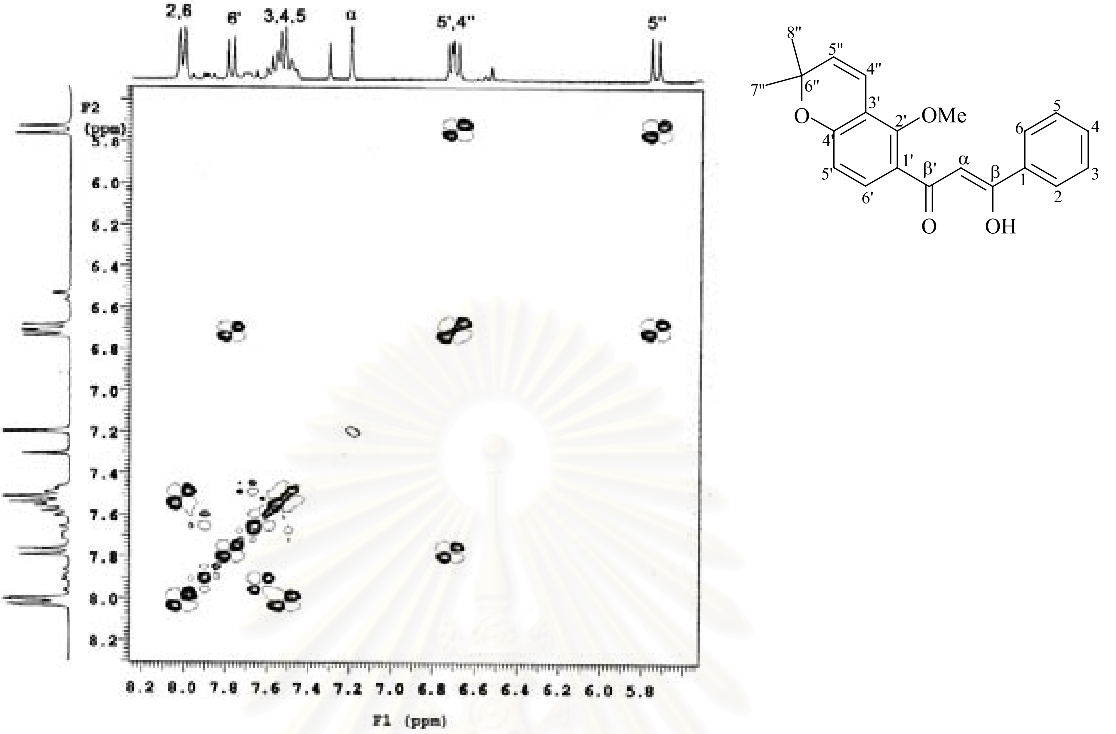


Figure 212 ^{1}H-^{1}H COSY Spectrum of compound ME21 ($CDCl_3$)

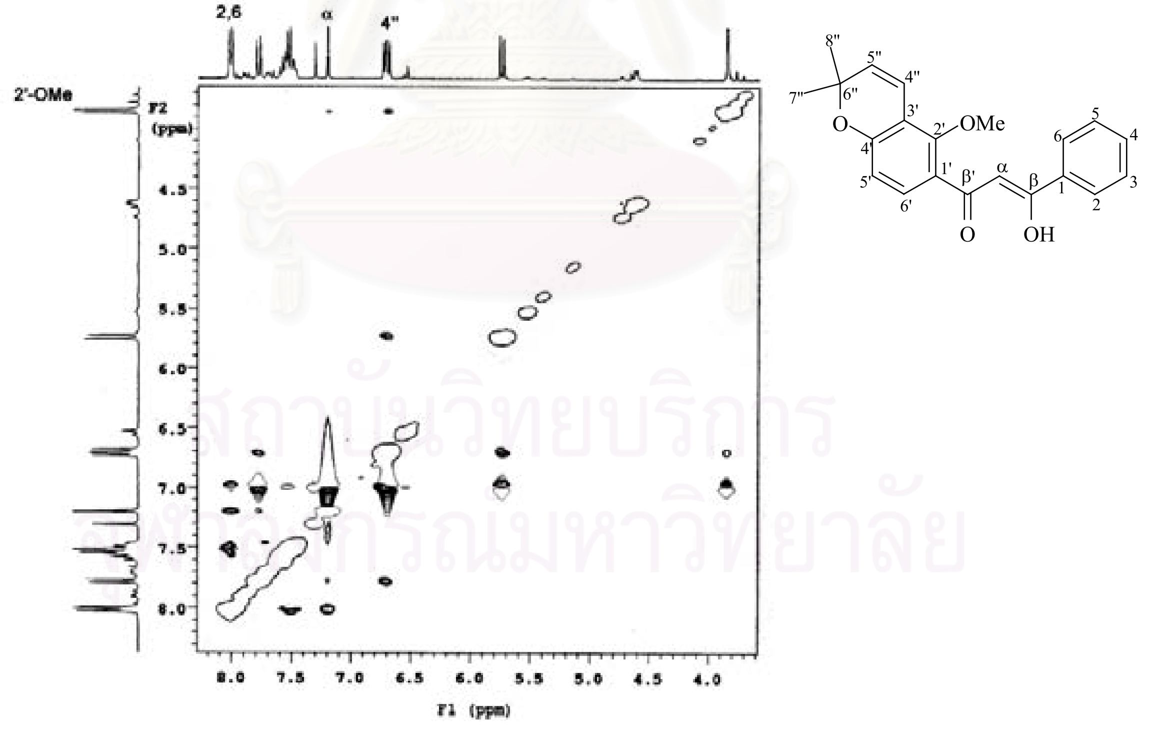


Figure 213 NOESY Spectrum of compound ME21 ($CDCl_3$)

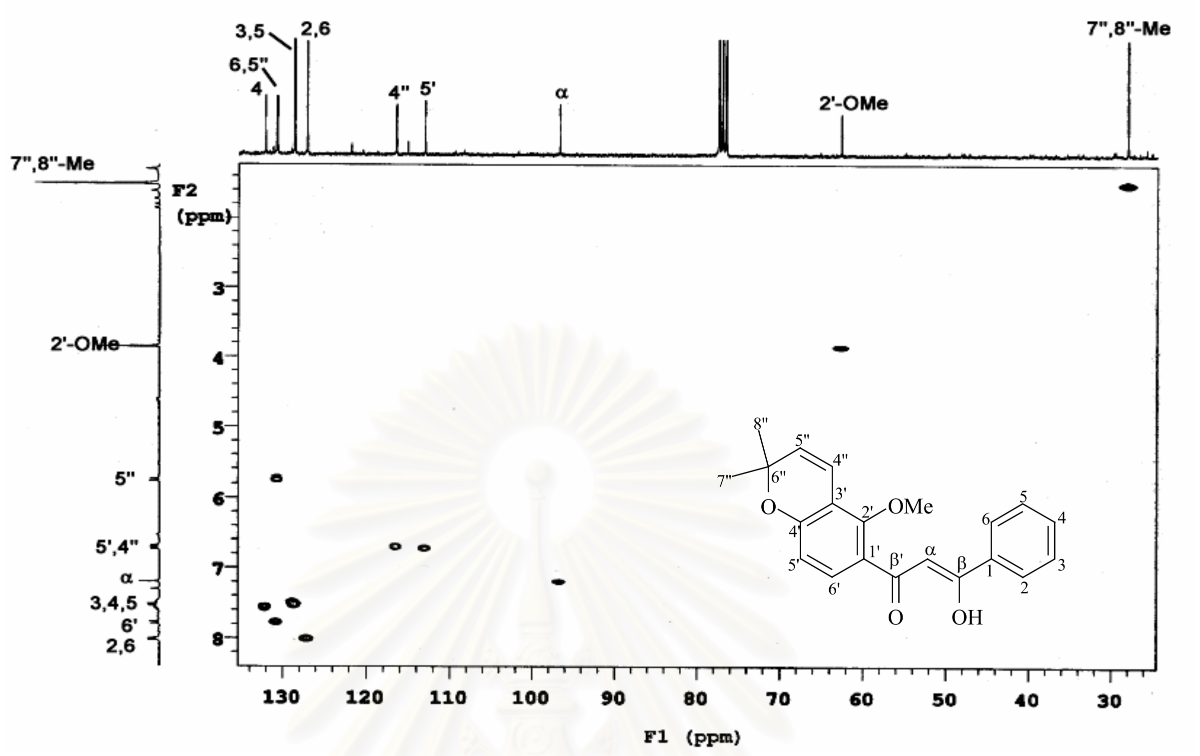


Figure 214 HSQC Spectrum of compound ME21 ($CDCl_3$)

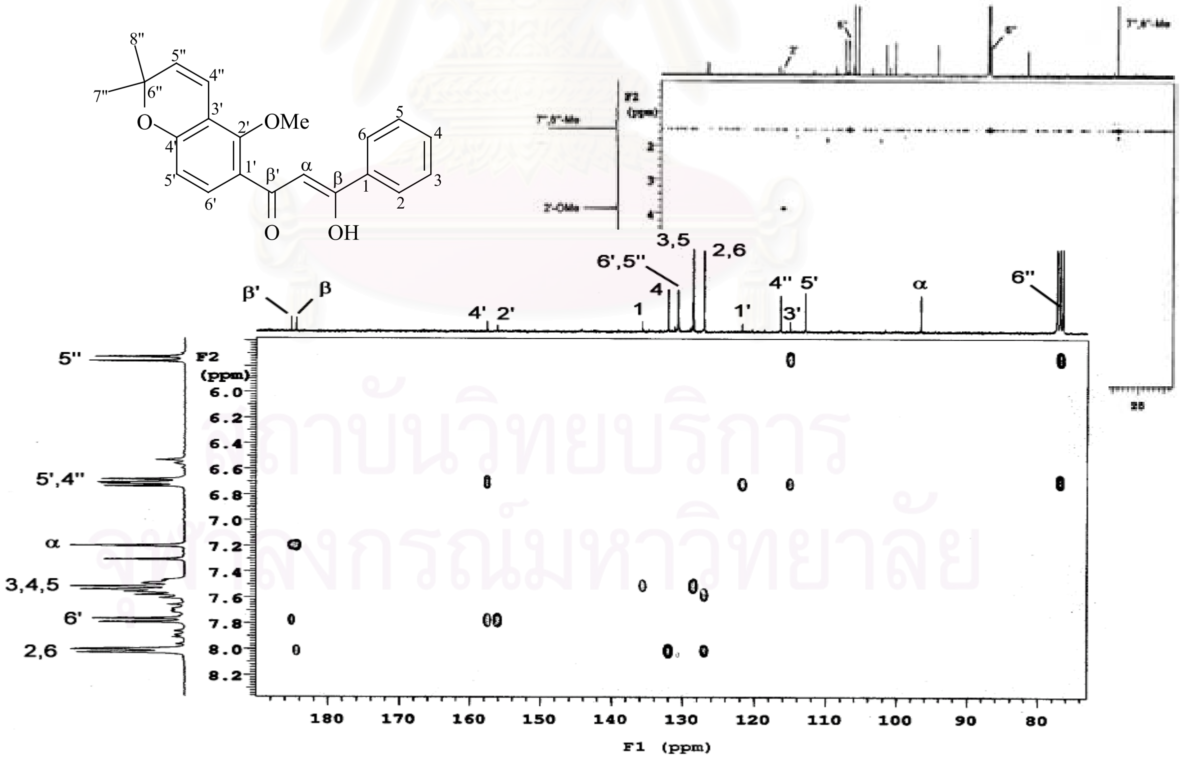


Figure 215 HMBC Spectrum of compound ME21 ($CDCl_3$)

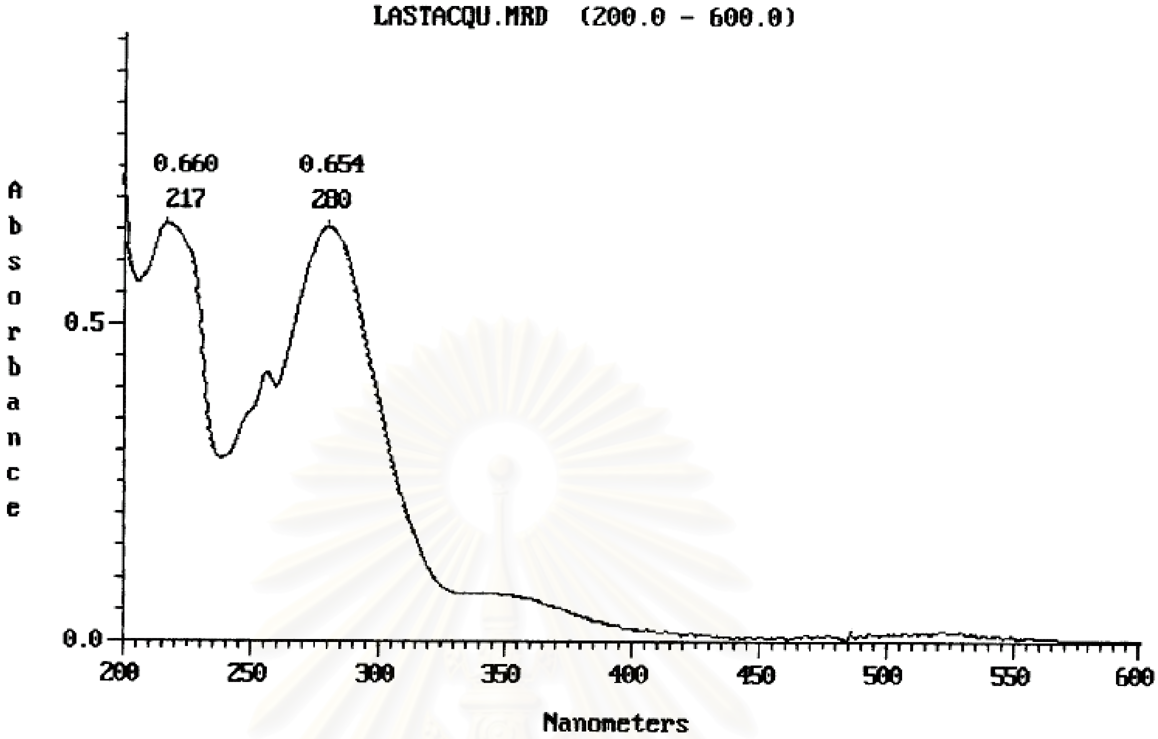


Figure 216 UV Spectrum of compound ME22 (methanol)

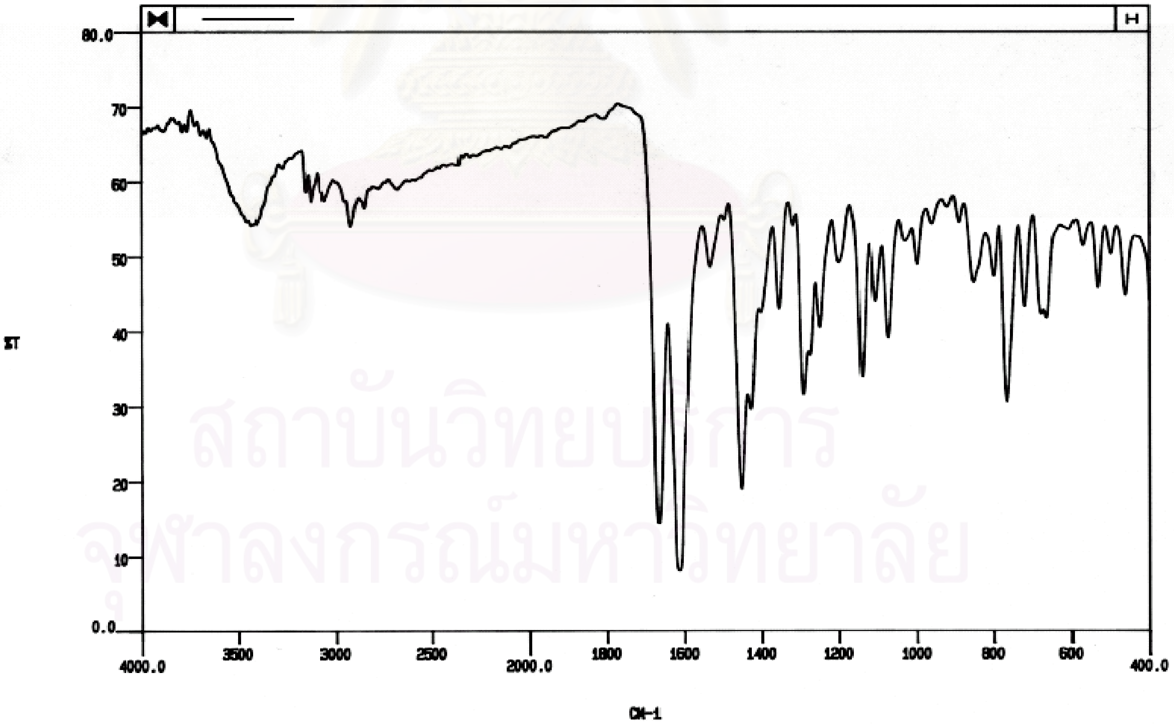
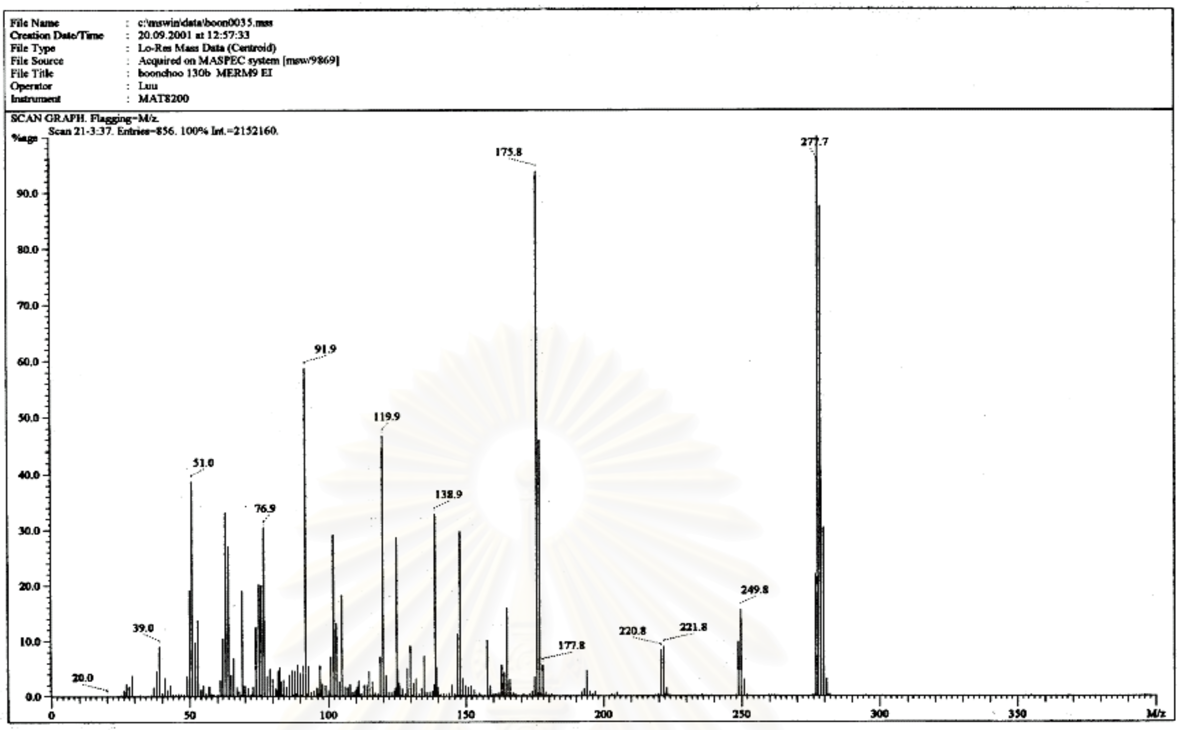


Figure 217 IR Spectrum of compound ME22 (KBr disc)



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Operator : Luu
Instrument : MAT8200

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Figure 218 EI Mass spectrum of compound ME22

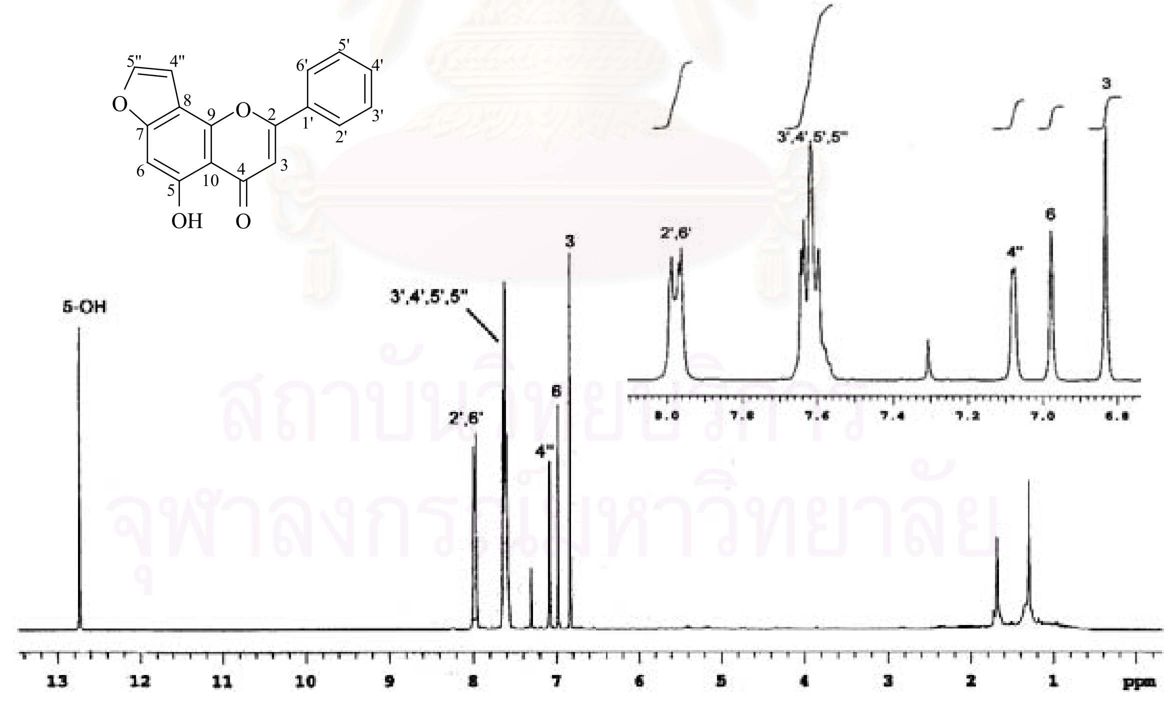


Figure 219 ^{1}H NMR (300 MHz) Spectrum of compound ME22 ($CDCl_3$)

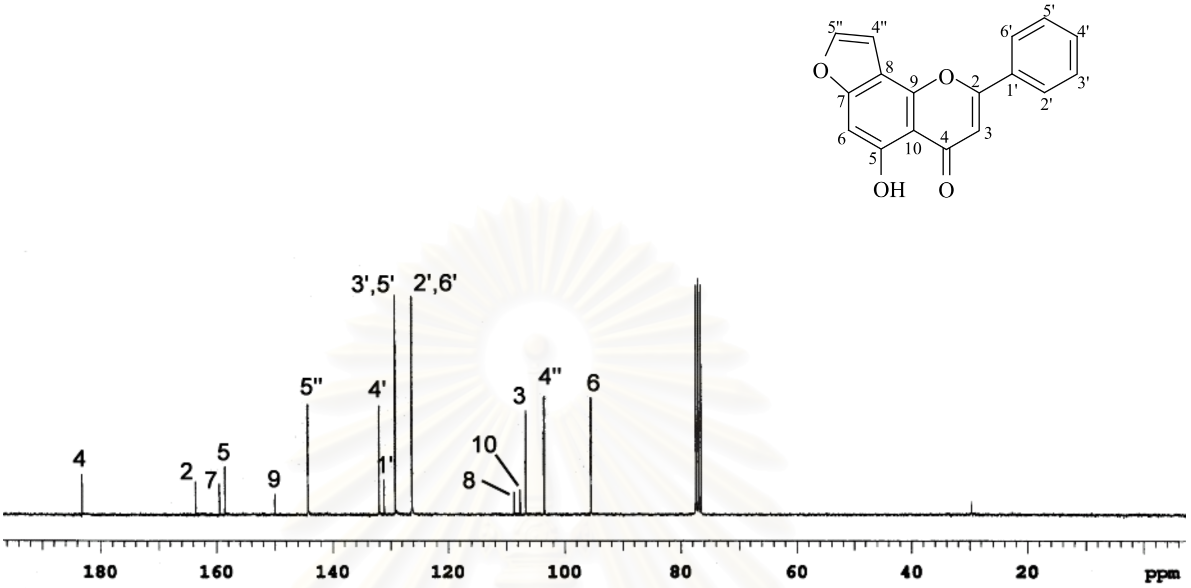


Figure 220 ^{13}C NMR (75 MHz) Spectrum of compound ME22 ($CDCl_3$)

