

ผลของรังสีแกมมาต่อสมบัติของวัสดุทดแทนไม้จากพอลิพรอพิลีนคอมพอสิต



นางสาวสุจิตรา วงษ์ทรงยศ

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรมหาบัณฑิต

สาขาวิชาวิศวกรรมเคมี ภาควิชาวิศวกรรมเคมี

คณะวิศวกรรมศาสตร์ จุฬาลงกรณ์มหาวิทยาลัย

ปีการศึกษา 2549

ลิขสิทธิ์ของจุฬาลงกรณ์มหาวิทยาลัย

EFFECTS OF GAMMA IRRADIATION ON THE PROPERTIES OF WOOD COMPOSITE  
FROM POLYPROPYLENE

Miss Sujitra Wongsongyot

A Thesis Submitted in Partial Fulfillment of the Requirements  
for the Degree of Master of Engineering Program in Chemical Engineering

Department of Chemical Engineering

Faculty of Engineering

Chulalongkorn University

Academic Year 2006

Copyright of Chulalongkorn University

**491686**

Thesis Title            EFFECTS OF GAMMA IRRADIATION ON THE PROPERTIES OF WOOD  
                                 COMPOSITE FROM POLYPROYLENE

By                            Miss Sujittra Wongsongyot

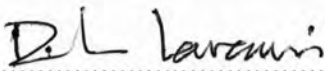
Field of Study            Chemical Engineering

Thesis Advisor           Assistant Professor Sarawut Rimdusit, Ph.D.


Thesis Co-advisor      Phiriyatorn Suwanmala, Ph.D.


---

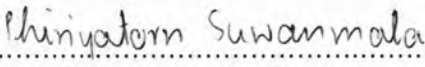
Accepted by the Faculty of Engineering, Chulalongkorn University in Partial  
Fulfillment of the Requirements for the Master's Degree

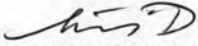
..... Dean of the Faculty of Engineering  
(Professor Direk Lavansiri, Ph.D.)

THESIS COMMITTEE

..... Chairman  
(Associate Professor Chirakarn Muangnapoh, Dr. Ing.)

..... Thesis Advisor  
(Assistant Professor Sarawut Rimdusit, Ph.D.)

..... Thesis Co-advisor  
(Phiriyatorn Suwanmala, Ph.D.)

..... Member  
(Associate Professor Siriporn Damrongsakkul, Ph.D.)

..... Member  
(Associate Professor Satok Chaikunchuensakun, Ph.D.)

สุจิตรา วงษ์ทรงยศ : ผลของรังสีแกมมาต่อสมบัติของวัสดุทดแทนไม้จากพอลิพรอพิลีนคอมพอสิต (EFFECTS OF GAMMA IRRADIATION ON THE PROPERTIES OF WOOD COMPOSITE FROM POLYPROPYLENE) อ. ที่ปรึกษา: ผู้ช่วยศาสตราจารย์ ดร. ศราวุธ ริมดุสิต, อ. ที่ปรึกษาร่วม: ดร. พิริยธร สุวรรณมาลา, 101 หน้า.