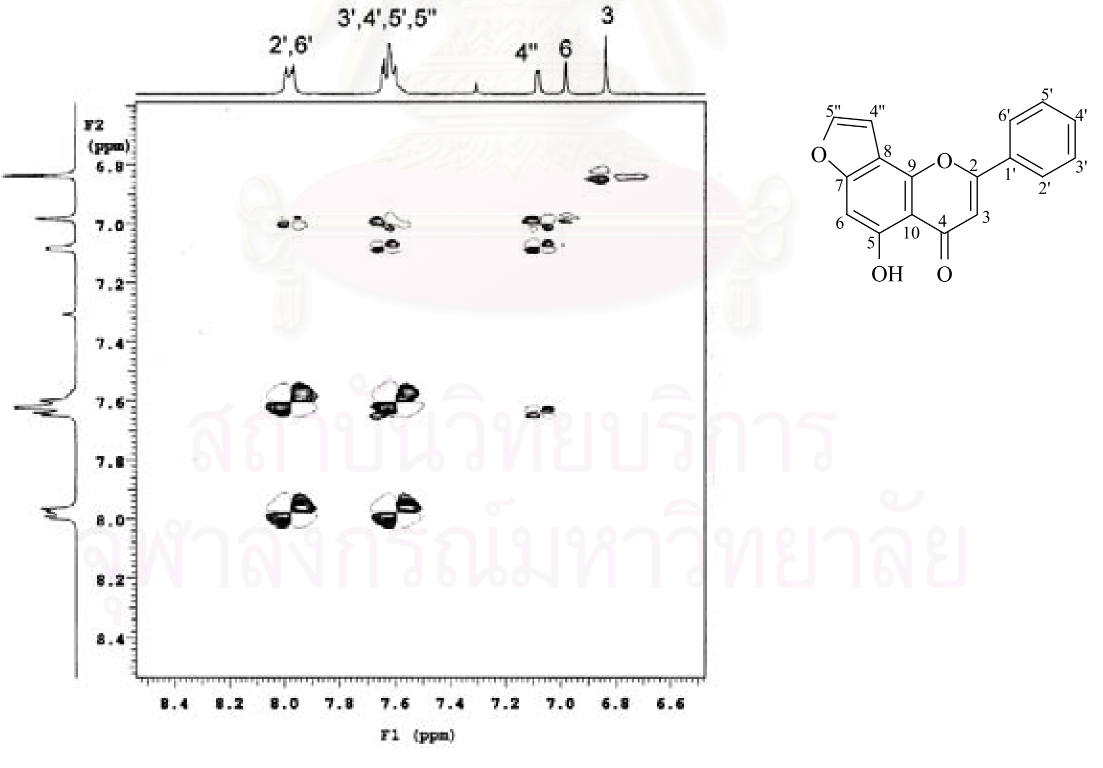


Figure 221 ^{1}H-^{1}H COSY Spectrum of compound ME22 ($CDCl_3$)

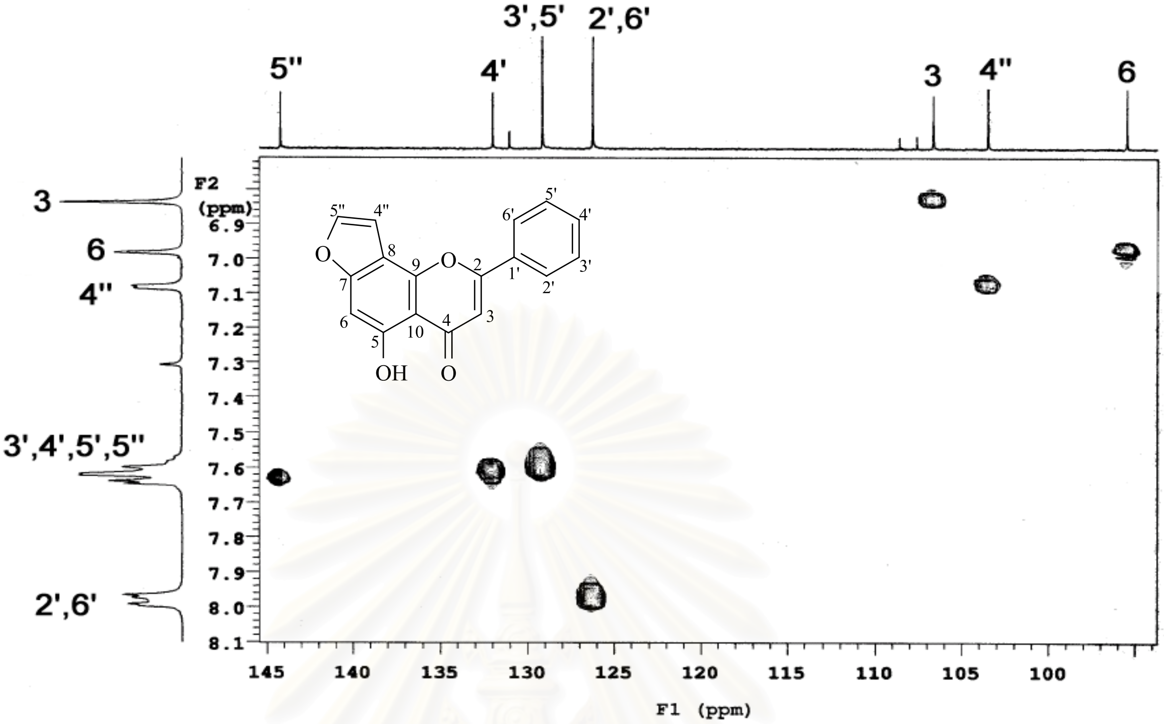


Figure 222 HSQC Spectrum of compound ME22 ($CDCl_3$)

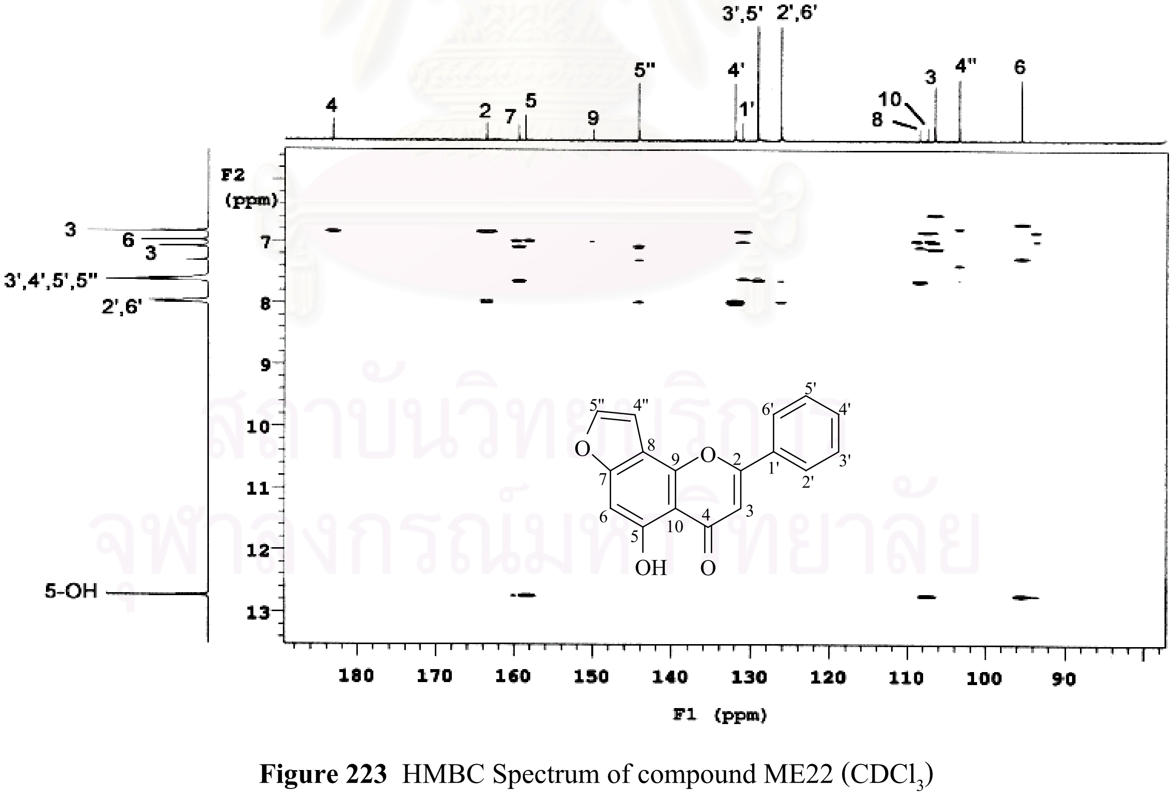


Figure 223 HMBC Spectrum of compound ME22 ($CDCl_3$)

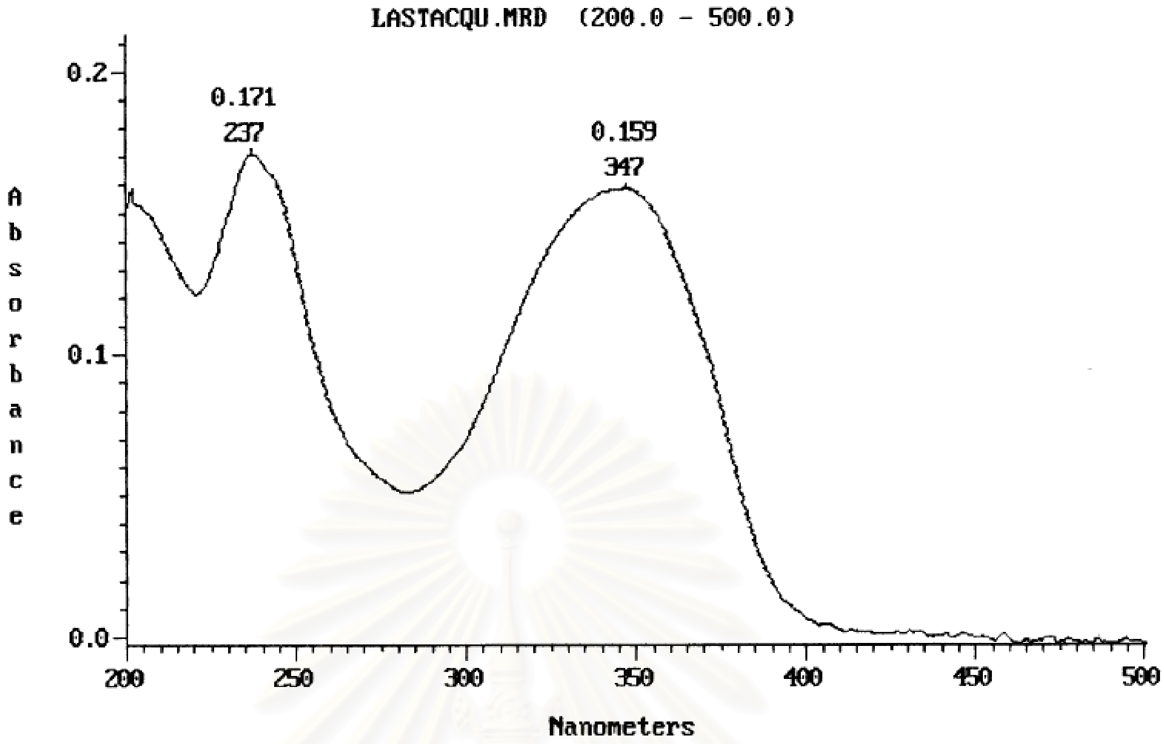


Figure 224 UV Spectrum of compound ME23 (methanol)

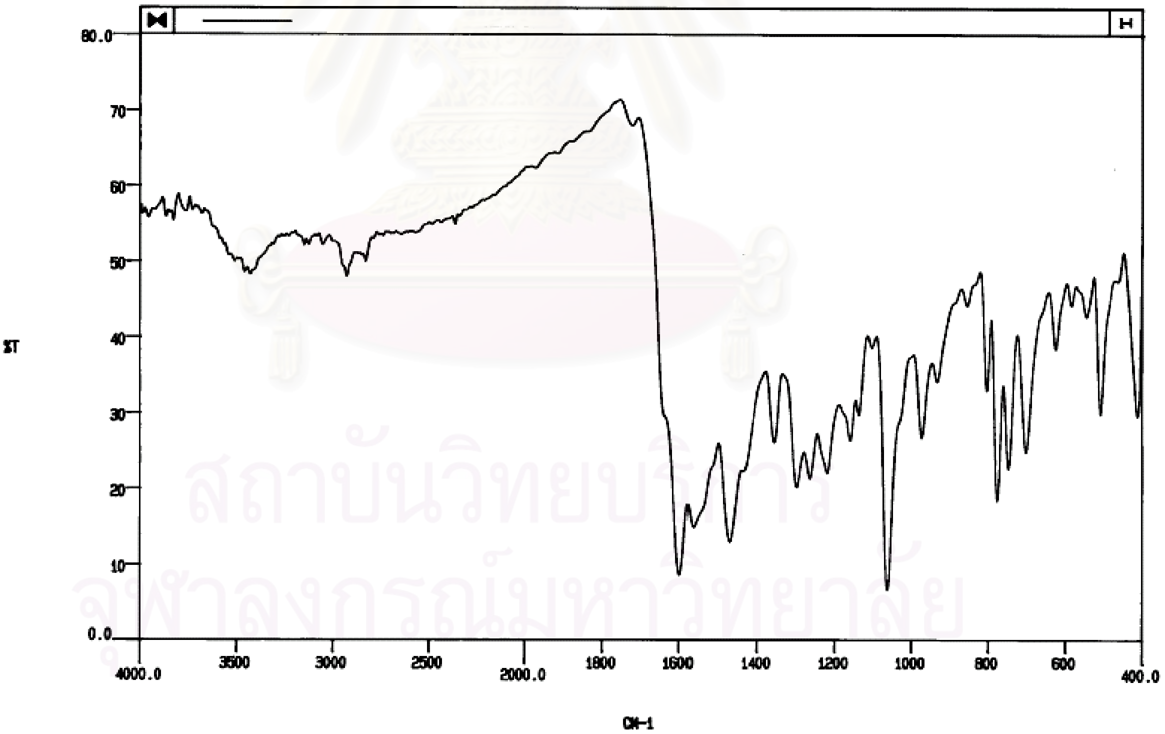
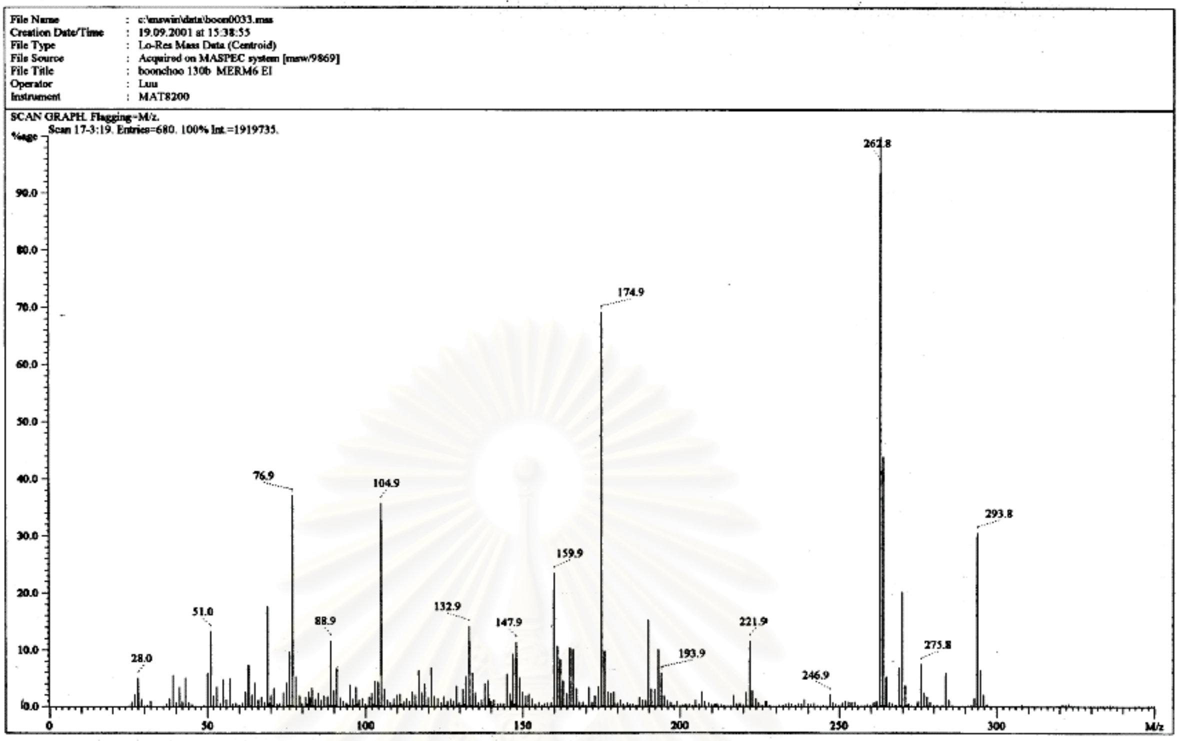


Figure 225 IR Spectrum of compound ME23 (KBr disc)



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Instrument : MAT8200

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Figure 226 EI Mass spectrum of compound ME23

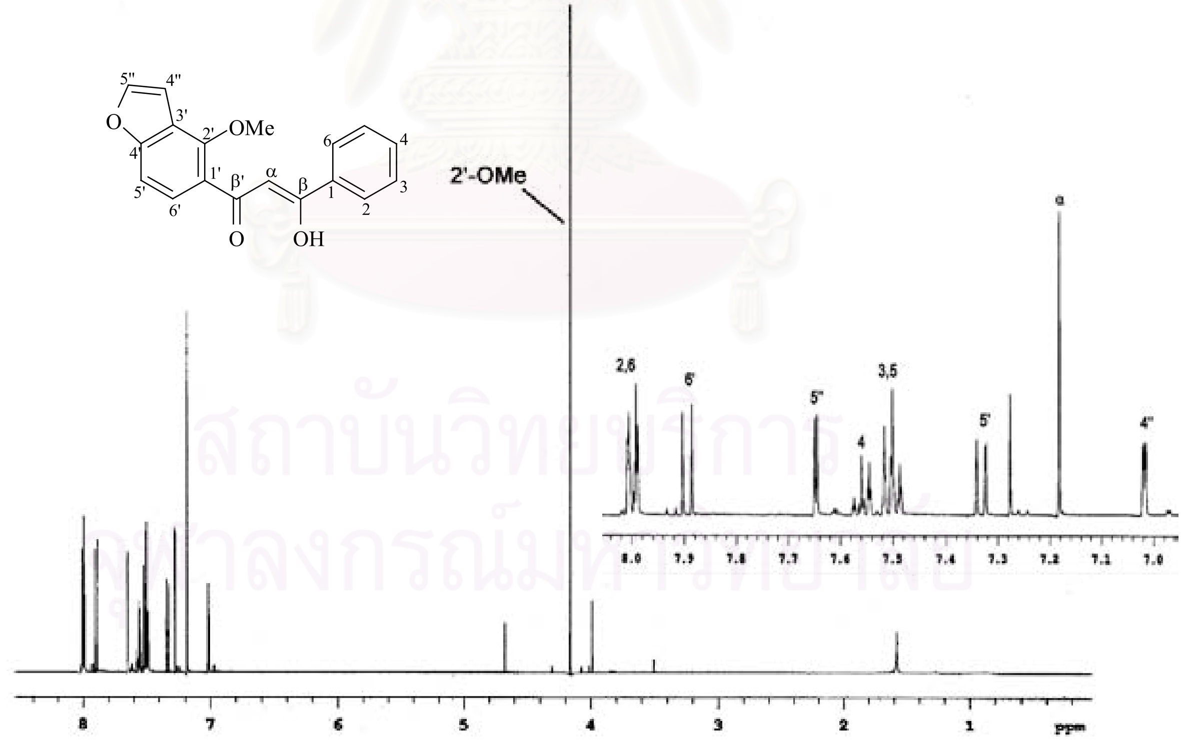


Figure 227 ^{1}H NMR (500 MHz) Spectrum of compound ME23 (CDCl$_3$)

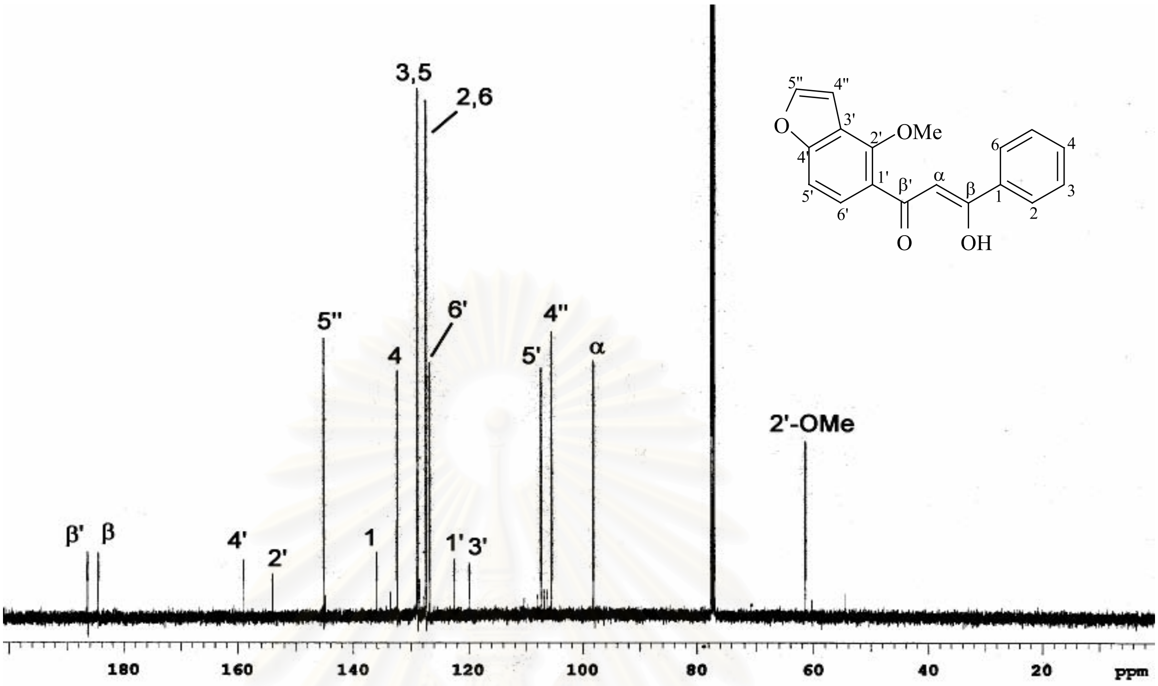


Figure 228 ^{13}C NMR (125 MHz) Spectrum of compound ME23 ($CDCl_3$)

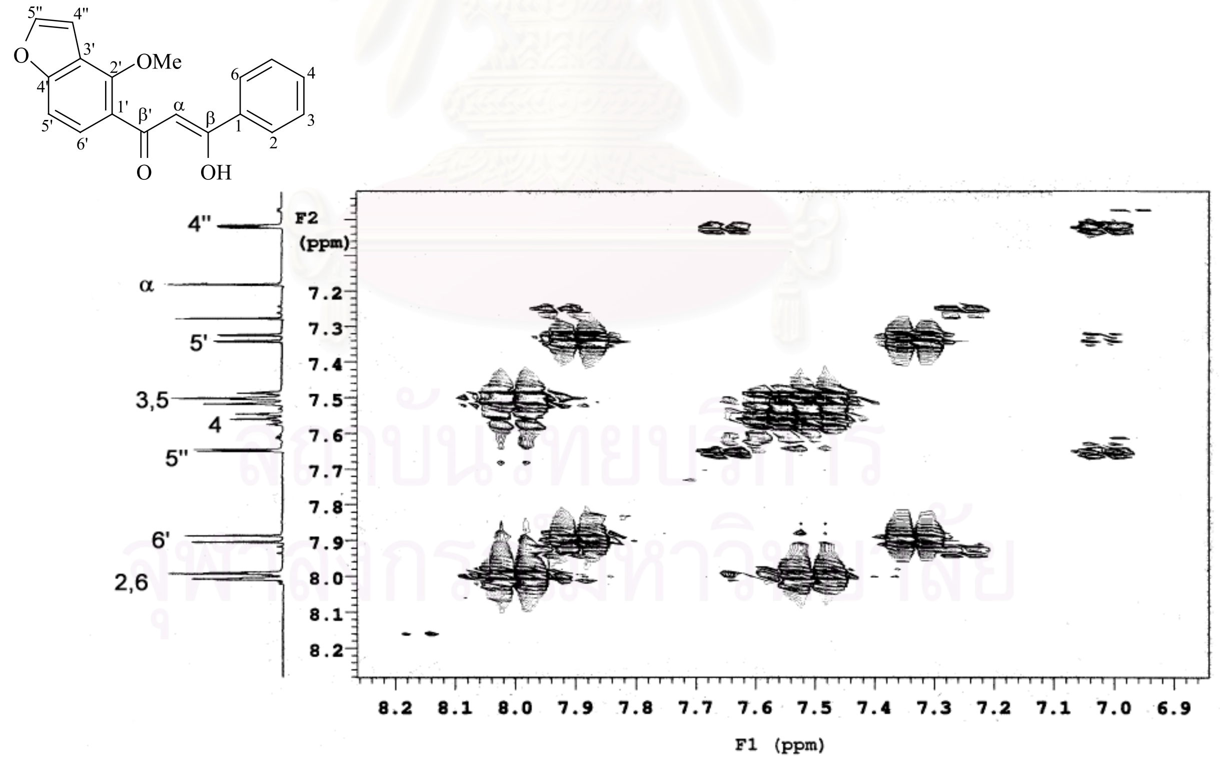


Figure 229 ^{1}H-^{1}H COSY Spectrum of compound ME23 ($CDCl_3$)

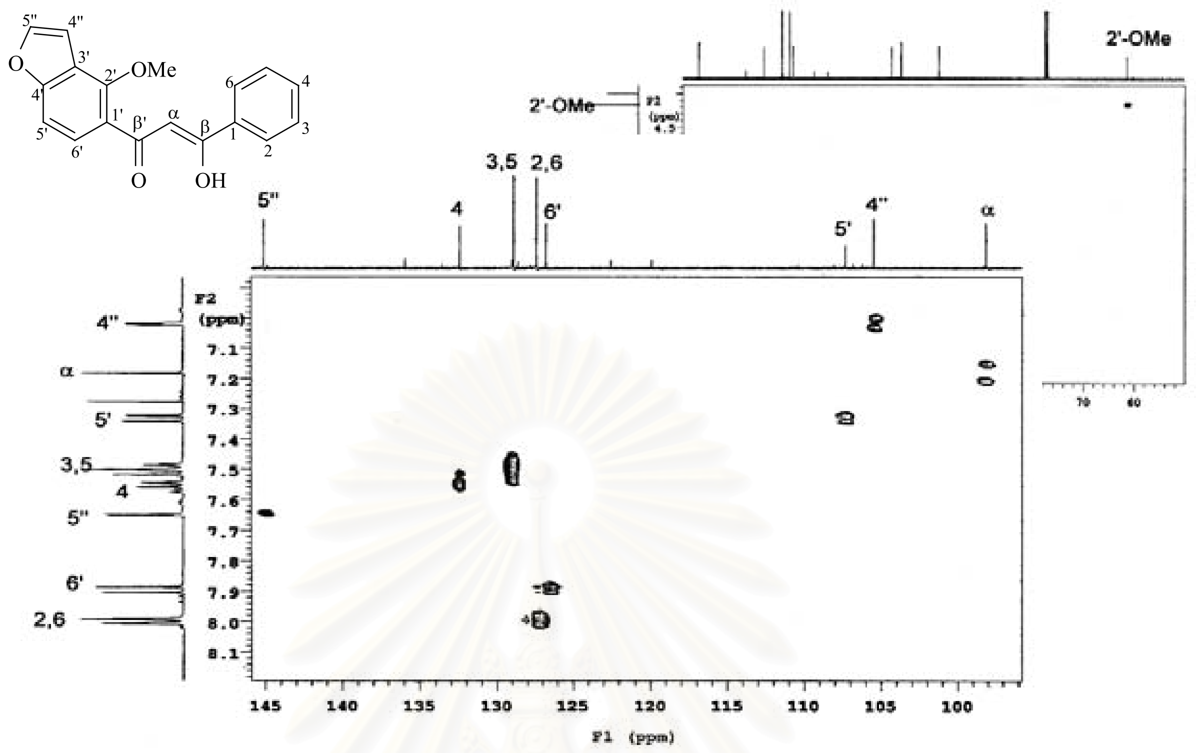


Figure 230 HSQC Spectrum of compound ME23 ($CDCl_3$)

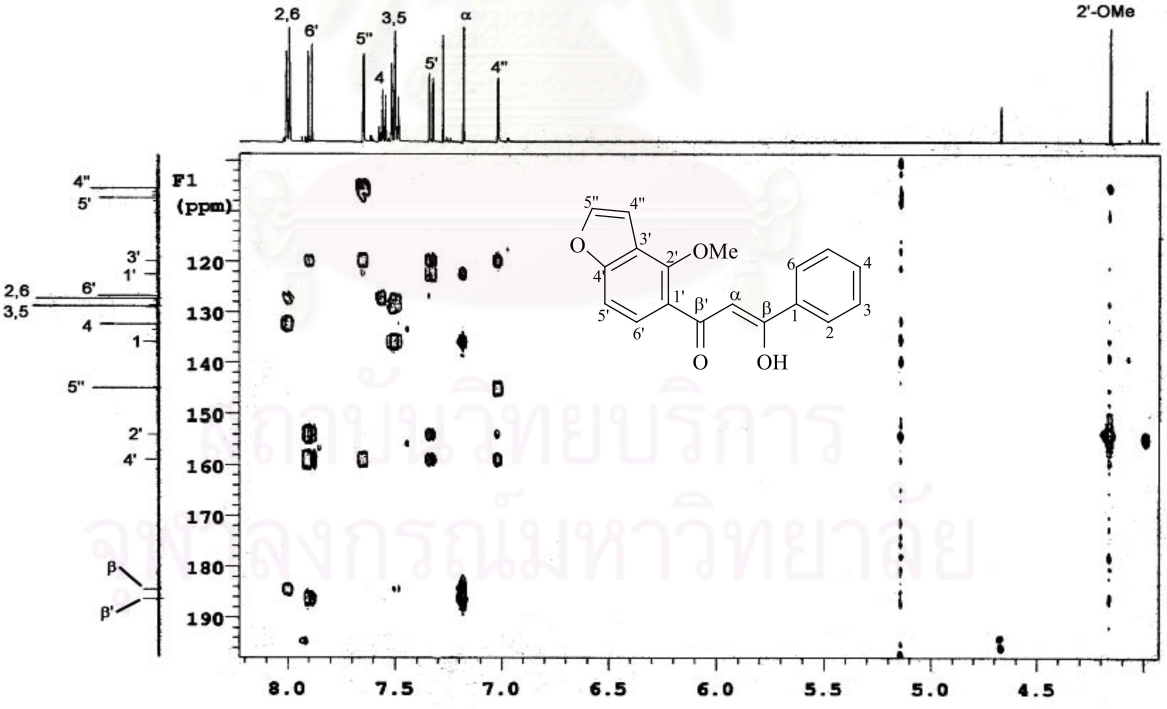


Figure 231 HMBC Spectrum of compound ME23 ($CDCl_3$)

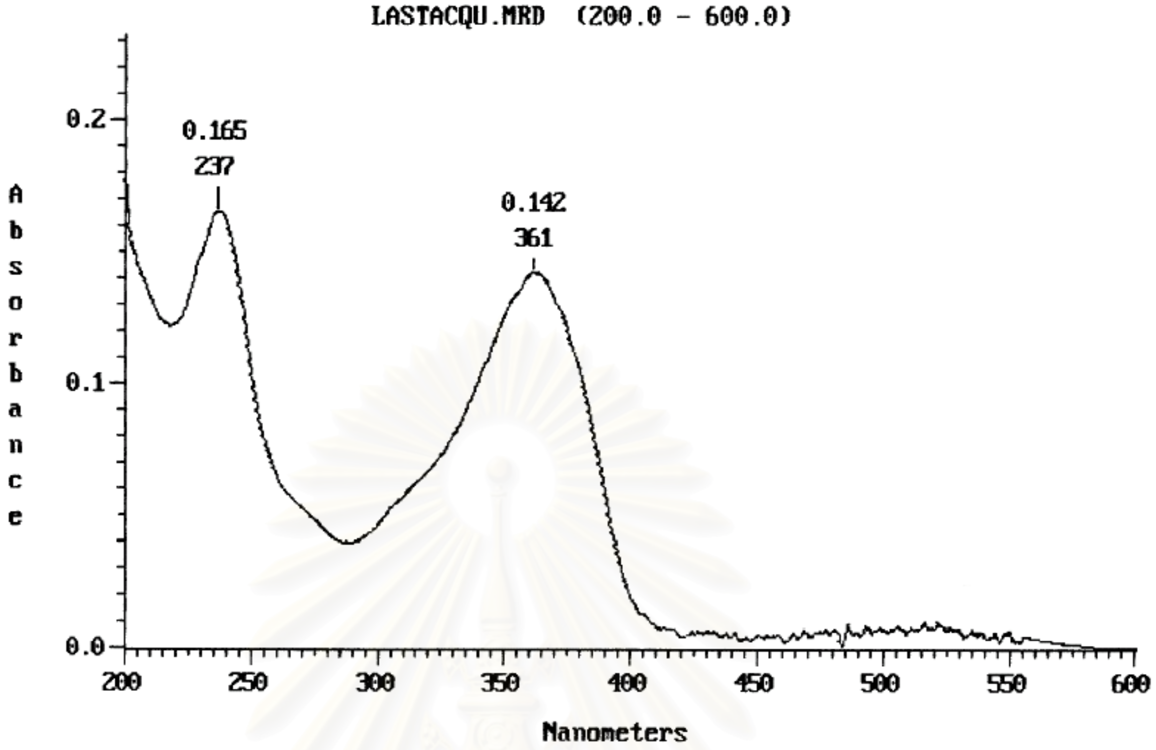


Figure 232 UV Spectrum of compound ME24 (methanol)

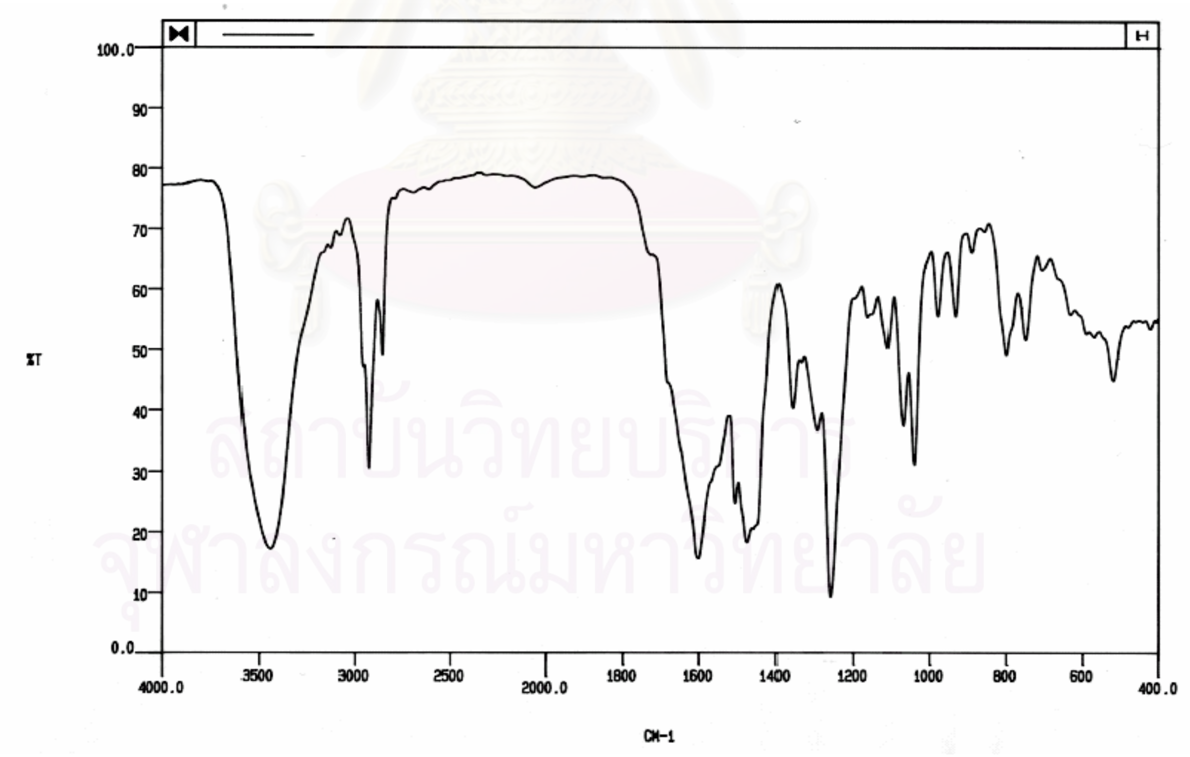
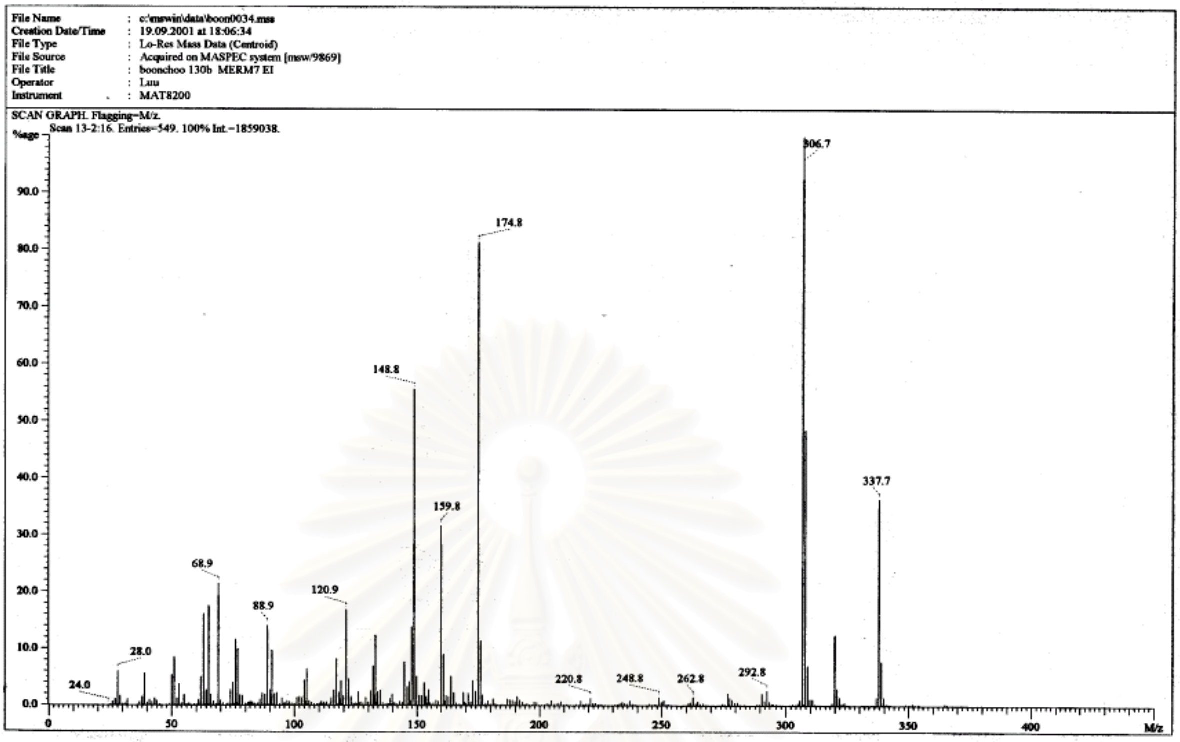


Figure 233 IR Spectrum of compound ME24 (KBr disc)



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Operator : Luu
Instrument : MAT8200

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Figure 234 EI Mass spectrum of compound ME24

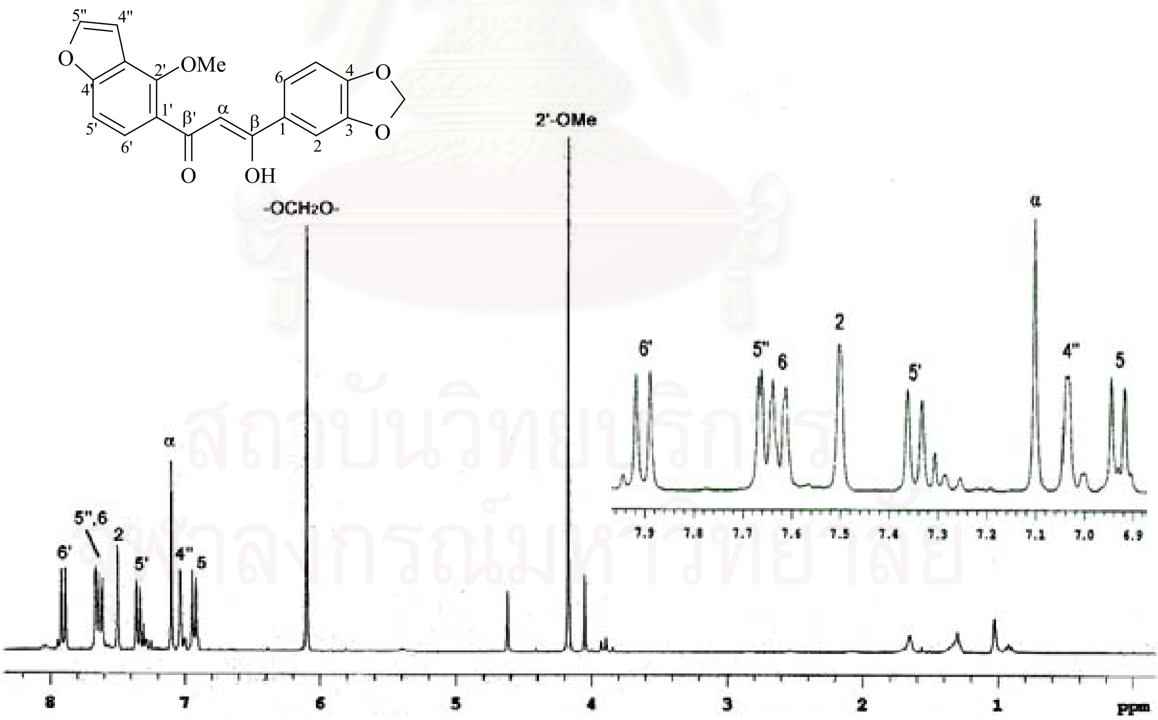


Figure 235 ^{1}H NMR (300 MHz) Spectrum of compound ME24 ($CDCl_3$)

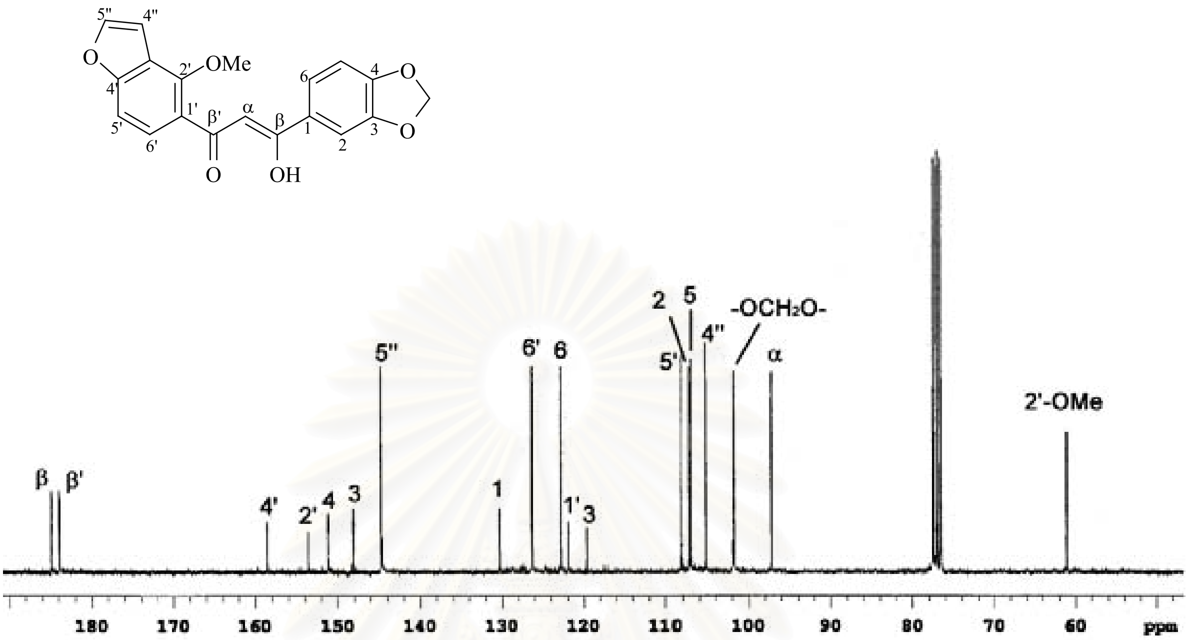


Figure 236 ^{13}C NMR (75 MHz) Spectrum of compound ME24 (CDCl$_3$)

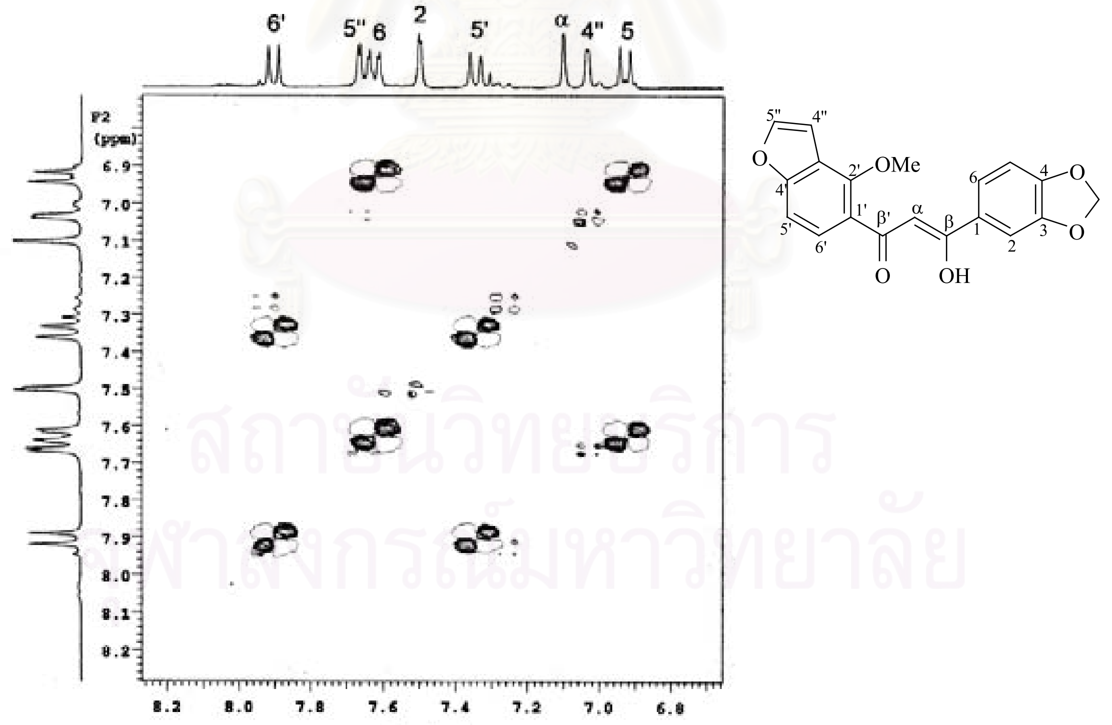


Figure 237 ^{1}H-^{1}H COSY Spectrum of compound ME24 (CDCl$_3$)

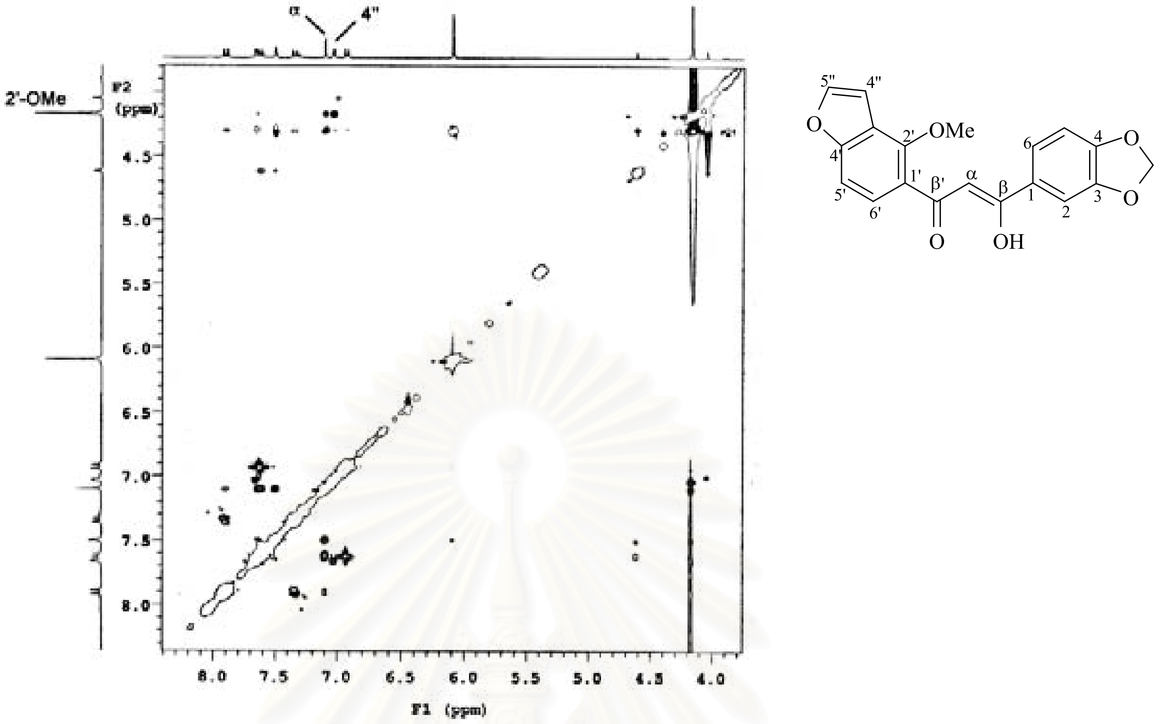


Figure 238 NOESY Spectrum of compound ME24 ($CDCl_3$)