การพัฒนาวัสดุประกอบแต่งสำหรับทดแทนไม้จากพอลิพรอพิลีนซึ่งทำหน้าที่เป็นเมตริกซ์และใช้ผงไม้ยางพาราเป็นสารเติมได้รับความสนใจมากขึ้น เนื่องจากสามารถผลิตได้ง่าย ใช้ต้นทุนต่ำและมีคุณสมบัติทางกลที่ดี อย่างไรก็ตาม ข้อเสียของวัสดุประกอบแต่งชนิดนี้คือคุณสมบัติด้านการต้านทานการเสียดสีและความสามารถในการเข้ากันระหว่างผงไม้ยางพาราและพอลิพรอพิลีนเมตริกซ์ งานวิจัยนี้ใช้รังสีแกมมาสำหรับปรับปรุงคุณสมบัติการคืบของวัสดุทดแทนไม้จากพอลิพรอพิลีน และศึกษาผลของรังสีแกมมาต่อคุณสมบัติของวัสดุทดแทนไม้จากพอลิพรอพิลีนทั้งที่เติมและไม่เติมสารช่วยผสม (PP-g-MA) ชิ้นงานทั้งหมดถูกฉายด้วยรังสีแกมมาที่ระดับรังสี 5, 10, 20 และ 30 กิโลเกรย์ ทั้งในสภาวะบรรยากาศและไนโตรเจน ผลการทดลองพบว่า มีความเป็นไปได้ในการใช้รังสีแกมมาที่ระดับ 10 กิโลเกรย์สำหรับปรับปรุงคุณสมบัติของพอลิพรอพิลีนและวัสดุทดแทนไม้จากพอลิพรอพิลีน ซึ่งการฉายรังสีที่ระดับ 10 กิโลเกรย์จะทำให้พอลิพรอพิลีน ผงไม้ และวัสดุคอมพอสิตมีความอดุลัสการดัดโค้ง (Flexural modulus) เพิ่มขึ้นประมาณ 23% อีกทั้งพอลิพรอพิลีนที่มีการฉายรังสีจะช่วยให้มีความแข็งแรง (Flexural strength) เพิ่มขึ้น วัสดุคอมพอสิตสามารถต้านทานการคืบได้มากที่สุดที่ระดับรังสี 10 กิโลเกรย์ เพราะรังสีระดับนี้จะทำให้เกิดพันธะเคมีหรือเกิดการเชื่อมโยงระหว่างโมเลกุลซึ่งสามารถยืนยันโดยการทดลองการสกัดด้วยตัวทำละลาย (Solvent extraction) นอกจากนี้ ผลของการใช้รังสีควบคู่กับการใช้สารช่วยผสมจะช่วยต้านทานการคืบ (Creep resistant) ของวัสดุคอมพอสิตได้มากขึ้น การฉายรังสีมีผลเพียงเล็กน้อยต่อคุณสมบัติทางความร้อนของพอลิพรอพิลีนและวัสดุคอมพอสิต เมื่อฉายรังสีในระดับที่สูงขึ้นจะช่วยเพิ่มคุณสมบัติด้านการยึดเกาะระหว่างผิวหน้าของพอลิพรอพิลีนเมตริกซ์กับผงไม้ซึ่งสังเกตจากภาพถ่าย scanning electron microscope (SEM)

ภาควิชา....วิศวกรรมเคมี.....  
สาขาวิชา....วิศวกรรมเคมี.....  
ปีการศึกษา....2549.....

ลายมือชื่อนิสิต.....  
ลายมือชื่ออาจารย์ที่ปรึกษา.....  
ลายมือชื่ออาจารย์ที่ปรึกษาร่วม.....

## 4870527821 : MAJOR CHEMICAL ENGINEERING

KEY WORD: COMPOSITES / POLYPROPYLENE / WOODFLOUR / GAMMA RADIATION / CROSSLINKING/ CREEP BEHAVIOR

SUJITTRA WONGSONGYOT: EFFECTS OF GAMMA IRRADIATION ON THE PROPERTIES OF WOOD COMPOSITE FROM POLYPROPYLENE.  
 THESIS ADVISOR: ASST. PROF. SARAWUT RIMDUSIT, Ph.D., THESIS COADVISOR: PHIRIYATORN SUWANMALA, Ph.D., 101 pp.

Polypropylene wood composites as wood-substituted materials have received increasing attention due to their easy processability, low cost as well as reasonable mechanical characteristics. However, the main problems encountered in PP/wood composites are poor creep resistant and low compatibility between woodflour and the PP matrix. In this work, gamma radiation was used to improve creep performance of the PP wood composites. Effects of gamma irradiation on properties of PP/wood composites with and without the presence of PP-g-MA compatibilizer are studied. The specimens are irradiated with gamma radiation at various doses of 5, 10, 20 and 30 kGy in both air and nitrogen atmospheres. The results demonstrated that it was possible to use low radiation doses 10 kGy to safely modify PP and its wood composites. The wood composite specimens radiated with 10 kGy provided about of less than 23% increase in flexural modulus compared with the unirradiated samples. The strength of the irradiated PP was 18.8% greater than that of the unirradiated PP. Creep strain of the irradiated composite has maximum decrease in the value due to the possible formation of crosslinking as confirmed by solvent extraction experiment. In addition, the combined effect of the use of gamma radiation and PP-g-MA rendered PP wood composite with maximum creep resistance. Furthermore, irradiation was found to show marginal effects on thermal properties of the PP and the PP/wood composites. Finally with increasing radiation dose, the interfacial adhesion between the woodflour and the polypropylene was enhanced as evidenced by SEM micrographs.