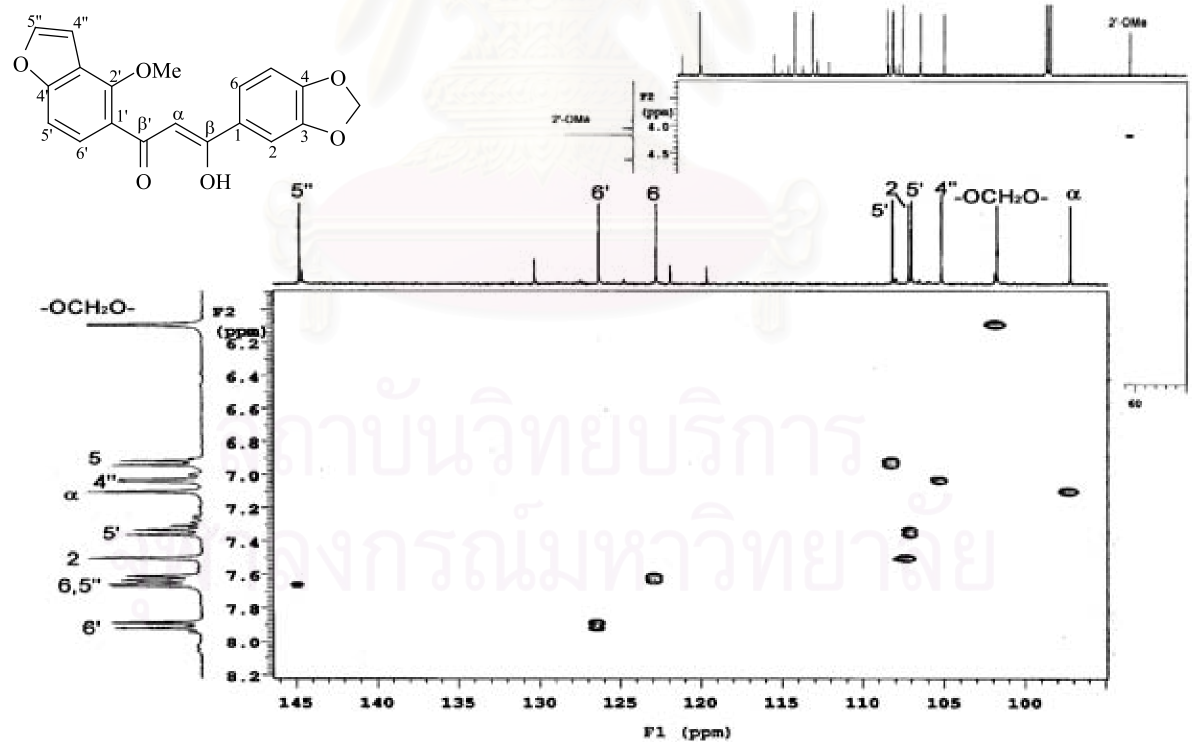


Figure 239 HSQC Spectrum of compound ME24 ($CDCl_3$)

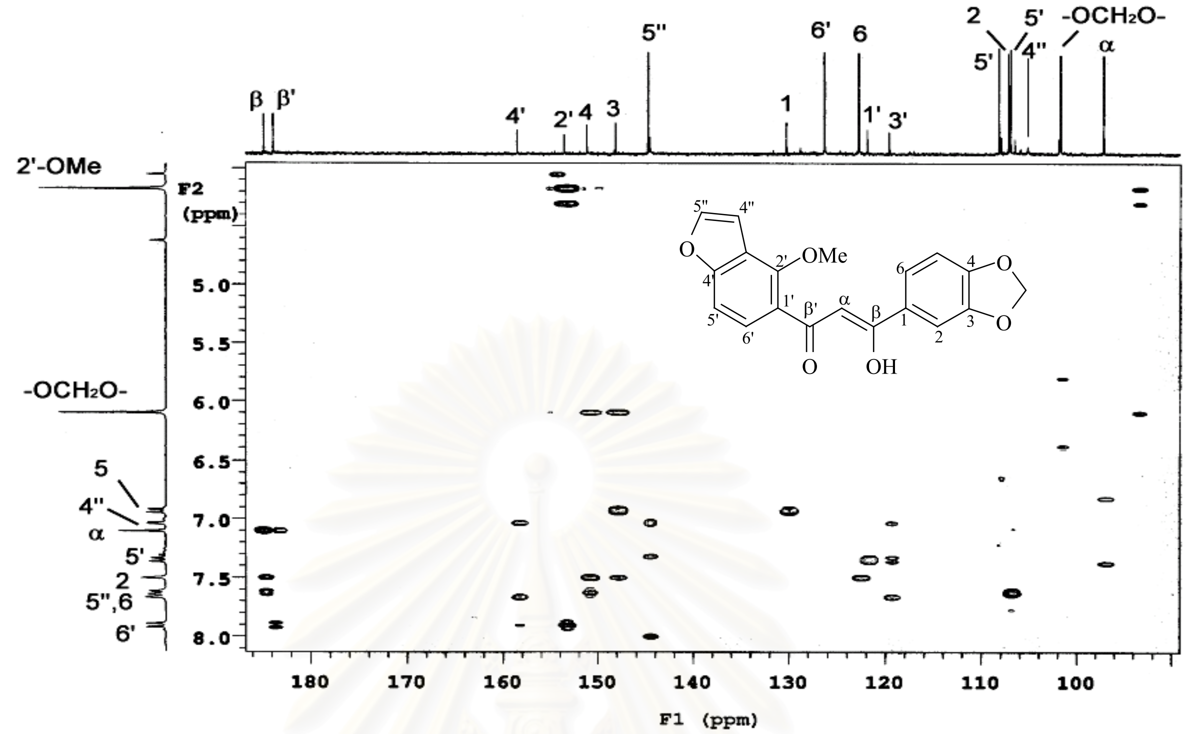


Figure 240 HMBC Spectrum of compound ME24 ($CDCl_3$) [δ_H 4.0-8.1 ppm, δ_C 90-186 ppm]

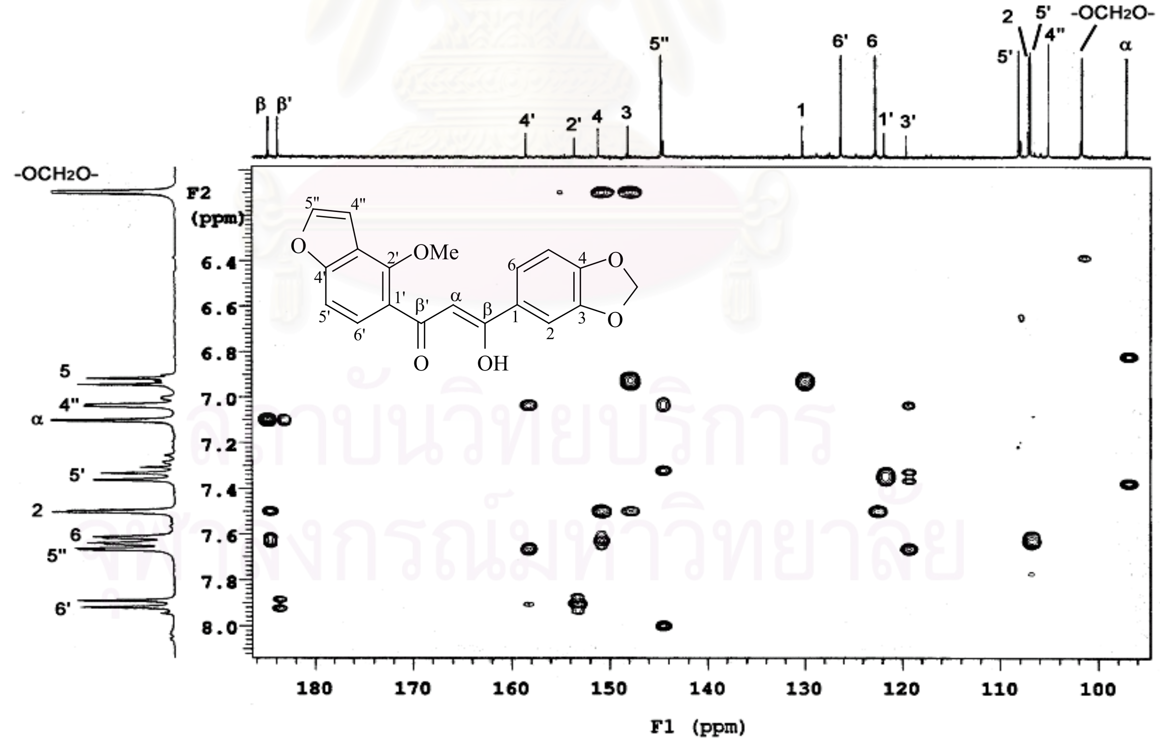


Figure 241 HMBC Spectrum of compound ME24 ($CDCl_3$) [δ_H 6.0-8.1 ppm, δ_C 90-186 ppm]

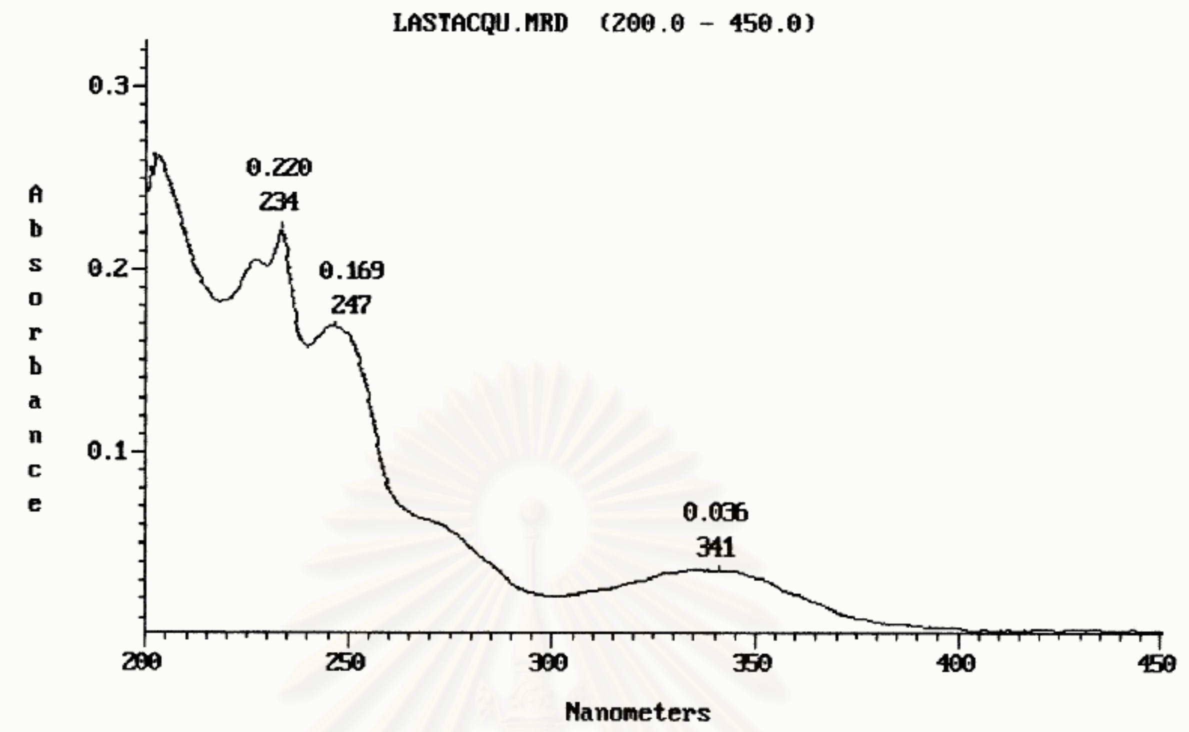


Figure 242 UV Spectrum of compound ME25 (methanol)

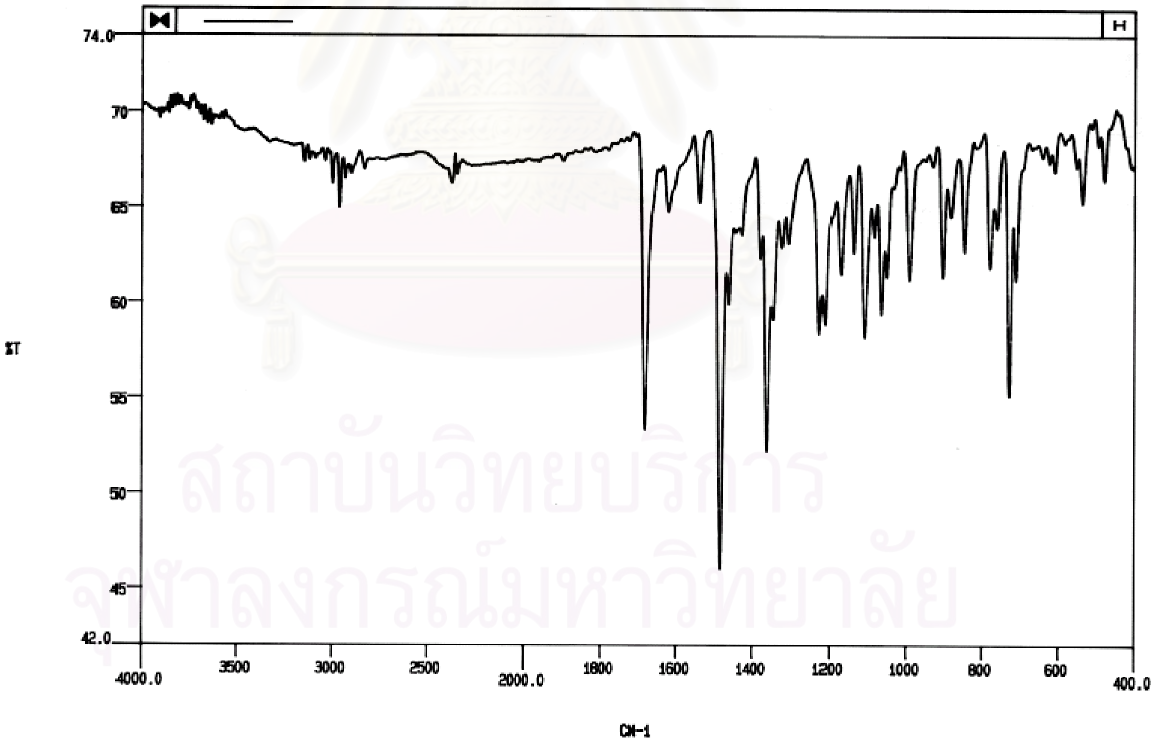
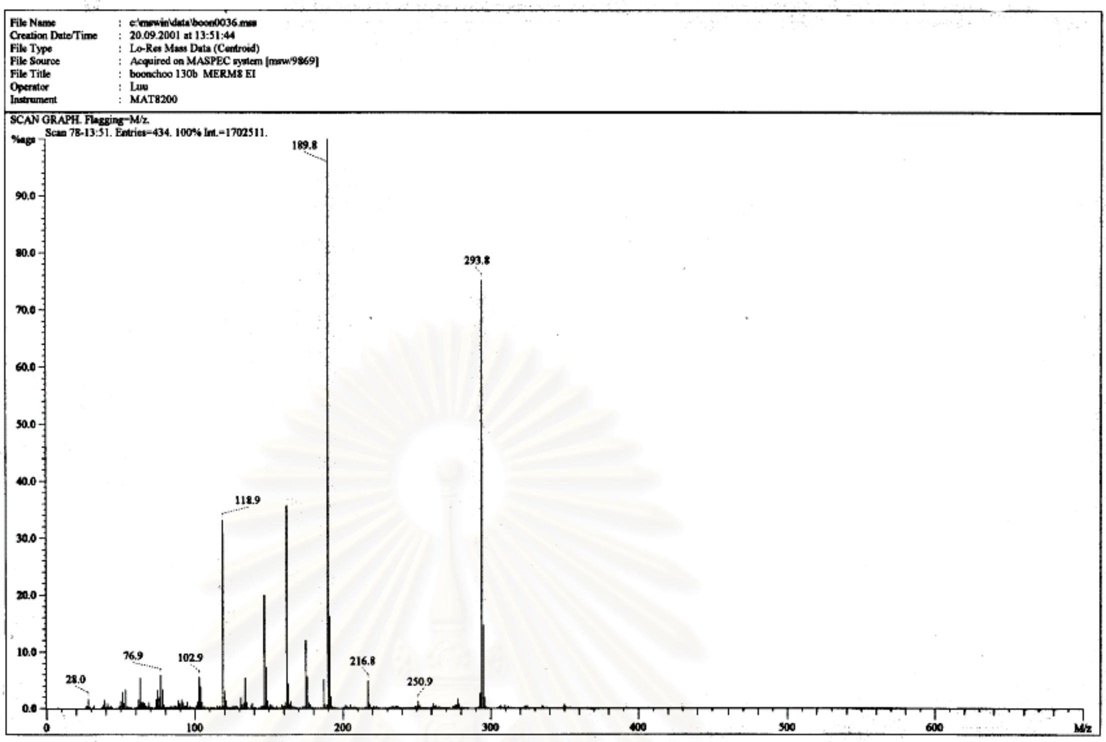


Figure 243 IR Spectrum of compound ME25 (KBr disc)



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Operator : Luu
Instrument : MAT8200

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Figure 244 EI Mass spectrum of compound ME25

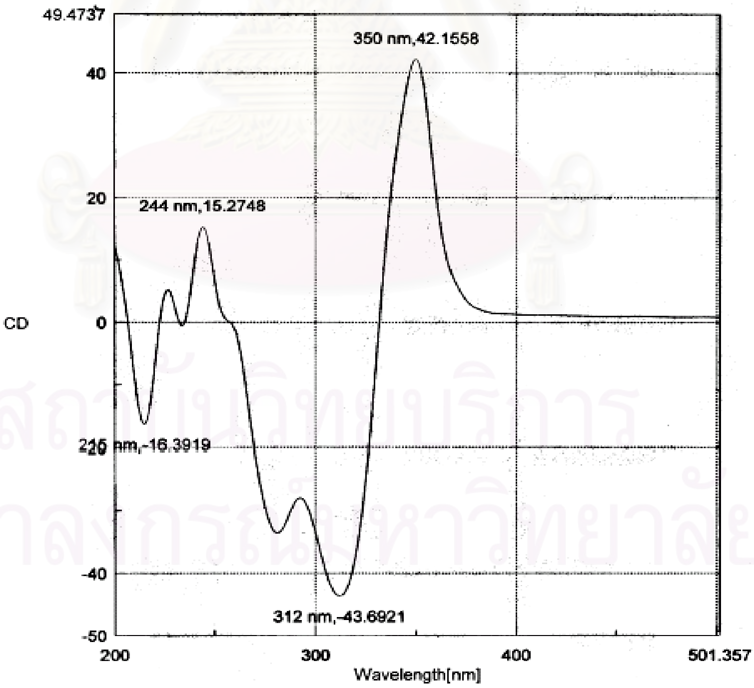


Figure 245 CD Spectrum of compound ME25 (methanol)

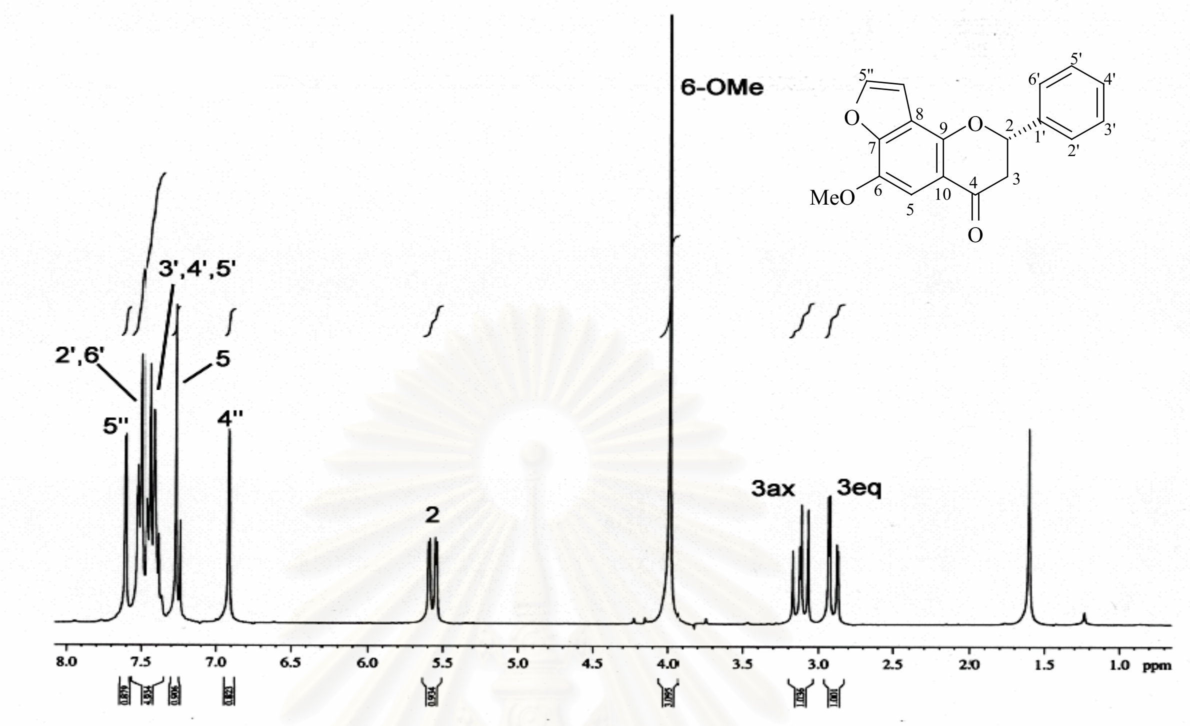


Figure 246 ^{1}H NMR (300 MHz) Spectrum of compound ME25 ($CDCl_3$)

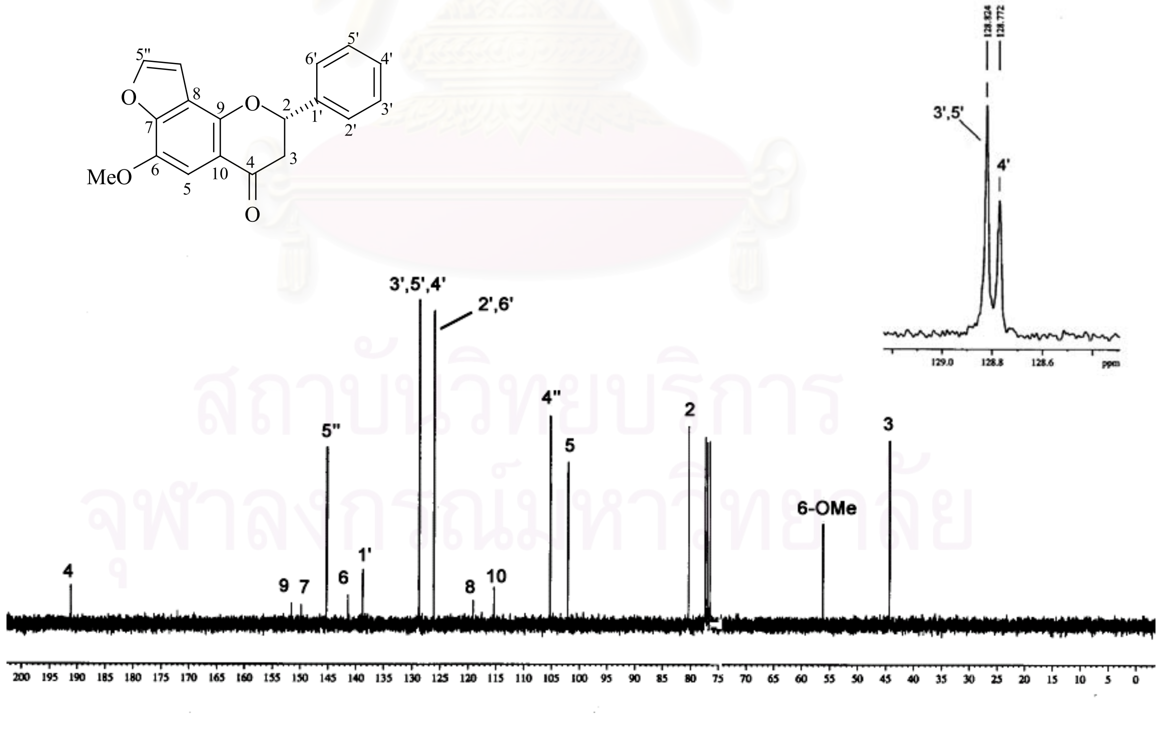


Figure 247 ^{13}C NMR (75 MHz) Spectrum of compound ME25 ($CDCl_3$)