Department...Chemical Engineering.....

Field of Study Chemical Engineering.....

Academic Year.....2006.....

Student's signature: *Sujittra Wongsongyot*

Advisor's signature: *Sarawut Rimdusit*

Co-advisor's signature: *Phiriyatorn Suwanmala*

## ACKNOWLEDGEMENTS

I wish to express my sincerest gratitude and deep appreciation to my advisor, Asst. Prof. Dr. Sarawut Rimdusit, and my co-advisor, Dr. Phiriyatorn Suwanmala, for their kindness, invaluable supervision, invaluable guidance, advice, and encouragement throughout the course of this study and editing of this thesis.

The Office of Atoms for Peace, Thailand, is also acknowledged for the  $^{60}\text{Co}$  gamma irradiator and TGA experiments. I wish to thank Dr. Phiriyatorn Suwanmala and Dr. Kasinee Hemvichian for her helpful and good advice on  $^{60}\text{Co}$  gamma irradiator and TGA analysis.

Gratefully thanks to Center of Excellence in Particle Technology, Chulalongkorn University, Thailand, for supporting ball mill and sieves.

Furthermore, thanks are due to HMC Polymer Company LTD., Thailand, and Dupont Company LTD., Thailand, for their raw material support.

Additionally, Many thanks to my senior colleagues especially Miss. Chanchira Jubsilp for her edit in the writing of my work and everyone in the Polymer Engineering Laboratory, Chulalongkorn University, for their discussion and friendly encouragement. Moreover, I would like to thank everyone here. I feel so fortunate having a chance to learn here.

Finally, I would like to affectionately give all gratitude to the members of my family for their wholehearted understanding, encouragement, and patient support throughout my entire study.

# CONTENTS

	<b>PAGE</b>
<b>ABSTRACT (THAI)</b> .....	iv
<b>ABSTRACT (ENGLISH)</b> .....	v
<b>ACKNOWLEDGEMENTS</b> .....	vi
<b>TABLE OF CONTENTS</b> .....	vii
<b>LIST OF TABLES</b> .....	xi
<b>LIST OF FIGURES</b> .....	xiii
<b>LIST OF SCHEMES</b> .....	xv
 <b>CHAPTER</b>	
<b>I INTRODUCTION</b> .....	1
1.1 General Introduction .....	1
1.2 Objectives of the Present Study .....	4
1.3 Scopes of the Present Study .....	4
<b>II THEORY</b> .....	5
2.1 Composites .....	5
2.1.1 Classification of Composite Materials .....	5
2.1.2 Theory of the Action of Filler and Reinforcements .....	6
2.1.3 Mechanical properties of Particulate Filled Polymer .....	7
2.1.3.1 Tensile Modulus .....	7
2.1.3.2 Tensile Strength .....	10
2.1.3.3 Aspect Ratio .....	11
2.1.3.4 Creep .....	12
2.2 Matrix .....	13
2.3 Filler (Woodflour) .....	14
2.4 Compatibilizer .....	16
2.5 Radiation .....	18



<b>CHAPTER</b>	<b>PAGE</b>
2.5.1 Radiological Units .....	18
2.5.2 Dosimetry of Electromagnetic Radiation .....	20
2.5.3 Gamma Rays .....	20
2.5.4 Radiation Effects on Polymers .....	21
2.5.5 Radiation Effects on Cellulose and Wood .....	23
2.5.6 Using of Gamma Radiation in Composites Technology .....	24
<b>III LITERATURE REVIEWS .....</b>	<b>25</b>
<b>IV EXPERIMENT .....</b>	<b>34</b>
4.1 Material .....	34
4.2 Methodology .....	34
4.2.1 Preparation of Polymer Matrix and Compatibilizer .....	34
4.2.2 Preparation of Woodflour .....	34
4.2.3 Preparation of Wood Composites .....	35
4.3 Wood Composite Characterization .....	35
4.3.1 Density .....	35
4.3.2 Mechanical Characterization .....	36
4.3.2.1 Flexural Property Measurement .....	36
4.3.2.2 Tension Property Measurement .....	36
4.3.2.3 Creep Property Measurement .....	37
4.3.3 Thermal Characterization .....	37
4.3.3.1 Differential Scanning Calorimetry (DSC) .....	37
4.3.3.2 Thermogravimetric Analysis (TGA) .....	38
4.3.4 Interfacial Interaction .....	38
4.3.5 Extraction Percentages Examination .....	38
<b>V RESULTS AND DISCUSSION .....</b>	<b>40</b>
5.1 Effect of Gamma Ray on Polypropylene (PP) Matrix. ....	40
5.1.1 Physical Characterization .....	40
5.1.2 Density of Polypropylene Matrix .....	40