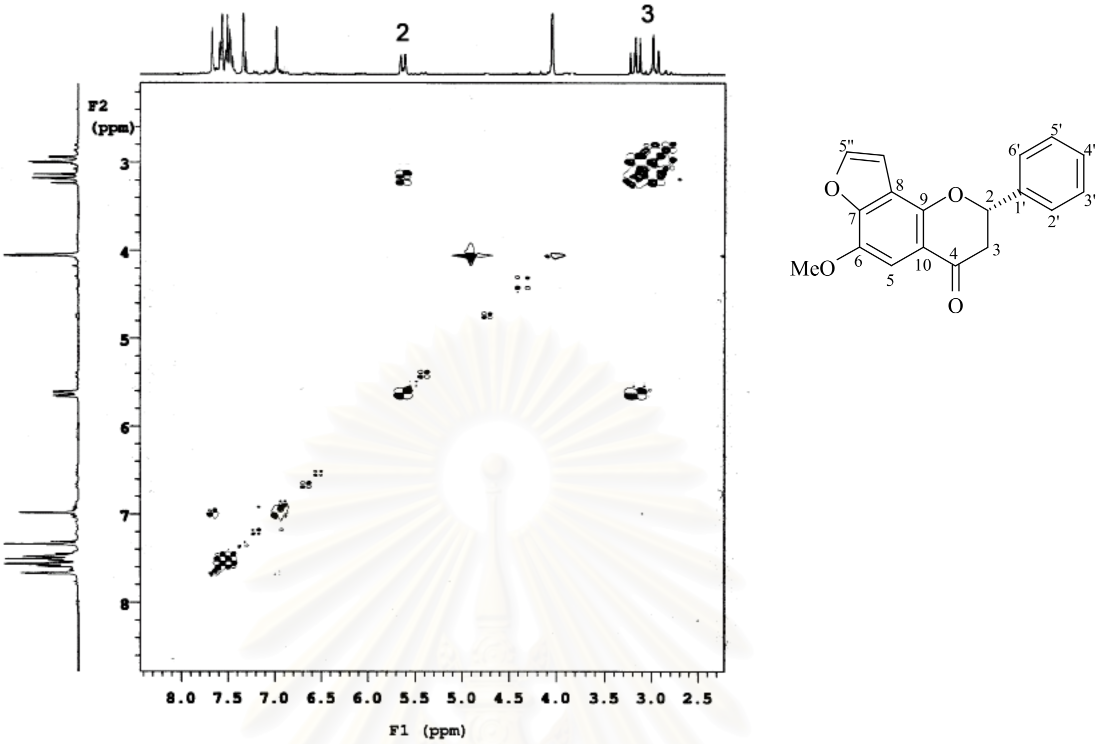


Figure 248 ^{1}H-^{1}H COSY Spectrum of compound ME25 (CDCl$_3$)

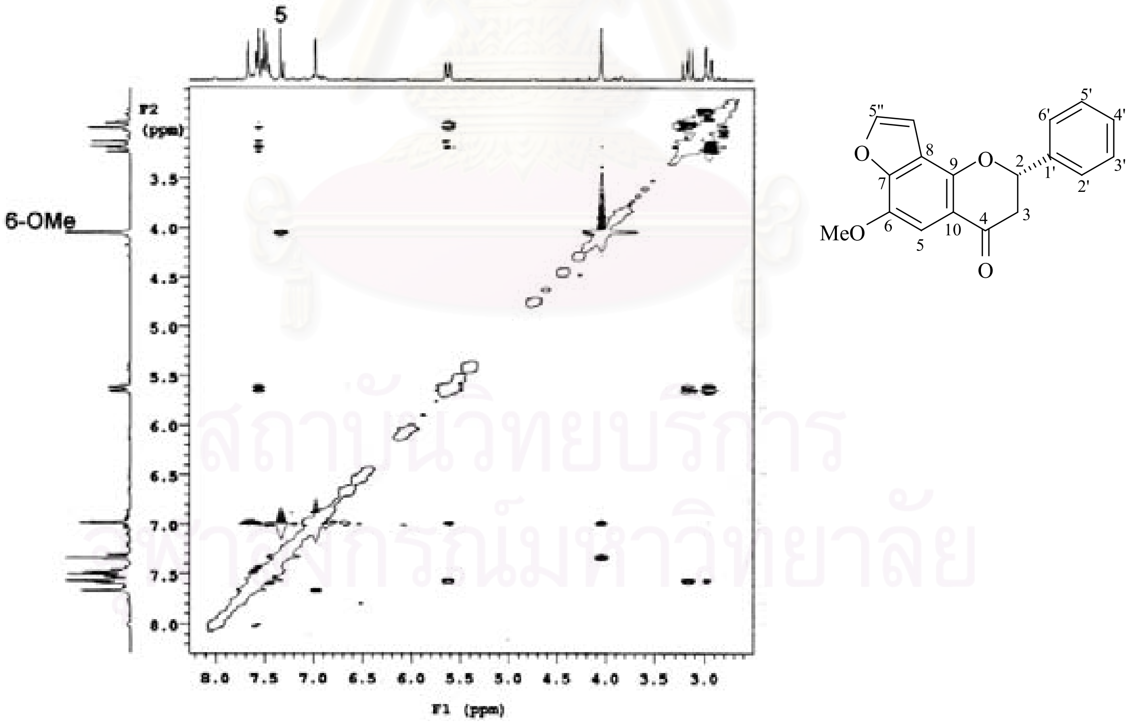


Figure 249 NOESY Spectrum of compound ME25 (CDCl$_3$)

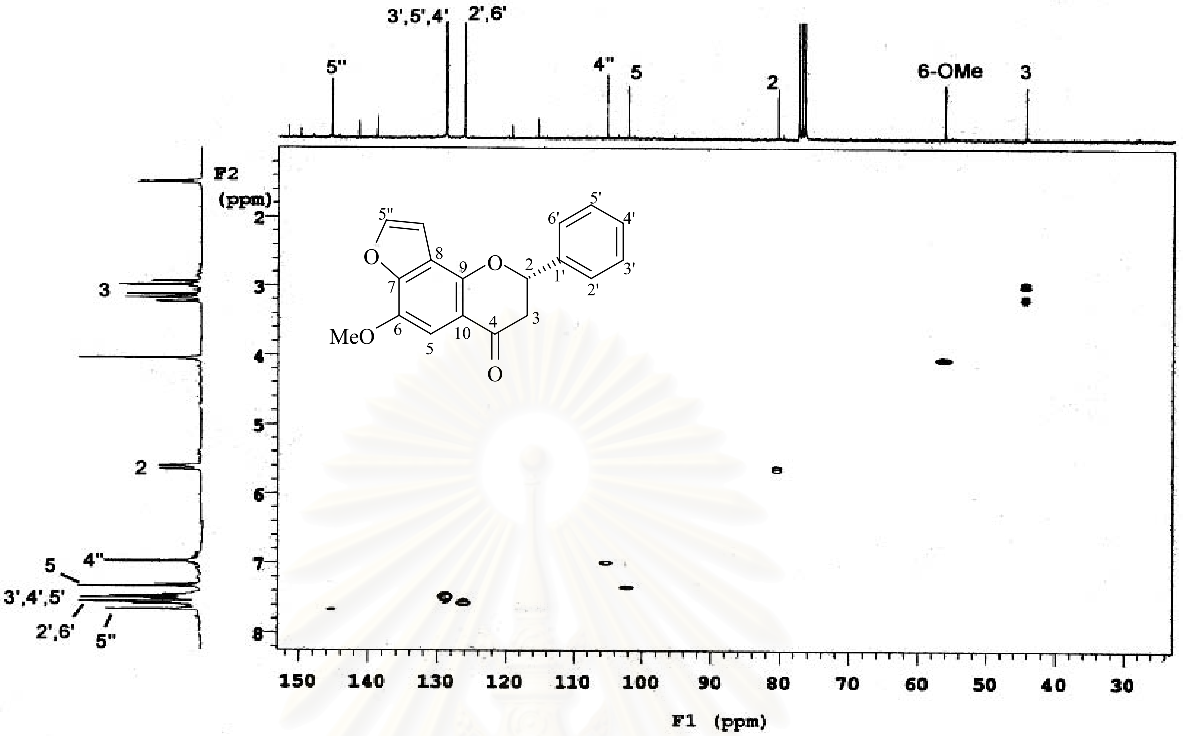


Figure 250 HSQC Spectrum of compound ME25 ($CDCl_3$)

Figure 251 HMBC Spectrum of compound ME25 ($CDCl_3$)

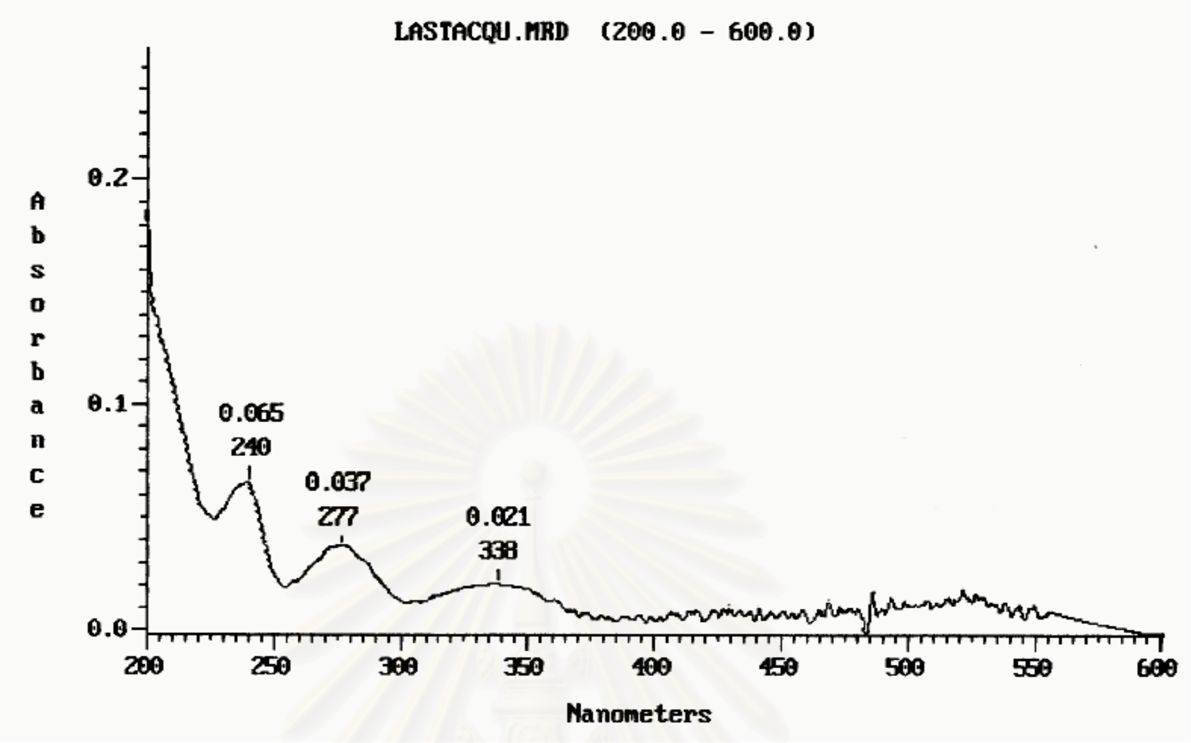


Figure 252 UV Spectrum of compound ME26 (methanol)

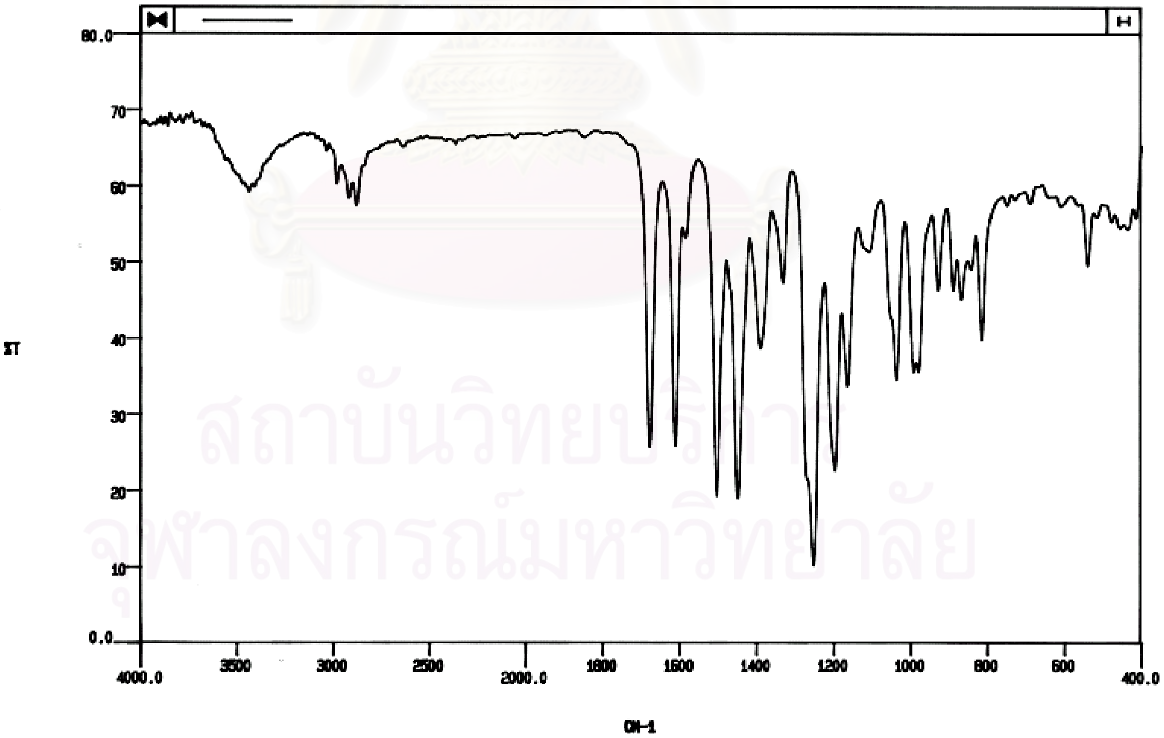
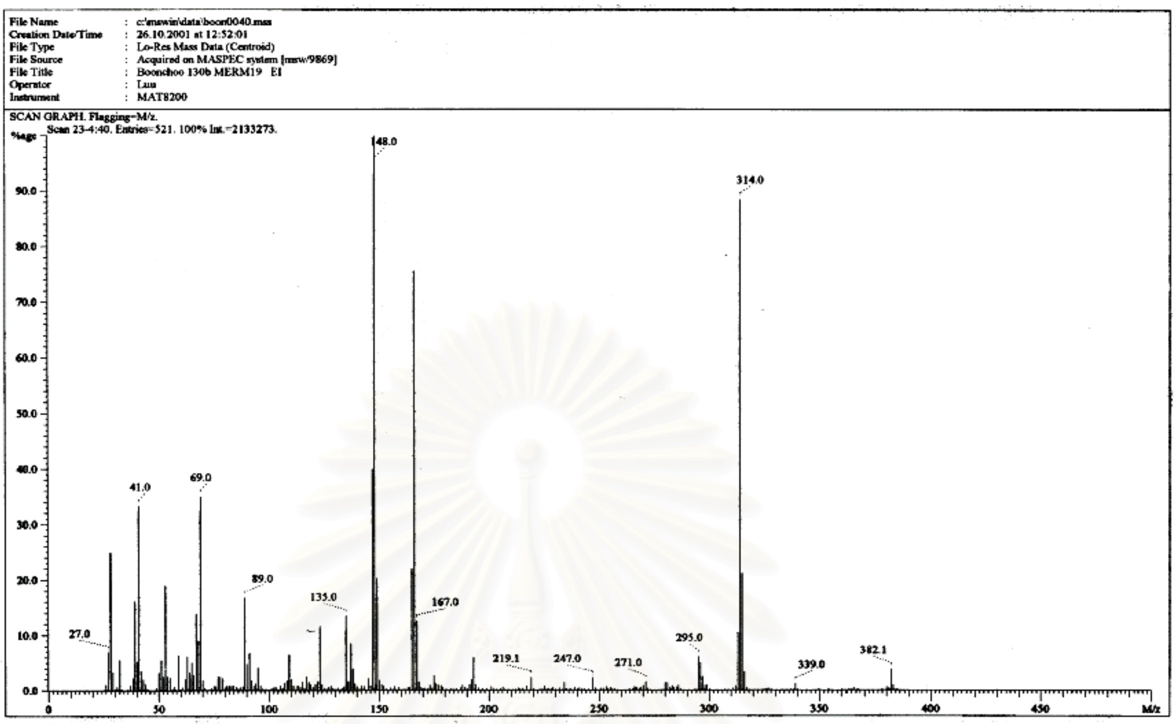


Figure 253 IR Spectrum of compound ME26 (KBr disc)



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Instrument : MAT8200

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Figure 254 EI Mass spectrum of compound ME26

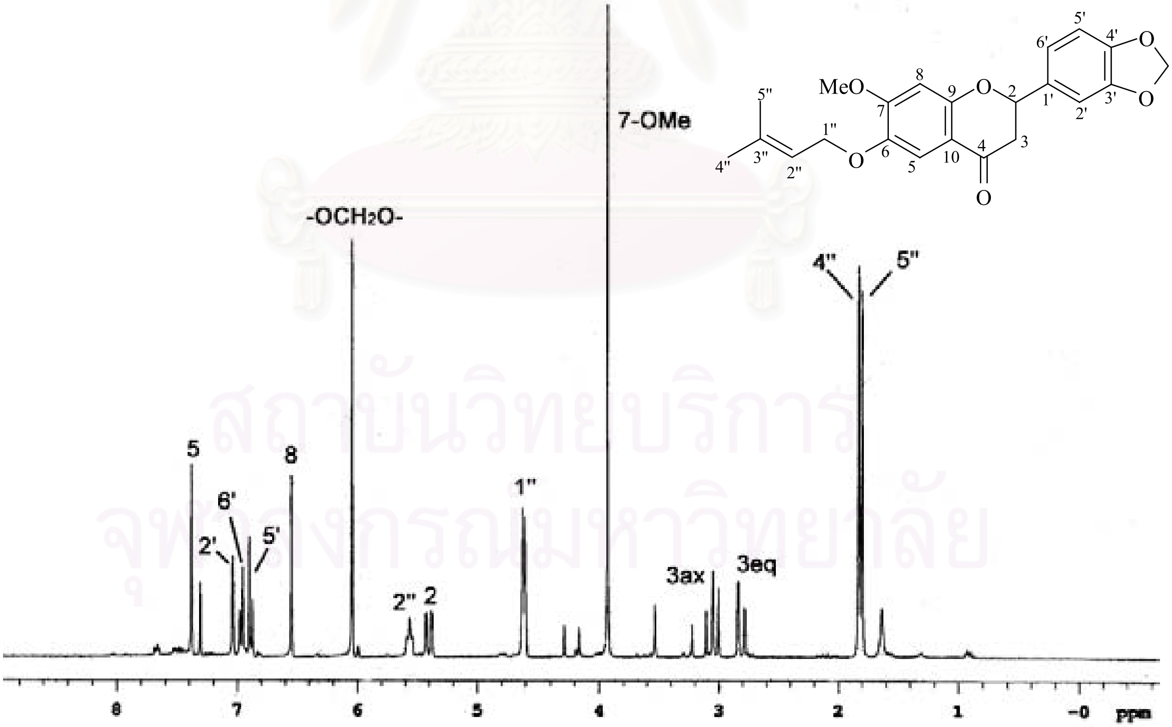


Figure 255 ^{1}H NMR (300 MHz) Spectrum of compound ME26 ($CDCl_3$)

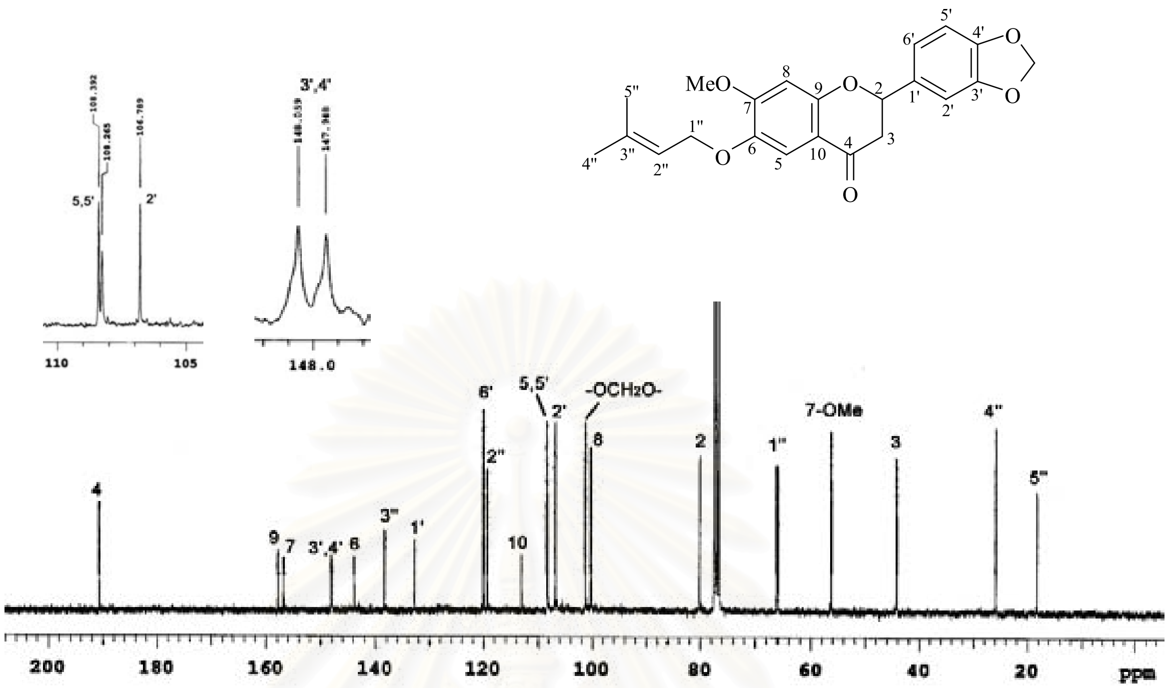


Figure 256 ^{13}C NMR (75 MHz) Spectrum of compound ME26 (CDCl$_3$)

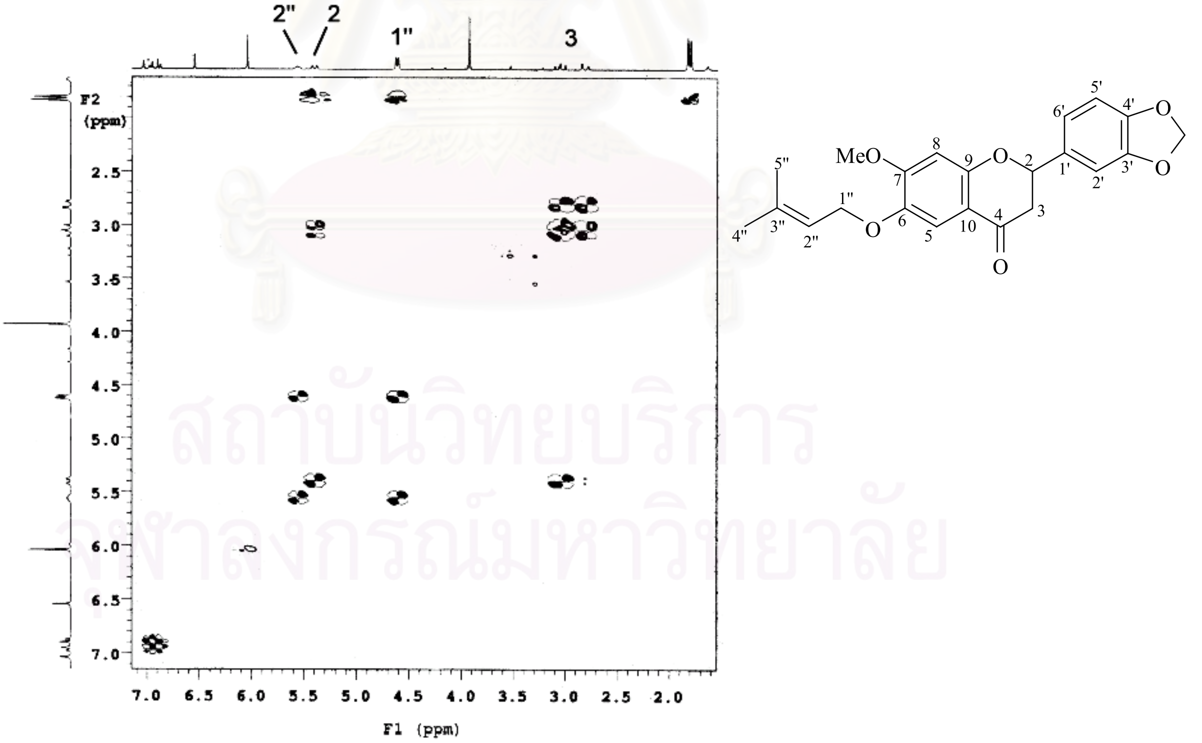


Figure 257 ^{1}H-^{1}H COSY Spectrum of compound ME26 (CDCl$_3$)

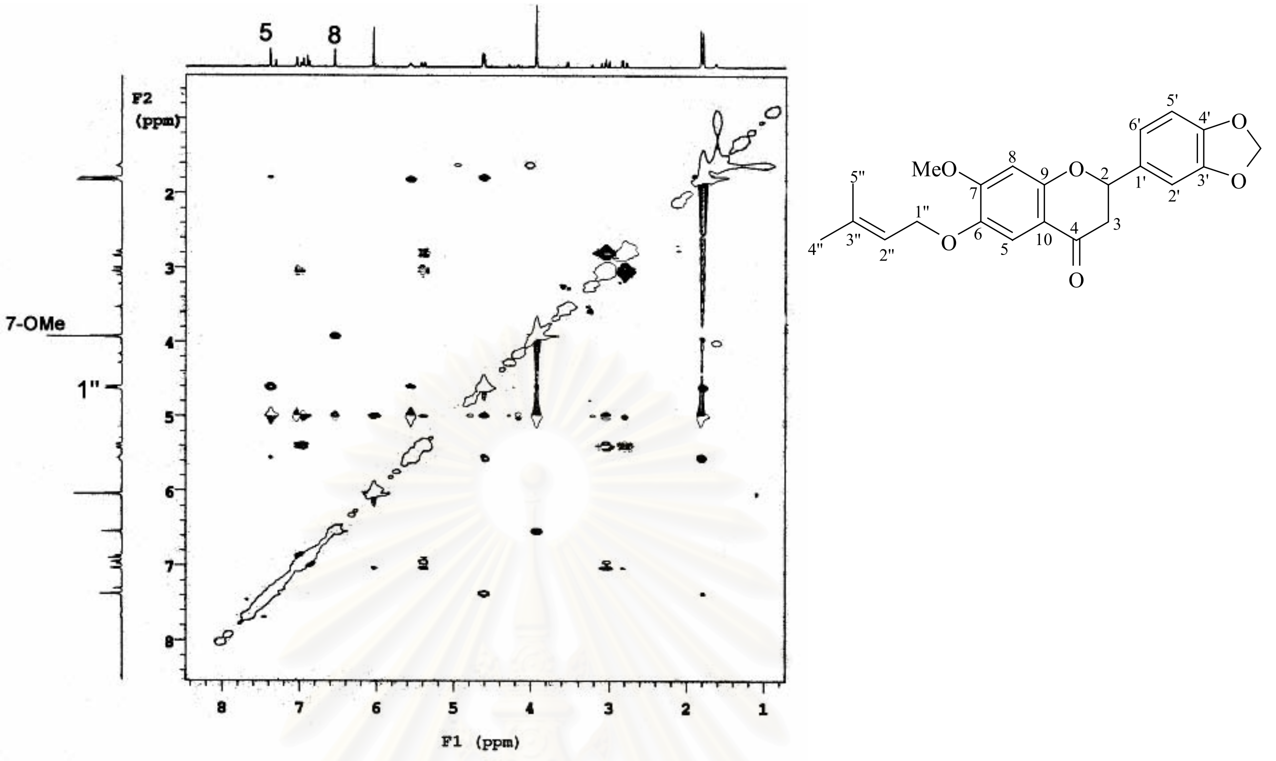


Figure 258 NOESY Spectrum of compound ME26 ($CDCl_3$)

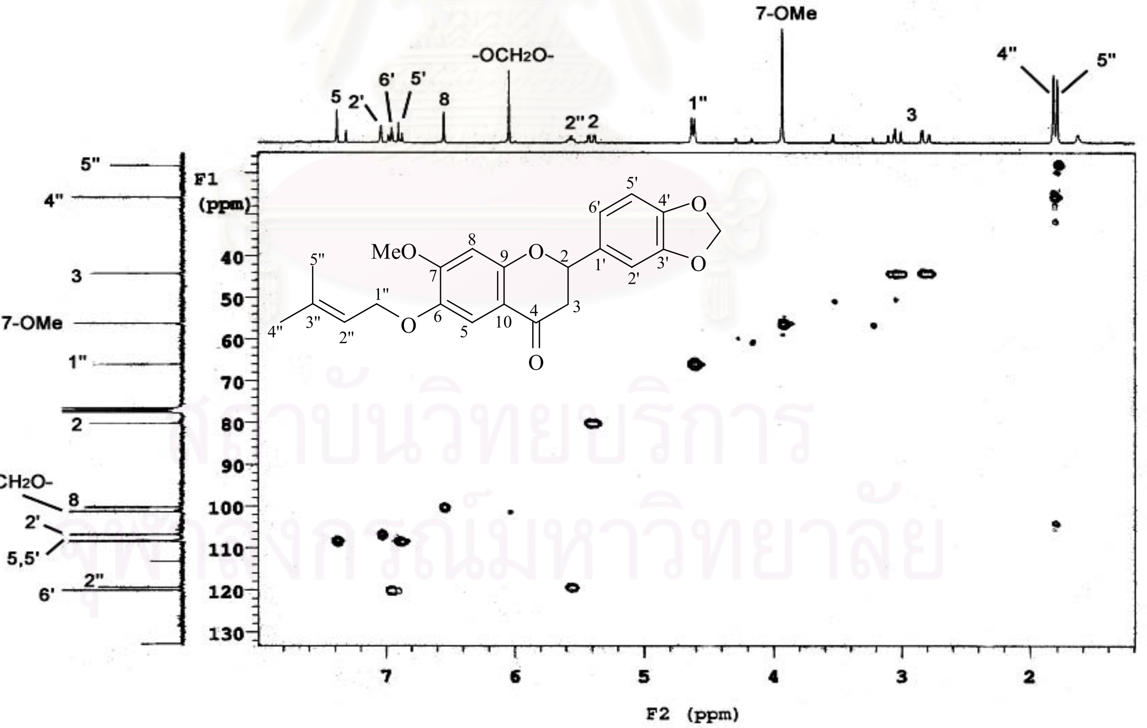


Figure 259 HSQC Spectrum of compound ME26 ($CDCl_3$)

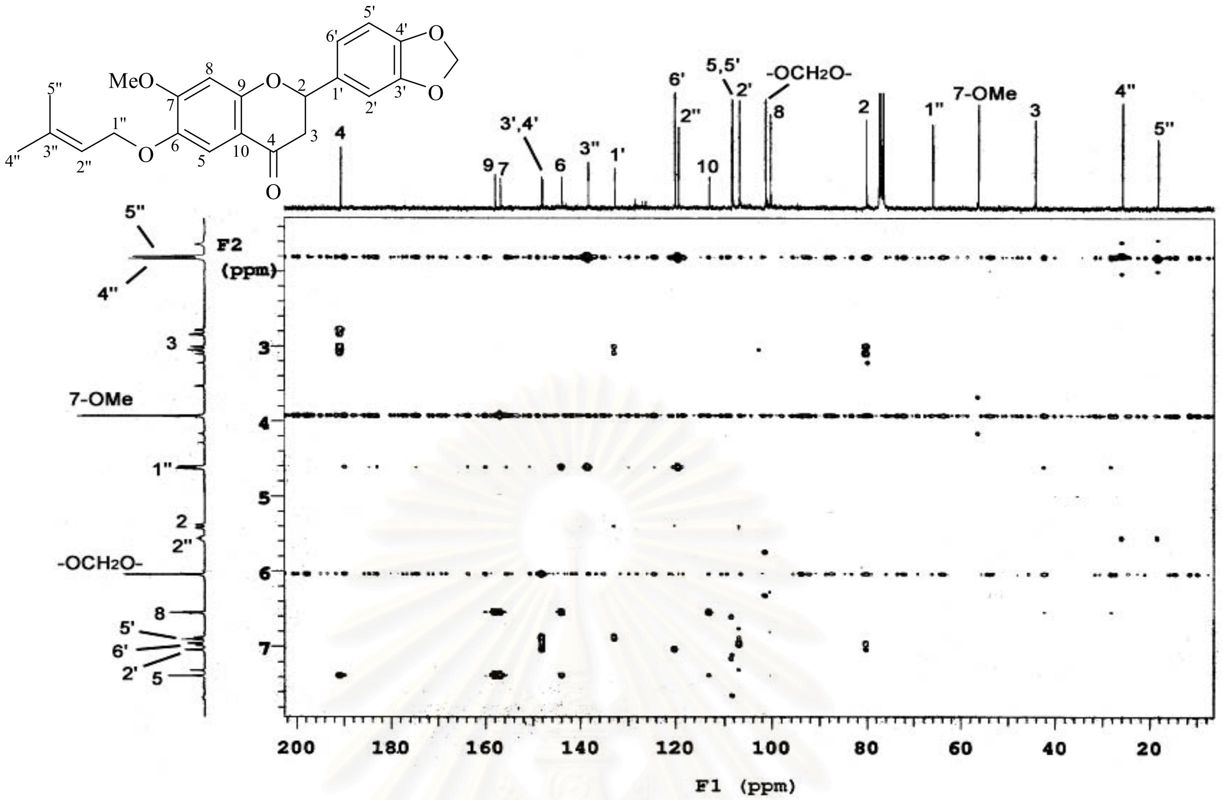


Figure 260 HMBC Spectrum of compound ME26 (CDCl$_3$)

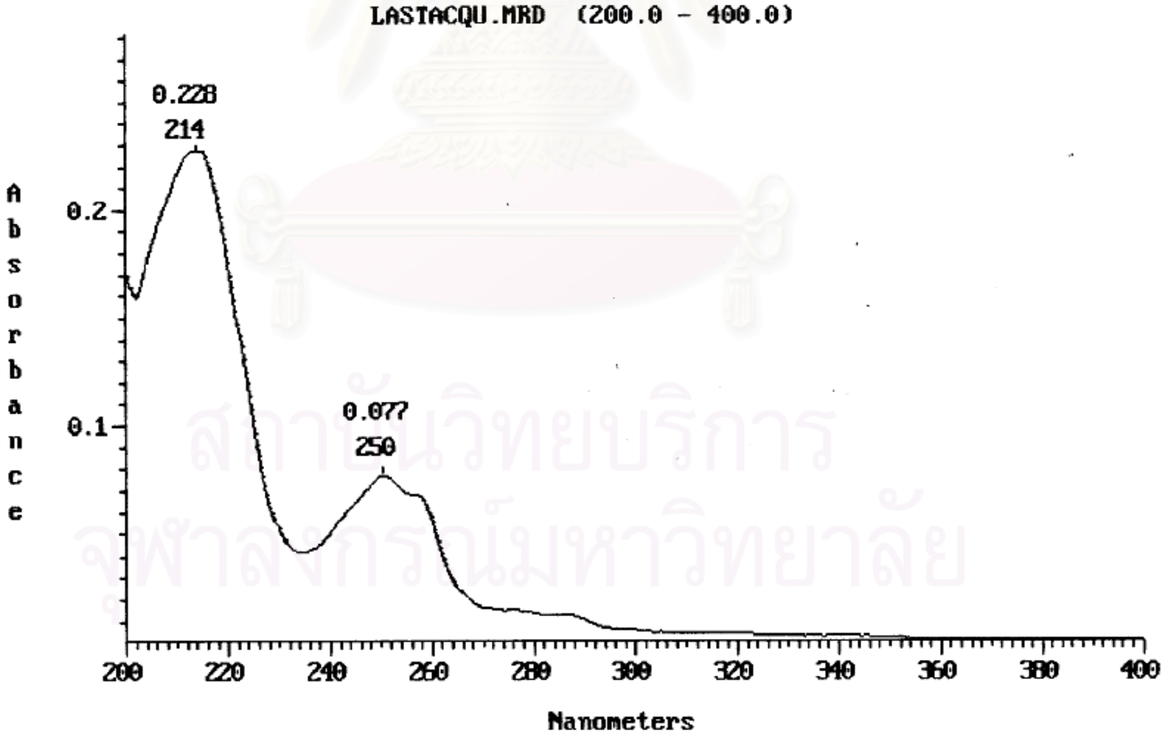


Figure 261 UV Spectrum of compound ME27 (methanol)

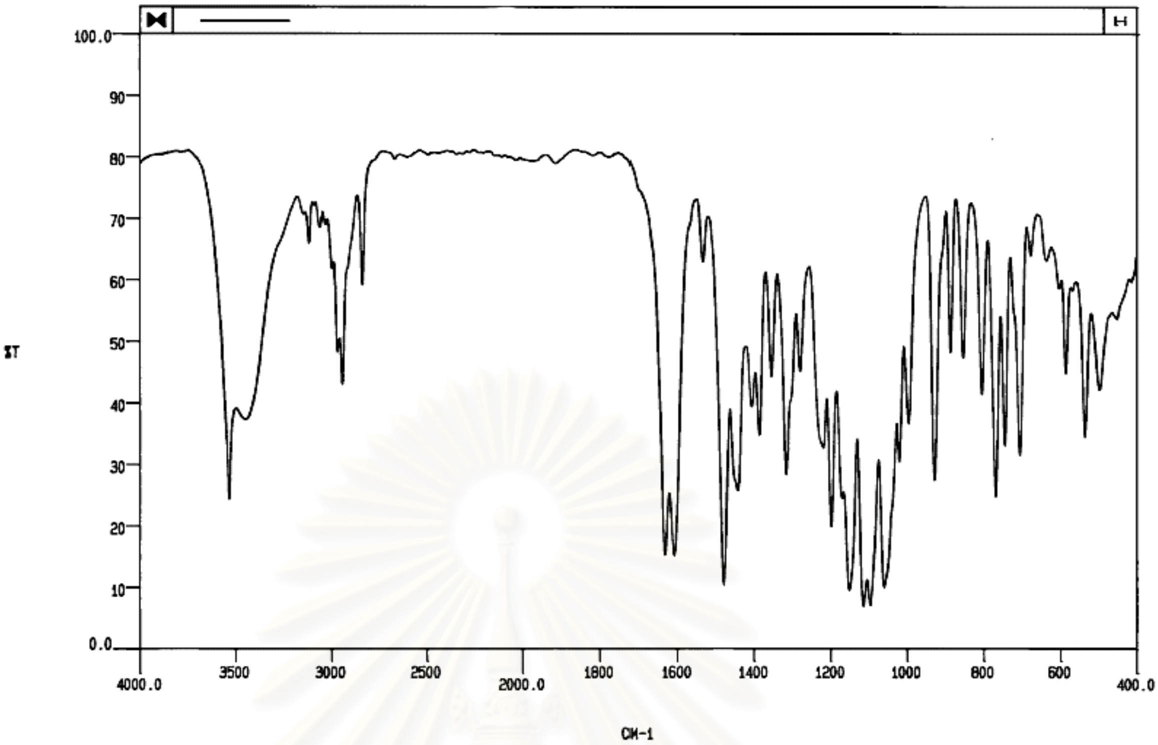
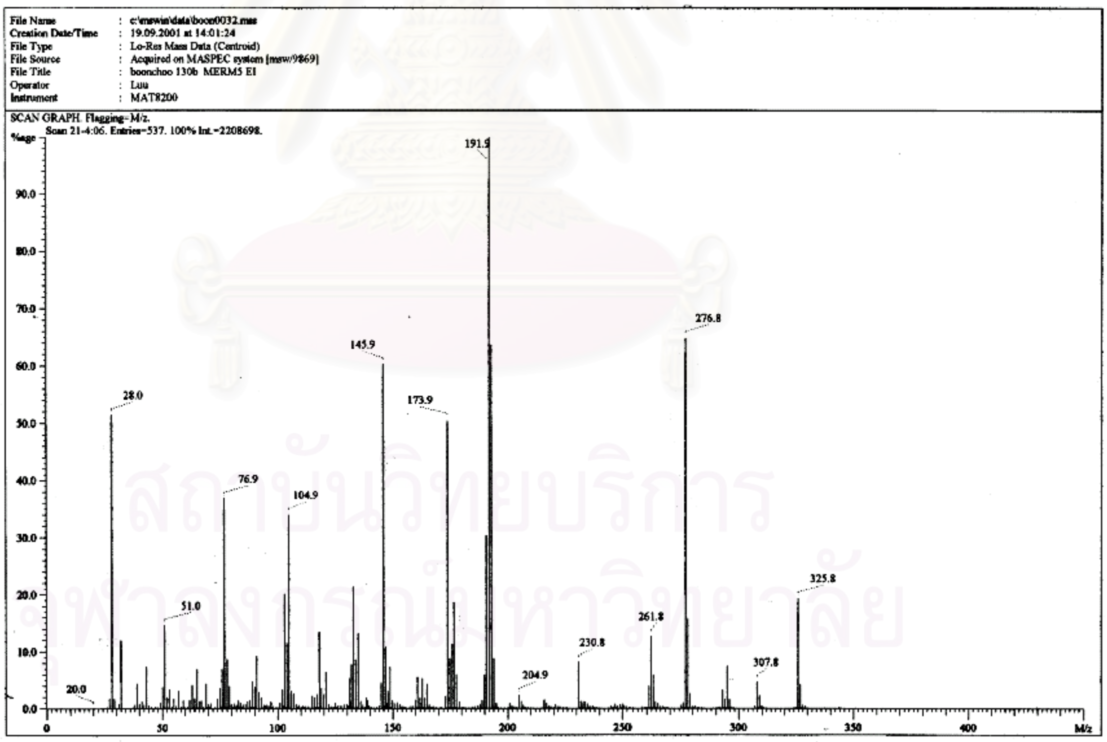


Figure 262 IR Spectrum of compound ME27 (KBr disc)



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Instrument : MAT8200

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Figure 263 EI Mass spectrum of compound ME27

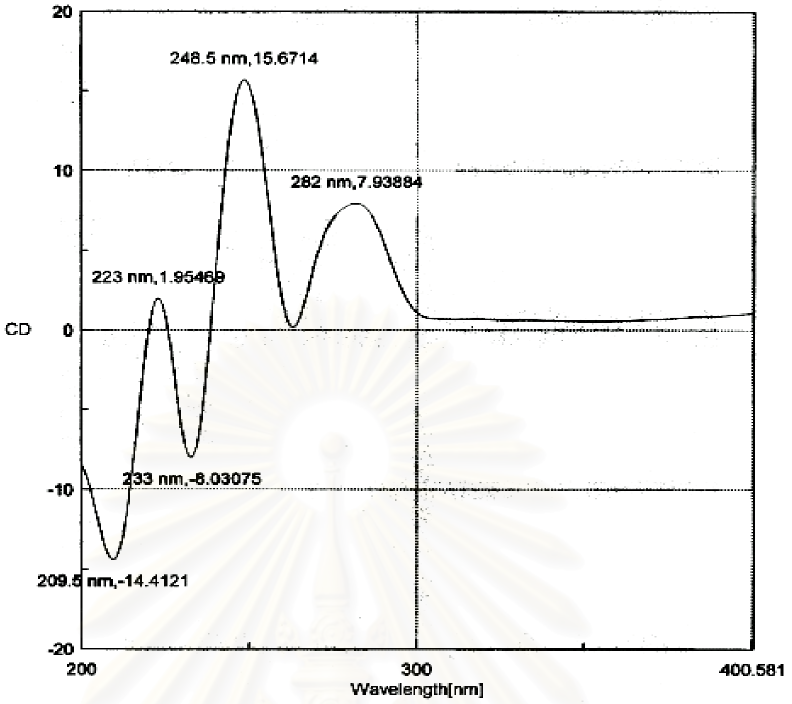


Figure 264 CD Spectrum of compound ME27 (methanol)

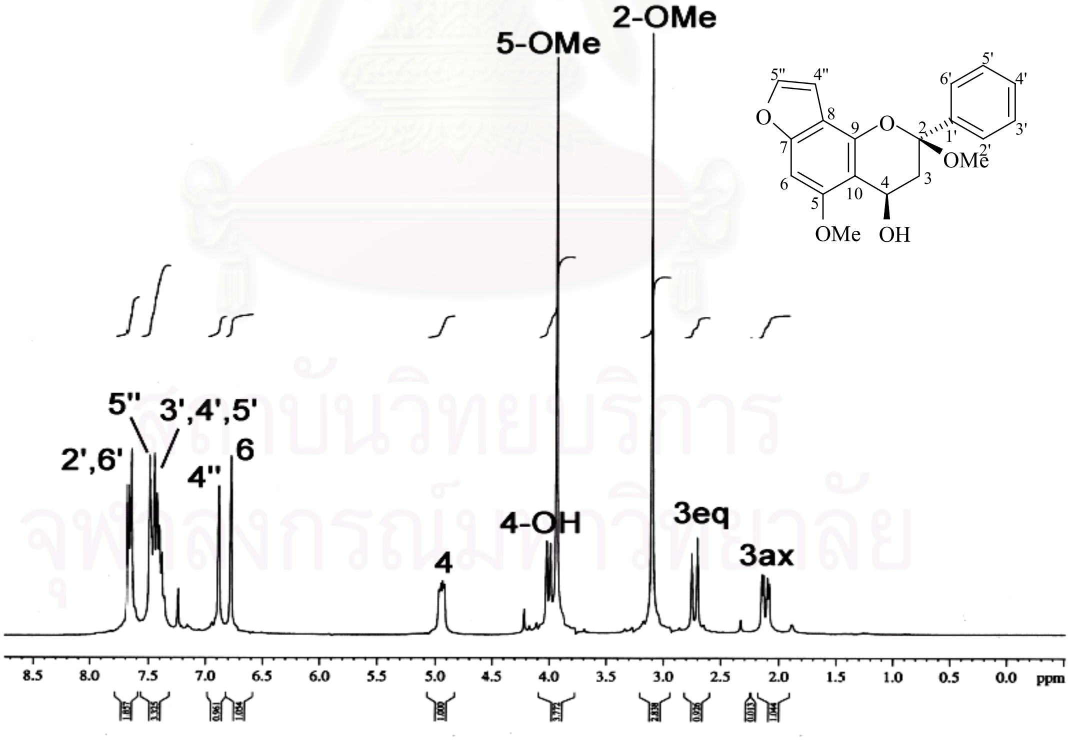


Figure 265 ^{1}H NMR (300 MHz) Spectrum of compound ME27 ($CDCl_3$)

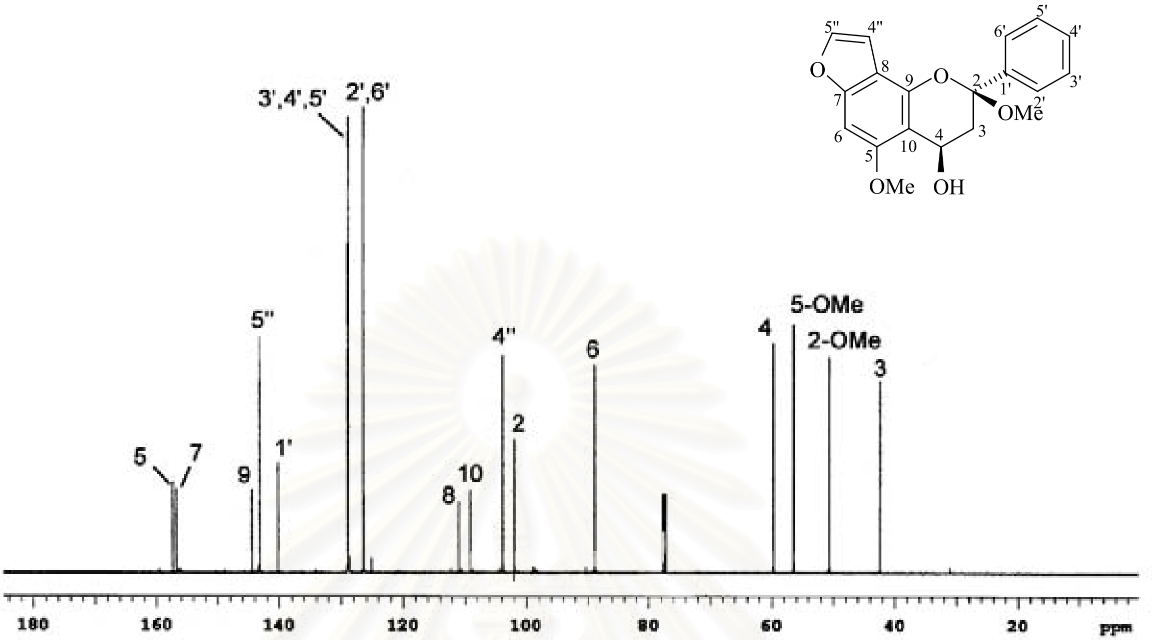


Figure 266 ^{13}C NMR (75 MHz) Spectrum of compound ME27 (CDCl$_3$)

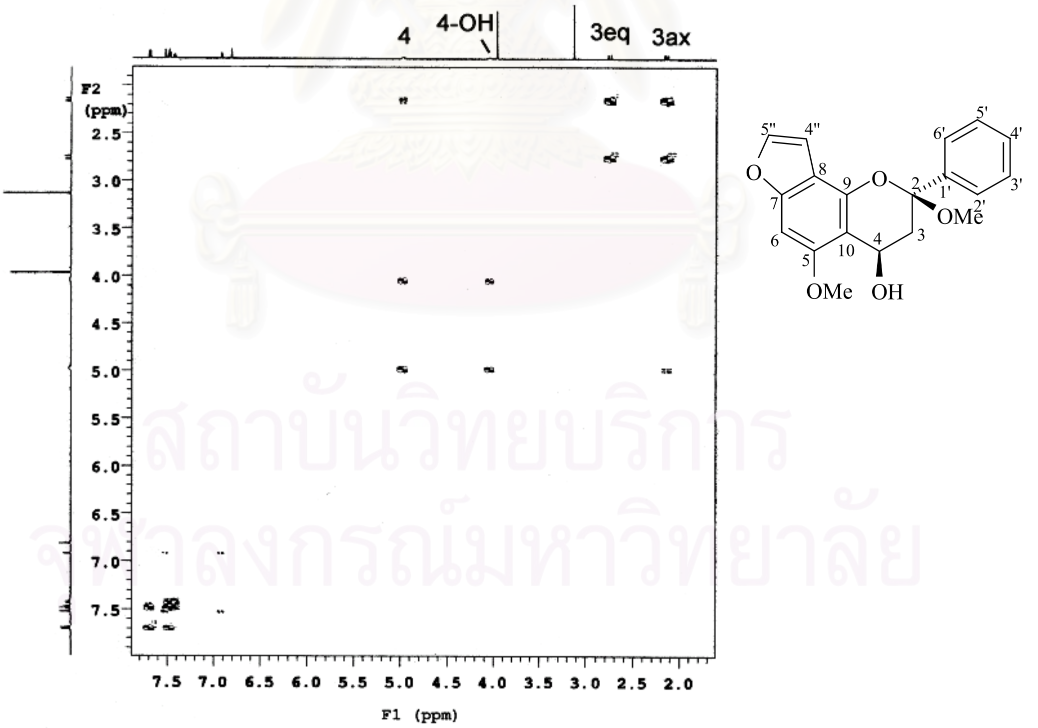


Figure 267 ^{1}H-^{1}H COSY Spectrum of compound ME27 (CDCl$_3$)