CHAPTER	PAGE
5.1.3 Mechanical Characterizations.....	41
5.1.3.1 Flexural Property Measurement .....	41
5.1.3.2 Tension Property Evaluation .....	42
5.1.3.3 Creep Property Measurement .....	43
5.1.4 Thermal Characterizations.....	43
5.1.4.1 Differential Scanning Calorimetry (DSC).....	43
5.1.4.2 Thermogravimetric Analysis (TGA).....	44
5.1.5 Interfacial Interaction.....	44
5.2 Effect of Gamma Ray on <i>Hevea Brasiliensis</i> Woodflour. ....	45
5.2.1 Physical Characterization .....	45
5.2.2 Mechanical Characterizations.....	45
5.2.2.1 Flexural and Tension Property Measurement .....	45
5.2.3 Thermal Properties.....	46
5.2.3.1 Thermogravimetric Analysis (TGA).....	46
5.3 Effects of Gamma Ray on Polypropylene <i>Hevea Brasiliensis</i> Woodflour Composites.....	47
5.3.1 Physical Characterization .....	47
5.3.2 Density of Polypropylene Woodflour Composites .....	47
5.3.3 Mechanical Characterizations.....	47
5.3.3.1 Flexural Property Measurement .....	47
5.3.3.2 Tension Property Evaluation .....	48
5.3.3.3 Creep Property Measurement .....	49
5.3.4 Thermal Characterizations.....	50
5.3.4.1 Differential Scanning Calorimetry (DSC).....	50
5.3.4.2 Thermogravimetric Analysis (TGA).....	51
5.3.5 Interfacial Interaction.....	51
5.4 Effects of Gamma Ray on Polypropylene/ <i>Hevea Brasiliensis</i> Woodflour Composites with and without a Compatibilizer. ....	52
5.4.1 Physical Characterization .....	52
5.4.2 Density of Polypropylene Woodflour Composites with PP-g-MA ..	52

<b>CHAPTER</b>	<b>PAGE</b>
5.4.3 Mechanical Characterizations.....	53
5.4.3.1 Flexural and tensile Property Measurement.....	53
5.4.3.2 Creep Property Measurement.....	54
5.4.4 Extraction Percentages Examination.....	54
5.5 Effects of Radiation Dose Rates of Gamma Ray on Polypropylene/ <i>Hevea</i> <i>Brasiliensis</i> Woodflour Composites.....	55
5.5.1 Mechanical Characterizations.....	55
5.5.1.1 Flexural and Tensile Property Measurement.....	55
5.5.1.2 Creep Property Measurement.....	56
<b>VI CONCLUSIONS .....</b>	<b>87</b>
<b>REFERENCES.....</b>	<b>88</b>
<b>APPENDICES .....</b>	<b>92</b>
<b>APPENDIX A</b> Theoretical Density Calculation.....	<b>93</b>
<b>APPENDIX B</b> Mechanical Characterizations .....	<b>95</b>
<b>APPENDIX C</b> Extraction Percentages .....	<b>99</b>
<b>VITA.....</b>	<b>101</b>

## LIST OF TABLES

TABLE	PAGE
2.1 Maximum packing fraction of several fillers types.....	9
2.2 Sharp factor for several filler types.....	9
2.3 Properties of polypropylene and other polyolefin.....	13
2.4 Chemical constituents of natural wood .....	15
2.5 Properties of rubber wood .....	15
2.6 Classification of irradiated polymers with gamma ray .....	21
3.1 Summary of mechanical properties of several species of woodflour filled polypropylene .....	26
5.1 The density of polypropylene irradiated with gamma rays at various doses.....	57
5.2 The melting temperature of polypropylene irradiated with gamma rays at various doses. ....	57
5.3 The degradation temperature and char yield of polypropylene irradiated with gamma rays at various doses.....	58
5.4 The density of woodflour composites irradiated with gamma rays at various doses. .....	58
5.5 The melting temperature of polypropylene woodflour composites irradiated with gamma rays at various doses.....	59
5.6 The degradation temperature and char yield of polypropylene woodflour composites irradiated with gamma rays at various doses.....	59
5.7 The density of PP/wood/PP-g-MA composites irradiated with gamma rays at various doses.....	60
5.8 The mechanical properties of PP wood composites irradiated with gamma rays at various doses.....	60
B-1 Flexural properties of polypropylene at various doses.....	95
B-2 Tensile properties of polypropylene at various doses. ....	95
B-3 Flexural properties of rubber wood at various doses. ....	96

B-4 Flexural properties of PP/woodflour composite at various doses.....	96
B-5 Tensile properties of PP/woodflour composite at various doses. ....	97
B-6 Flexural properties of PP/wood/PP-g-MA composite at various doses. ....	97
B-7 Tensile properties of PP/wood/PP-g-MA composite at various doses.....	98
C-1 Extraction percentages of polypropylene at various doses.....	99
C-2 Extraction percentages of polypropylene/woodflour composites at various doses .....	100
C-3 Extraction percentage of polypropylene/ woodflour composites with PP-g-MA at various doses.....	100

## LIST OF FIGURES

FIGURE	PAGE
2.1 Typical tensile strength-concentration curves for filled polymers showing upper and lower bound responses .....	10
2.2 Regions of creep behavior strain vs. time.....	12
2.3 Shapes of reinforcing fillers.....	16
3.1 The effects of stress level on creep behavior of WPC.....	27
3.2 Modification scheme for esterification reaction between wood particles and maleated polyolefins.....	28
3.3 SEM micrographs of PP/wood composites .....	30
3.4 Tensile strength of PP/wood and sisal fiber composite vs. irradiation dose .....	31
3.5 Tensile modulus of PP/wood flour and PP/sisal fibre composites vs. irradiation dose.....	32
3.6 Elongation at break of PP/wood and sisal fiber composite vs. irradiation dose ..	32
3.7 The effect of $\gamma$ -irradiation on the DSC thermograms (second heating) of PP/wood flour .....	33
5.1 Effect of gamma irradiation on discoloration of polypropylene.....	61
5.2 The flexural properties of polypropylene at various doses.....	62
5.3 The tension properties of polypropylene at various doses.....	63
5.4 Creep behavior of polypropylene at various doses .....	64
5.5 DSC thermograms of polypropylene at various doses. ....	65
5.6 TGA thermograms of polypropylene at various doses.....	66
5.7 SEM micrographs of polypropylene at various radiation doses .....	67
5.8 The average particles size of rubber wood flour.....	68
5.9 The flexural modulus of rubber wood at various doses.....	69
5.10 The flexural strength of rubber wood at various doses .....	70
5.11 TGA thermogram of rubber wood .....	71
5.12 Effect of gamma irradiation on discoloration of woodflour composites.....	72

5.13 The flexural properties of polypropylene woodflour composites at various doses .....	73
5.14 The tension properties of polypropylene woodflour composites at various doses .....	74
5.15 Creep behavior of polypropylene and composites .....	75
5.16 Creep behavior of polypropylene woodflour composites at various doses .....	76
5.17 DSC thermograms of polypropylene woodflour composites at various doses ....	77
5.18 TGA thermograms of polypropylene, rubber wood, and polypropylene wood composites at various doses .....	78
5.19 SEM Micrographs of polypropylene woodflour composites at various radiation doses .....	79
5.20 Effect of gamma irradiation on discoloration of woodflour composites treated with maleic anhydride graft polypropylene .....	80
5.21 The flexural properties of polypropylene woodflour composites treated with maleic anhydride graft polypropylene at various doses .....	81
5.22 The tension properties of polypropylene woodflour composites treated with maleic anhydride graft polypropylene at various doses .....	82
5.23 Creep behavior of polypropylene woodflour composites treated with maleic anhydride graft polypropylene at various doses.....	83
5.24 Xylene extraction of polypropylene and polypropylene woodflour composites at various doses .....	84
5.25 Xylene extraction of polypropylene woodflour composites at various doses treated with maleic anhydride graft polypropylene at various doses .....	85
5.26 Creep behavior of polypropylene wood composites at various dose rates.....	86



## LIST OF SCHEMES

SCHEME	PAGE
2.1 Classifications of polymer-based composites.....	6
2.2 Important models for predicting modulus value of filled polymer.....	8
2.3 Classifications of radiations.....	18