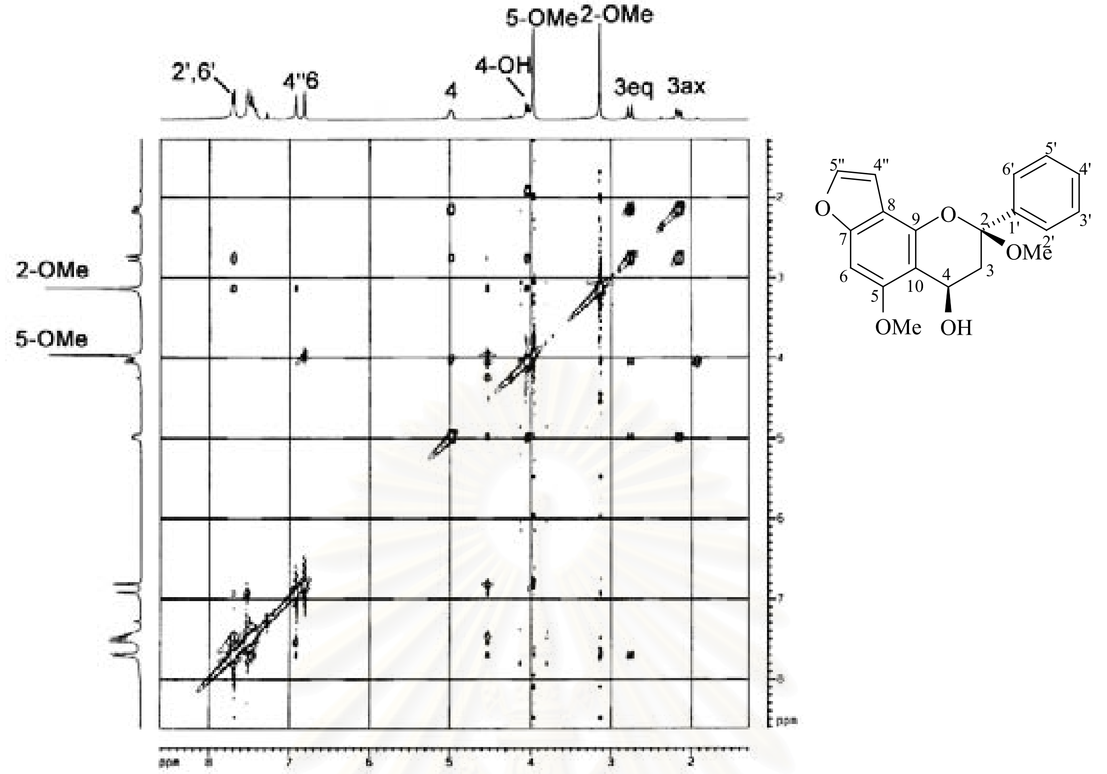


Figure 268 NOESY Spectrum of compound ME27 ($CDCl_3$)

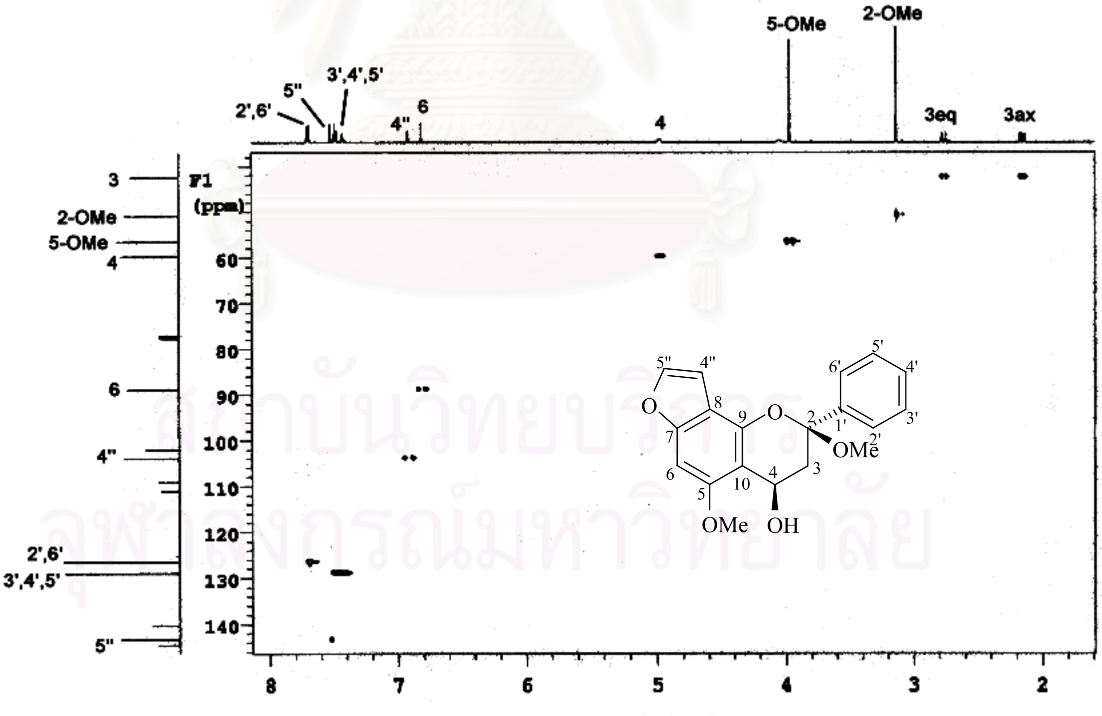


Figure 269 HSQC Spectrum of compound ME27 ($CDCl_3$)

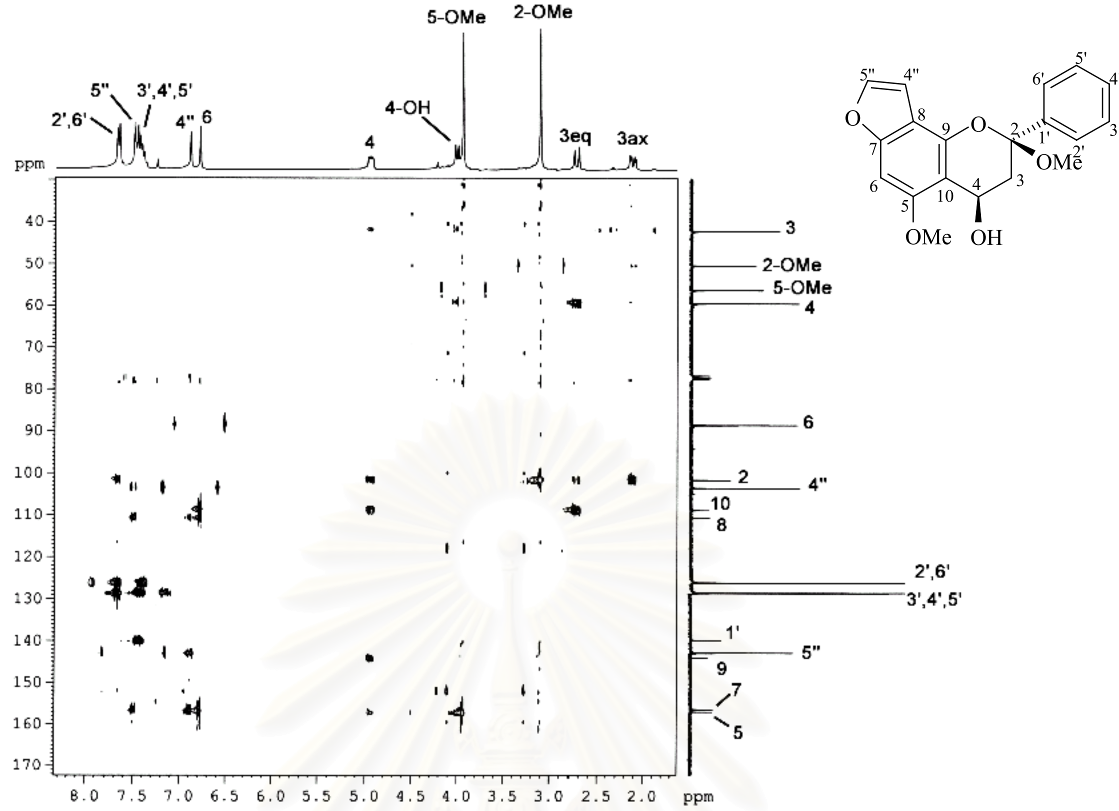


Figure 270 HMBC Spectrum of compound ME27 ($CDCl_3$)

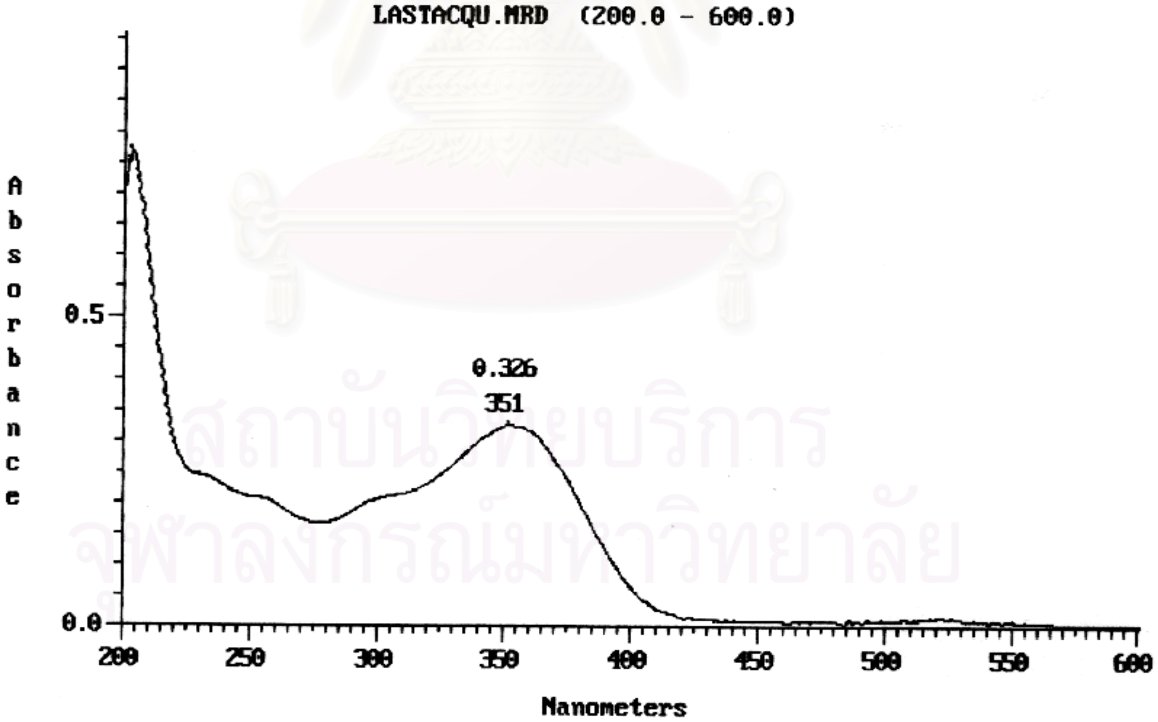


Figure 271 UV Spectrum of compound ME28 (methanol)

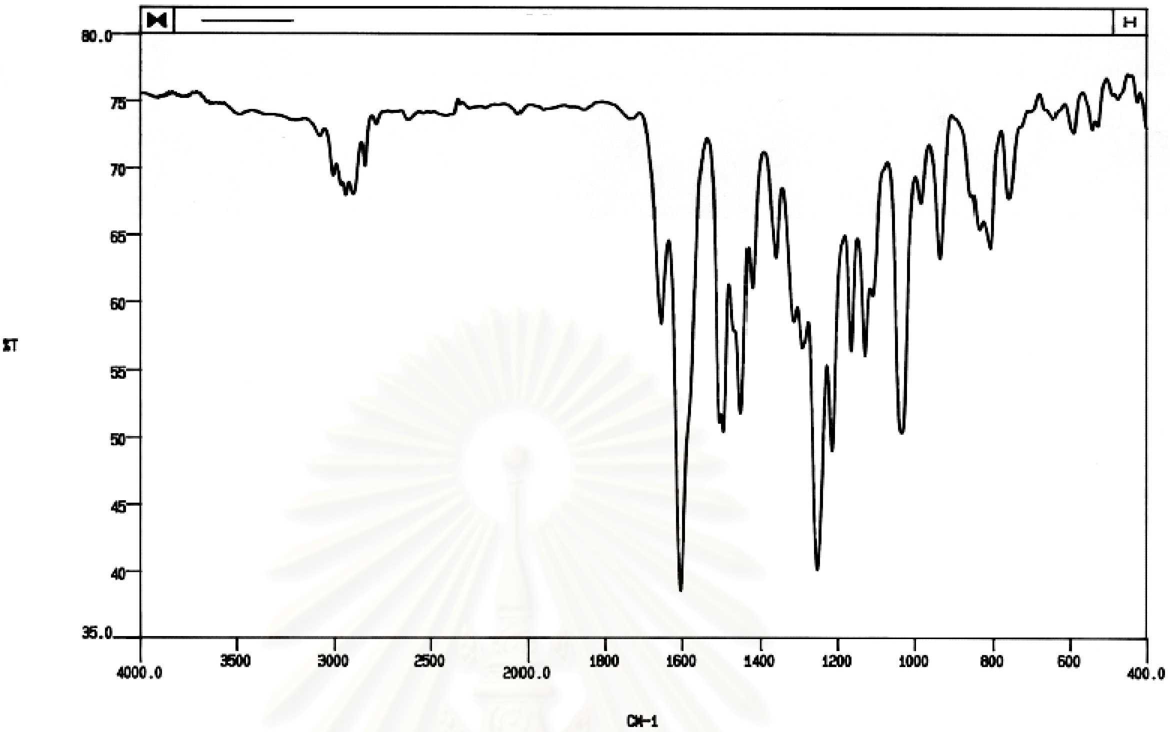
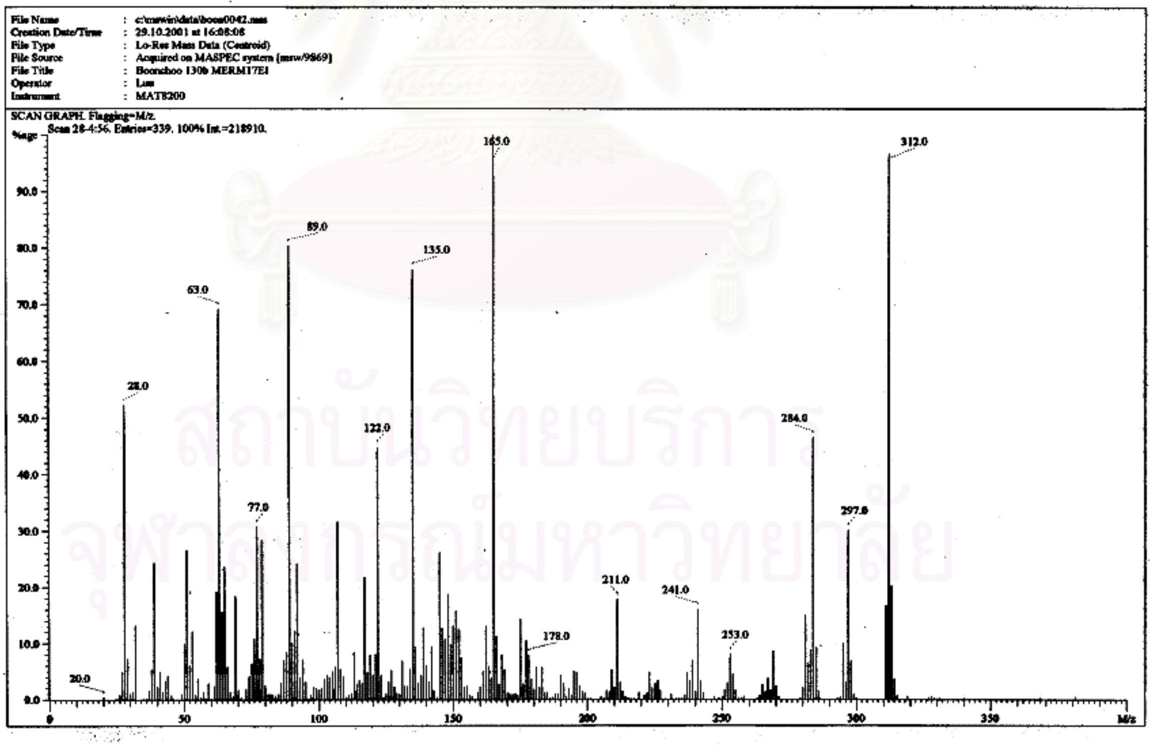


Figure 272 IR Spectrum of compound ME28 (KBr disc)



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Operator : Lam
Instrument : MAT8200

Figure 273 EI Mass spectrum of compound ME28

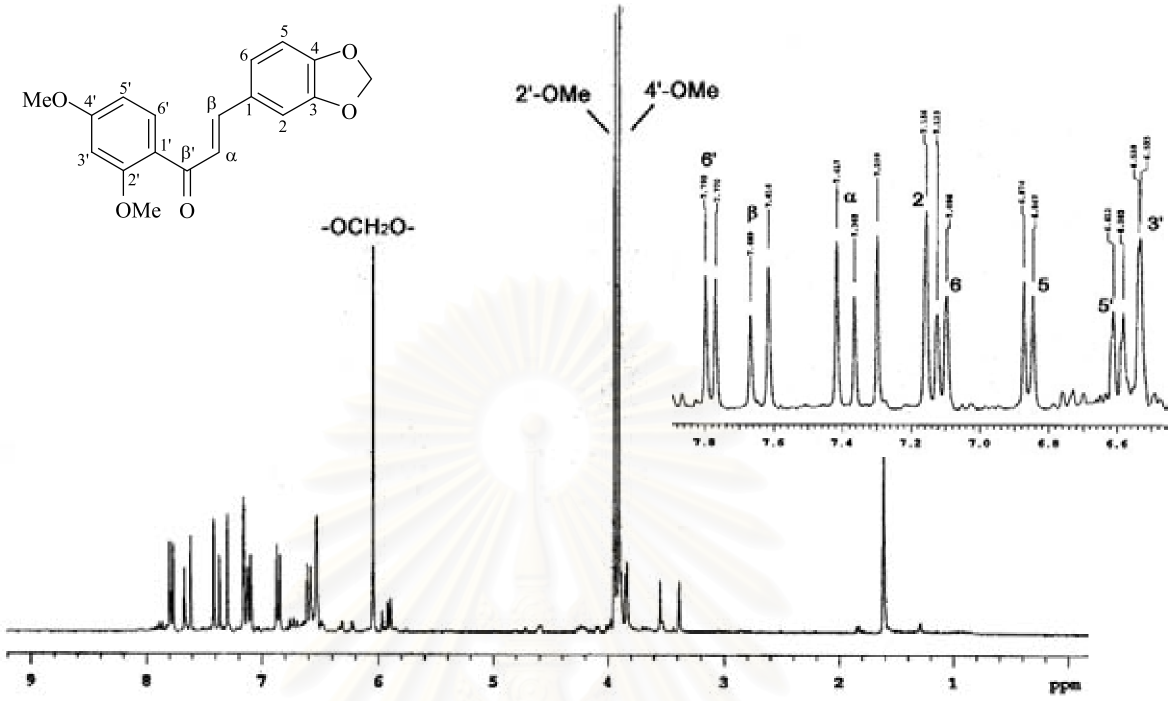

Figure 274 ^{1}H NMR (300 MHz) Spectrum of compound ME28 (CDCl$_3$)

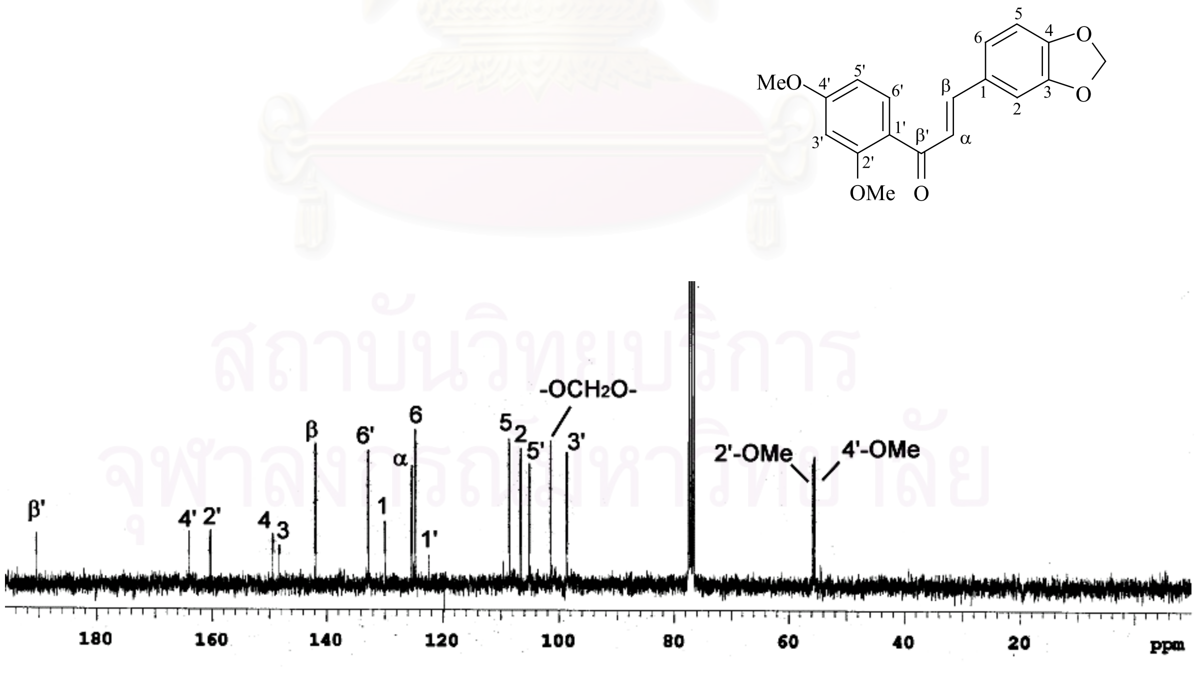

Figure 275 ^{13}C NMR (75 MHz) Spectrum of compound ME28 (CDCl$_3$)

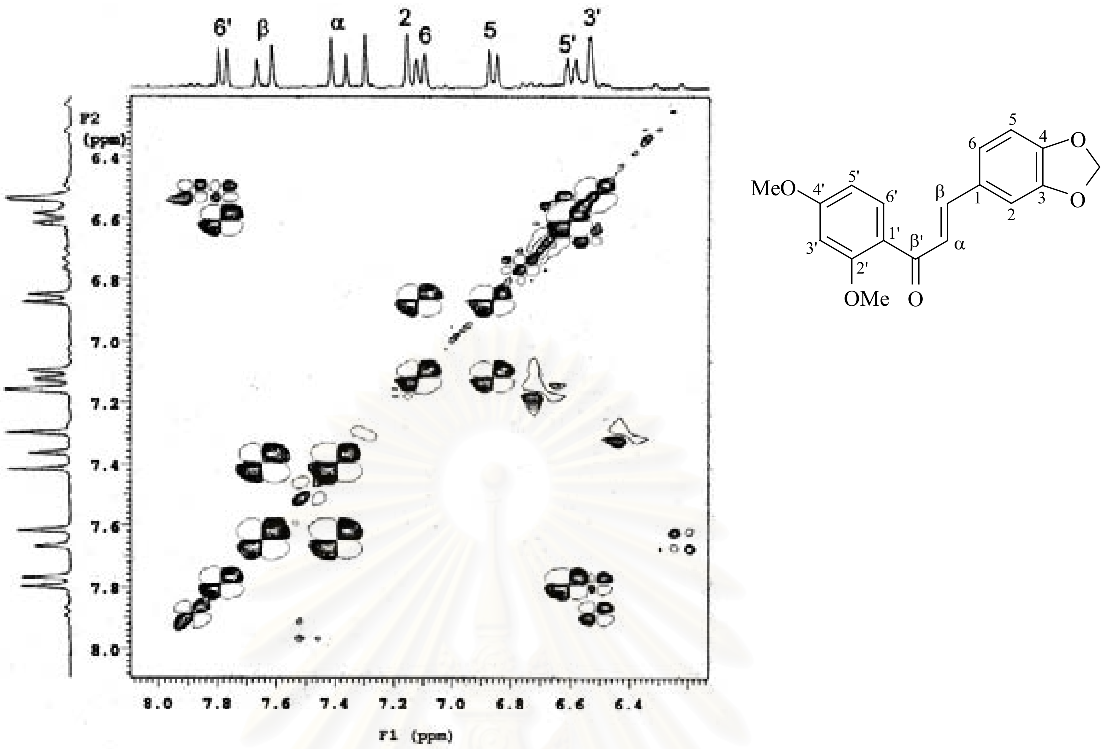

Figure 276 ^{1}H-^{1}H COSY Spectrum of compound ME28 ($CDCl_3$)

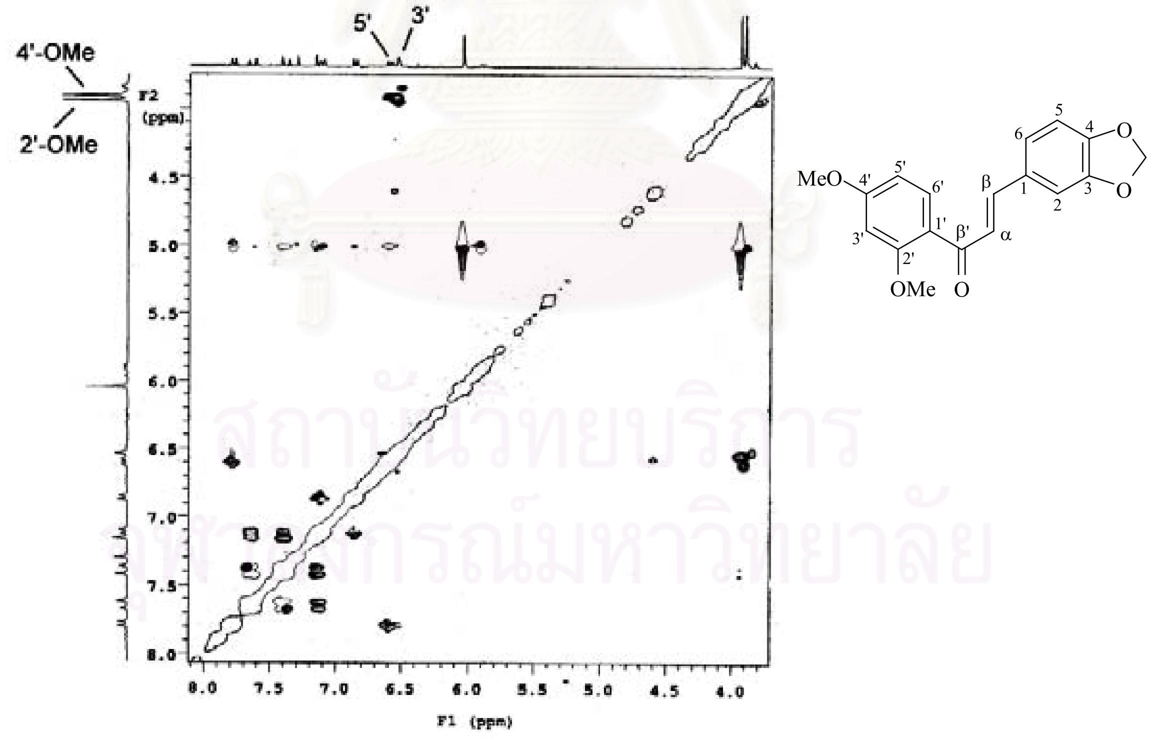

Figure 277 NOESY Spectrum of compound ME28 ($CDCl_3$)

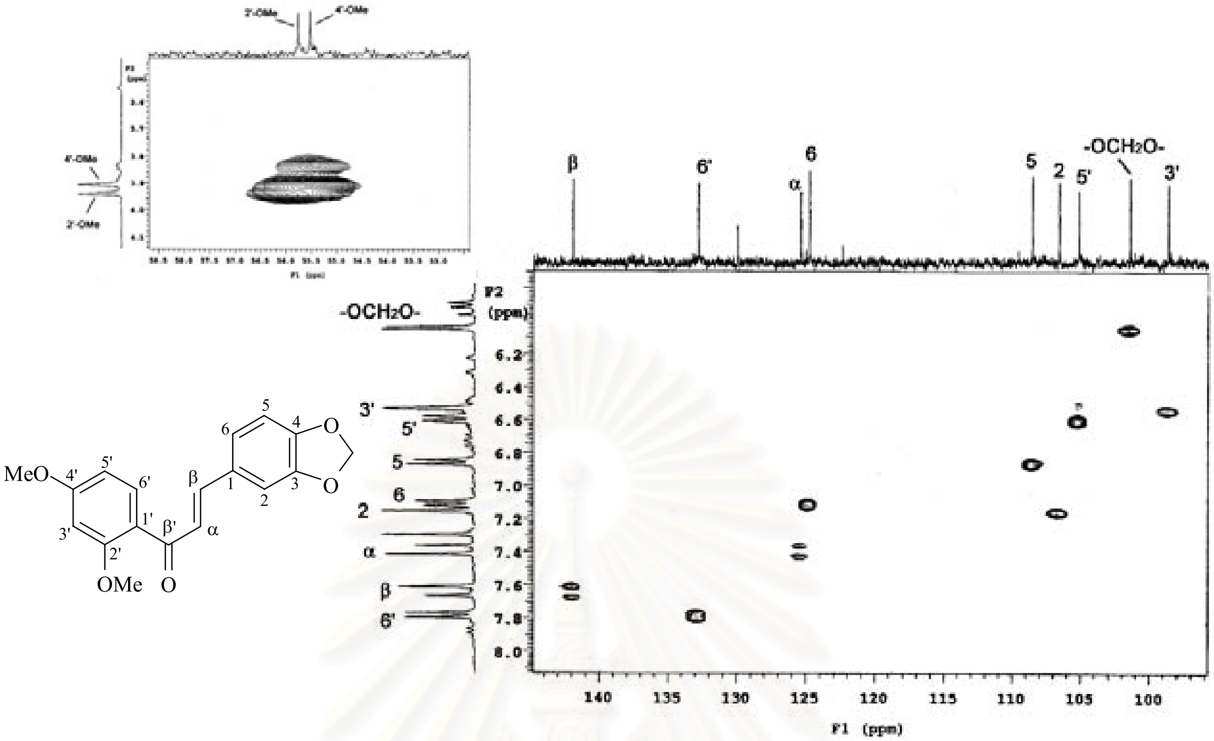

Figure 278 HSQC Spectrum of compound ME28 ($CDCl_3$)

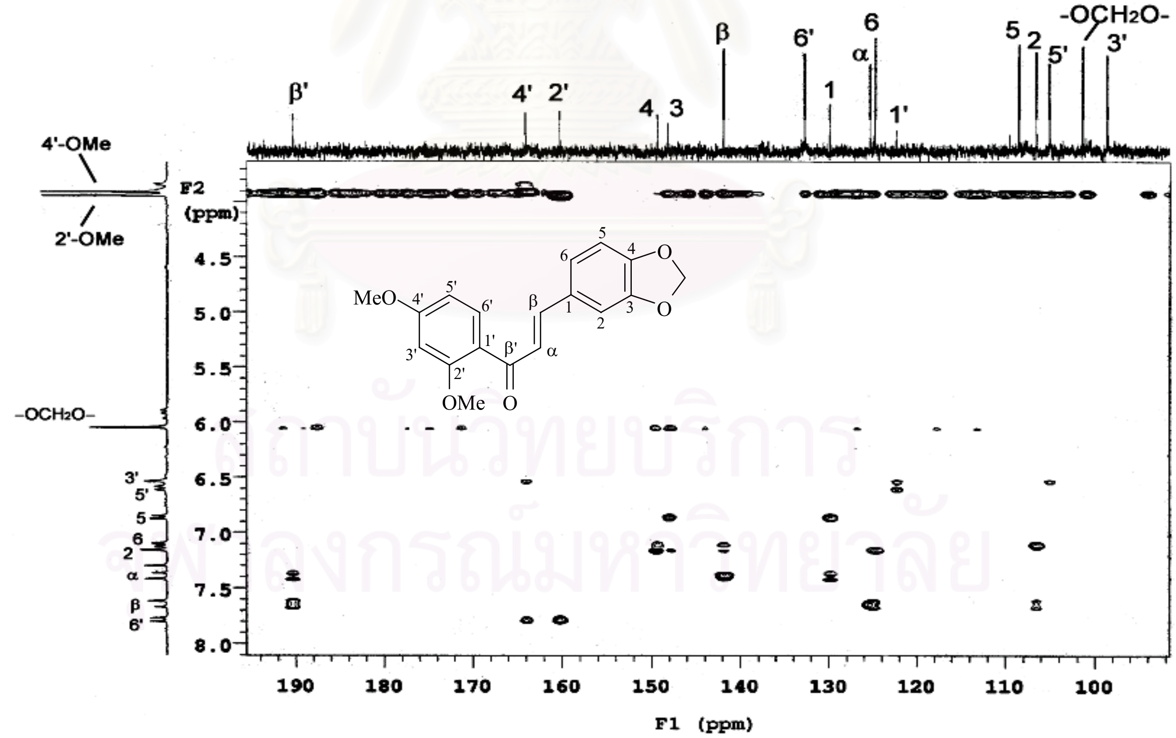

Figure 279 HMBC Spectrum of compound ME28 ($CDCl_3$)

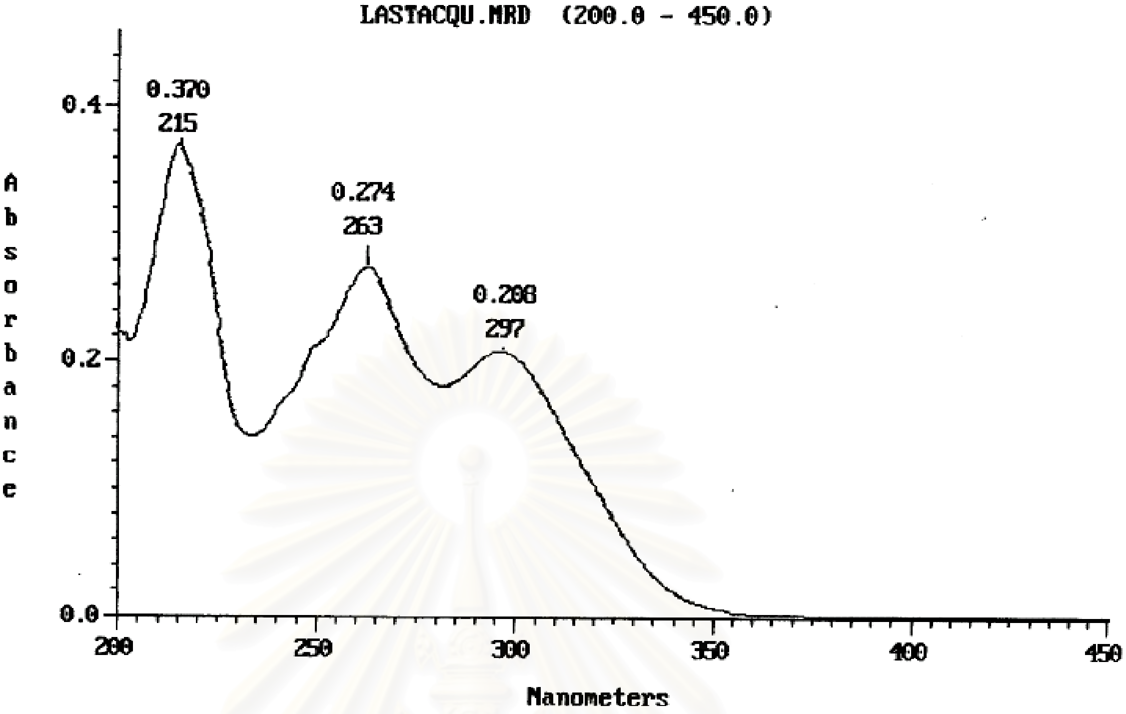

Figure 280 UV Spectrum of compound ME29 (methanol)

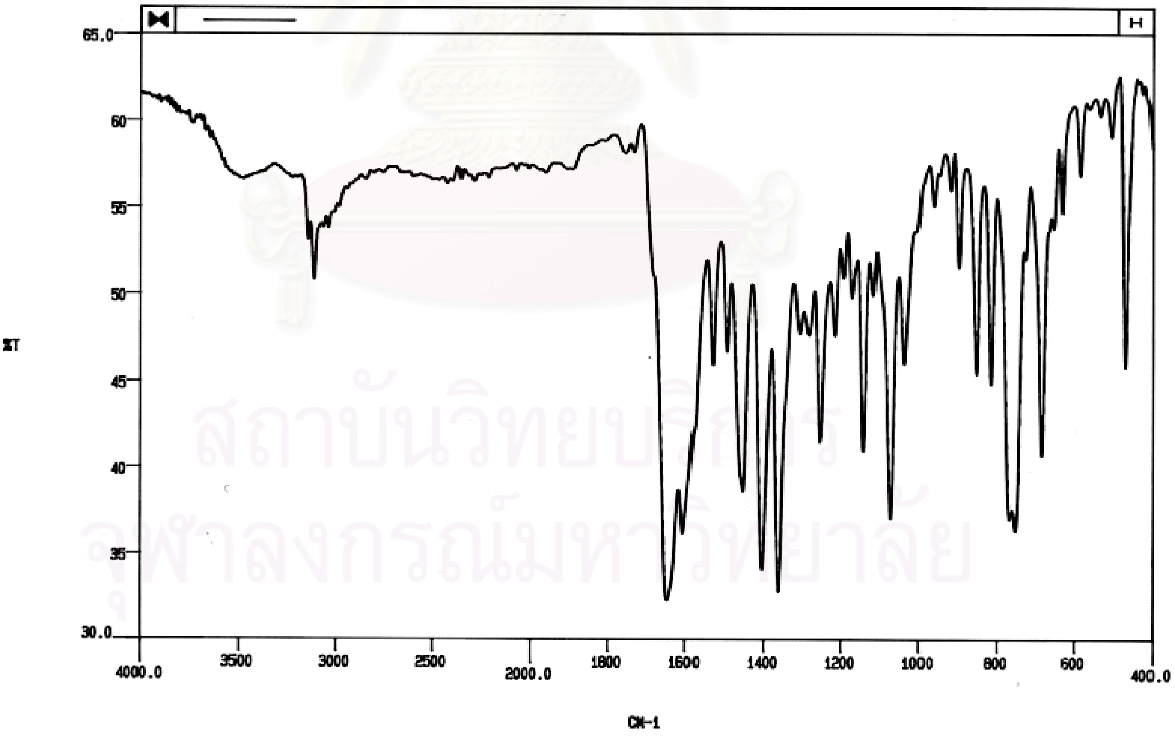

Figure 281 IR Spectrum of compound ME29 (KBr disc)

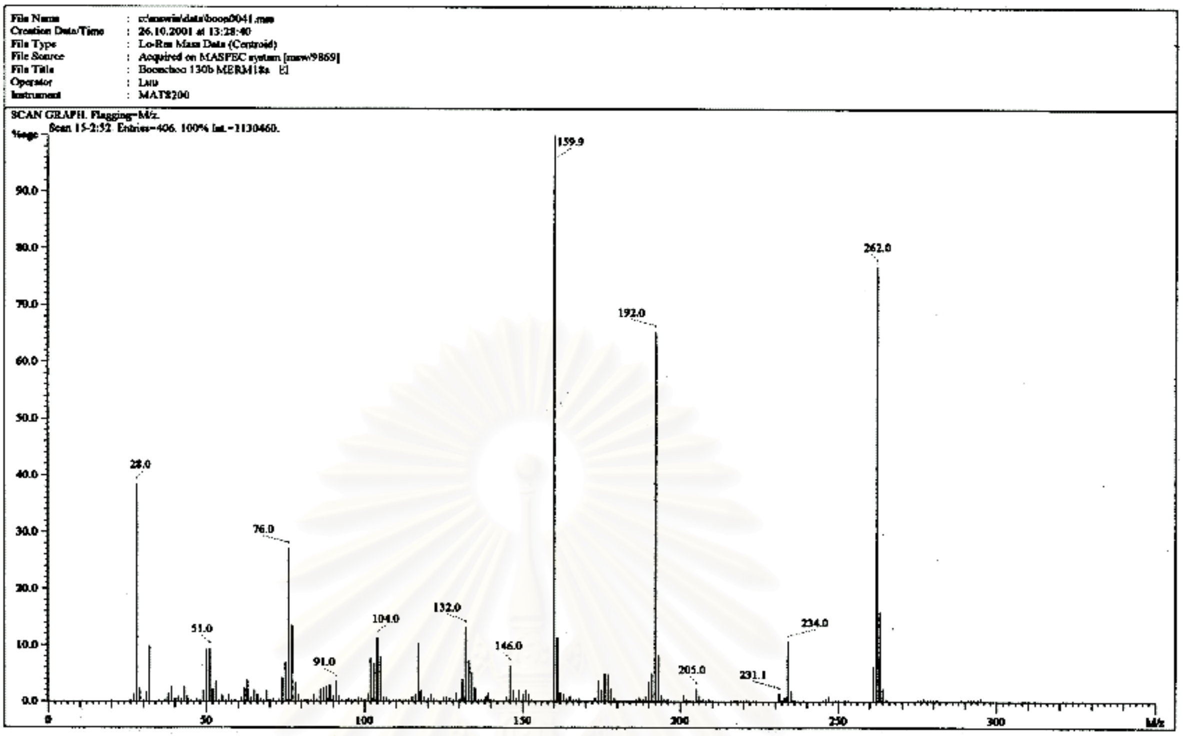

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Figure 282 EI Mass spectrum of compound ME29

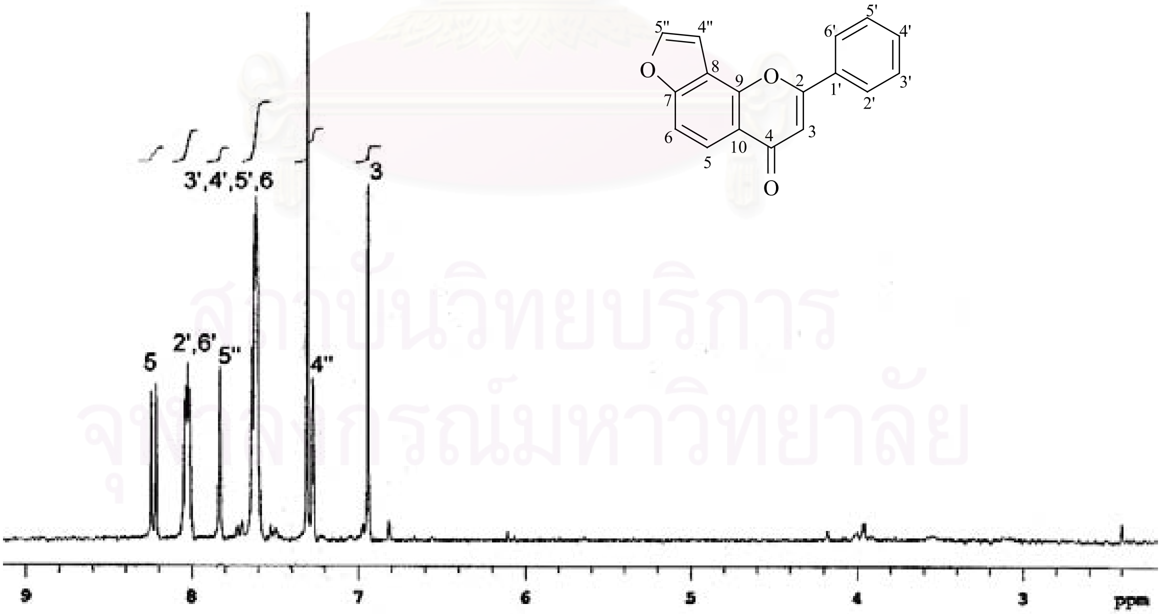

Figure 283 ^{1}H NMR (300 MHz) Spectrum of compound ME29 ($CDCl_3$)

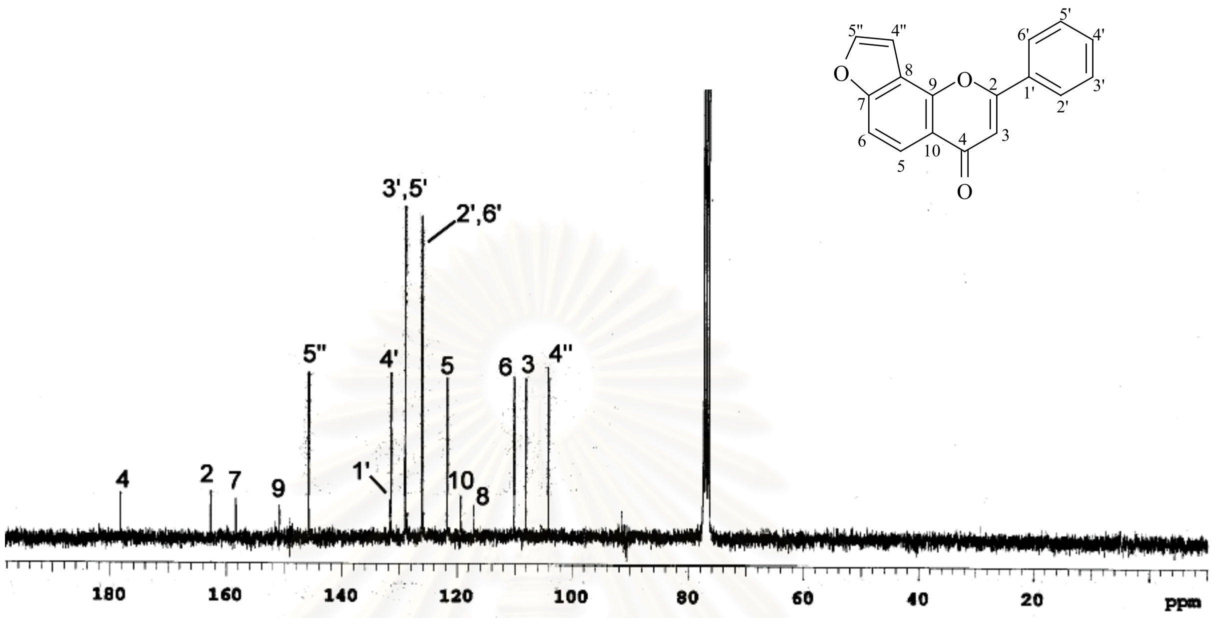

Figure 284 ^{13}C NMR (75 MHz) Spectrum of compound ME29 ($CDCl_3$)

VITA

Mr. Boonchoo Sritularak was born on January 5, 1973 in Phuket, Thailand. He received his Bachelor's degree of Science in Pharmacy in 1996 from the Faculty of Pharmaceutical of Sciences, Prince of Songkla University and Master's degree of Science in Pharmacy in 1998 from the Faculty of Pharmaceutical of Sciences, Chulalongkorn University, Thailand. He was awarded a 1999 Royal Golden Jubilee Scholarship from the Thailand Research Fund and a 2001 research grant from the German Academic Exchange Service (DAAD).

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1. Sritularak, B., De-Eknamkul, W., and Likhitwitayawuid, K. 1998. Tyrosinase inhibitors from *Artocarpus lakoocha*. Thai J. Pharm. Sci. 22: 149-155.
2. Likhitwitayawuid, K., Sritularak, B., and De-Eknamkul, W. 2000. Tyrosinase inhibitors from *Artocarpus gomezianus*. Planta Med. 66: 275-277.
3. Likhitwitayawuid, K., and Sritularak, B. 2001. A new dimeric stilbene with tyrosinase inhibitory activity from *Artocarpus gomezianus*. J. Nat. Prod. 64: 1457-1459.
4. Sritularak, B., Likhitwitayawuid, K., Conrad, J., Vogler, B., Reeb, S., Klaiber, I., and Kraus, W. 2002. New flavones from *Millettia erythrocalyx*. J. Nat. Prod. 65: 589-591.
5. Sritularak, B., Likhitwitayawuid, K., Conrad, J., and Kraus, W. 2002. Flavonoids from the roots of *Millettia erythrocalyx*. Phytochemistry 61: 943-947.

Poster Presentations

1. Sritularak, B., De-Eknamkul, W., and Likhitwitayawuid, K. Tyrosinase inhibitors from *Artocarpus lakoocha* Roxb. pp. 462-463. 24th Congress on Science and Technology of Thailand, October 19-21, 1998, Queen Sirikit National Convention Center, Bangkok.
2. Sritularak, B., and Likhitwitayawuid, K. A new dimeric stilbene from the roots of *Artocarpus gomezianus*. p. 54. The 16th Annual Research Meeting in Pharmaceutical Sciences, December 8, 1999, Faculty of Pharmaceutical Sciences, Chulalongkorn University, Bangkok.
3. Sritularak, B., Likhitwitayawuid, K., and Kraus, W. New dimeric stilbenes from *Artocarpus gomezianus*. p.127. NRCT-JSPS Core University System on Pharmaceutical Sciences The Fifth Joint Seminar Natural Medicines, November 15-17, 2000, Faculty of Pharmaceutical Sciences, Chulalongkorn University, Bangkok.
4. Sritularak, B., Likhitwitayawuid, K., Conrad, J., Vogler, B., Reeb, S., Klaiber, I., and Kraus, W. New flavones from stem bark of *Millettia hemsleyana*. p. P046. Lead Compound from Higher Plants, September 12-14, 2001, University of Lausanne, Switzerland.
5. Sritularak, B., Likhitwitayawuid, K., Conrad, J., and Kraus, W. 2002. New Flavonoids from the roots of *Millettia erythrocalyx*. p. 23 28th Congress on Science and Technology of Thailand, October 24-26, 2002, Queen Sirikit National Convention Center, Bangkok.
6. Sritularak, B., Likhitwitayawuid, K., Conrad, J., and Kraus, W. 2002. New Flavonoids from the roots of *Millettia erythrocalyx*. p. 8 The 19th Annual Research Meeting in Pharmaceutical Sciences, December 4, 2002, Faculty of Pharmaceutical Sciences, Chulalongkorn University, Bangkok.

Oral Presentation

1. Sritularak, B., Likhitwitayawuid, K., Conrad, J., Vogler, B., Reeb, S., Klaiber, I., and Kraus, W. Bioactive phenolics from *Artocarpus gomezianus* and *Millettia erythrocalyx*. p. 109. RGJ-Ph.D. Congress III, April 25-27, 2002, Chomtien Beach Resort Hotel, Chonburi.