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นางสาว อินธิรา สังวรารณ์

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**IRON-CONTAINING MFI CATALYSTS FOR CONVERSION OF METHANOL TO  
OLEFINS**

**Miss Intira Sangvaraporn**

**ศูนย์วิทยทรัพยากร  
จุฬาลงกรณ์มหาวิทยาลัย**

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..... Dean of Faculty of Science  
(Professor Piamsak Menasveta, Ph.D.)

THESIS COMMITTEE

Siri Varothai ..... Chairman  
(Associate Professor Siri Varothai, Ph.D.)

A. Chaisuwan ..... Thesis Advisor  
(Aticha Chaisuwan, Ph.D.)

Vudhichai Parasuk ..... Member  
(Associate Professor Vudhichai Parasuk, Ph.D.)

T. Tuntulani ..... Member  
(Assistant Professor Thawatchai Tuntulani, Ph.D.)

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ได้สังเคราะห์ตัวเร่งปฏิกิริยาเอ็มเอฟไอที่มีเหล็กเป็นองค์ประกอบที่มีอัตราส่วนซิลิกอนต่อเหล็กหลายอัตราส่วน (10 ถึง 90) โดยตรงภายใต้สภาวะไฮโดรเทอร์มอล ได้ศึกษาผลของพื้นผิวเจลผสมเริ่มน้ำ (8.0 ถึง 11.0) ต่อความเป็นผลึกของเหล็ก-เอ็มเอฟไอ (อัตราส่วนซิลิกอนต่อเหล็กในเจล 90) และตำแหน่งของเหล็กในเฟรมเวิร์กหรือนอกเฟรมเวิร์กด้วยเทคนิคการเลี้ยวเบนของรังสีเอกซ์ กล้องจุลทรรศน์อิเล็กตรอนแบบส่องกระดาษ เครื่องวิเคราะห์ขนาดอนุภาค อิเล็กตรอนสปีนเรโซナンซ์ และการคายรังสีของอะตอม พบว่าพื้นผิวของเจลมีผลต่อขนาดของอนุภาค รูปร่าง และปริมาณเหล็ก แต่ละตำแหน่ง ข้อมูลการเลี้ยวเบนของรังสีเอกซ์แสดงว่าสารที่สังเคราะห์ได้ทุกตัวมีโครงสร้างแบบเอ็มเอฟไอ และความเป็นผลึกของเหล็ก-เอ็มเอฟไอลดลงเมื่อปริมาณเหล็กเพิ่มขึ้น ได้เตรียมไฮโดรเจน เหล็ก-เอ็มเอฟไอโดยการแยกเปลี่ยนไฮอนกับสารละลายแอมโมเนียม คลอไรด์ ที่อุณหภูมิจุดเดือดด้วยการคนข้ามคืนตามด้วยการเผา และทำซ้ำทั้งหมดอีกหนึ่งครั้ง การนีโอเดียมไฮอน อุณหภูมิ (300 ถึง 500 องศาเซลเซียส) อัตราส่วนซิลิกอนต่อเหล็ก และรูปร่างมีผลต่อปรอร์เซ็นต์การเปลี่ยนเมทานอล การทำให้เกิดและการเลือกจำเพาะต่อผลิตภัณฑ์ ไฮเดียมไฮอนมีผลต่อประสิทธิภาพของตัวเร่งปฏิกิริยา เนื่องจากไฮเดียมไฮอนบดบังตำแหน่งความเป็นกรด ได้การเปลี่ยนเมทานอลเกือบสมบูรณ์และความเลือกจำเพาะต่อพรอพิลีนสูงสุดที่ 450 องศาเซลเซียสบนเหล็ก-เอ็มเอฟไอ (อัตราส่วนซิลิกอนต่อเหล็ก 87) ขณะที่การเกิดโค้กสะสมบนตัวเร่งปฏิกิริยาน้อยใน การผ่านการเกิดปฏิกิริยานั่นเอง การลดปริมาณเหล็กทำให้ได้การเลือกจำเพาะต่อโอลีฟินนิดเบา เพิ่มขึ้น และลดการเกิดโค้กสะสมบนตัวเร่งปฏิกิริยา ได้การเกิดผลิตภัณฑ์ที่เป็นแก๊สสูงสุดเมื่อใช้เหล็ก-เอ็มเอฟไอทรงกลมเมื่อเทียบกับเหล็ก-เอ็มเอฟไอเครื่องหมายบวก

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The iron-containing-MFI catalysts with various Si/Fe ratios (10 to 90) were synthesized by direct synthesis under hydrothermal condition. The effect of pH (8.0 to 11.0) of the starting gel mixture on the crystallinity of the selected Fe-MFI (Si/Fe ratio in gel of 90), and sites of framework or non-framework sites incorporated irons, were investigated using X-ray diffractometer, scanning electron microscope, particle size analyzer, electron spin resonance and atomic absorption. It was found that the pH of gel affects the particle size, morphology, and the amount of iron in each site. XRD data show that all products have MFI structure and the crystallinity of Fe-MFI decreases with the increase in iron content. Hydrogen Fe-MFI was prepared by ion exchange with ammonium chloride solution at boiling temperature with stirring overnight followed by calcinations, and the treatment was repeated once more. The present sodium ions, temperature (300-500°C), Si/Fe ratio, and morphology have strong influence on the %conversion of methanol, yield and selectivity of products. Sodium ions affect the activity of the catalyst because they blocked the acid sites. Nearly complete conversions of methanol and highest selectivity to propylene are obtained at 450°C over Fe-MFI (Si/Fe ratio = 87), where coke deposition on the single-run catalysts was a low. Decreasing of iron content leads to higher selectivity to light olefins and less coke deposited on the catalyst. The highest yield of gas products was obtained using spherically-shape compared to plus-like Fe-MFI.

# ศูนย์วิทยทรัพยากร จุฬาลงกรณ์มหาวิทยาลัย

Department.....Chemistry..... Student's signature.....*Intira Sangvaraporn*

Field of study.....Chemistry..... Advisor's signature.....*A. Chaisuwan*

Academic year....2003.....

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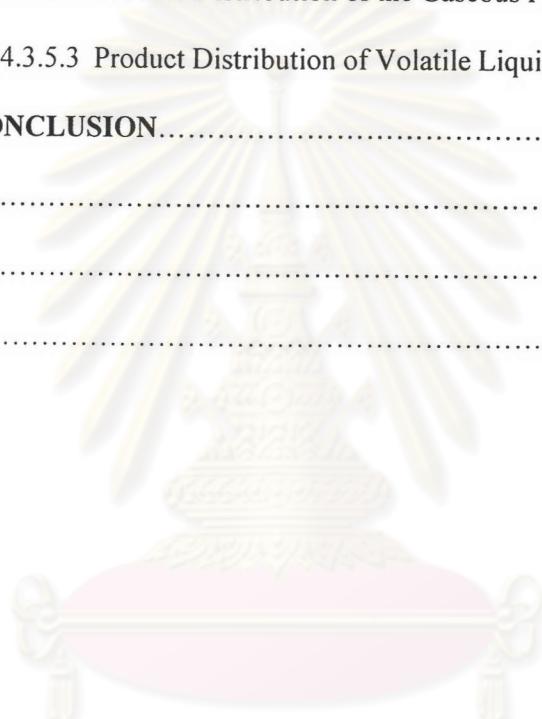
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## LIST OF ABBREVIATIONS

AAS	=	Atomic Absorption Spectrometer
BET	=	Brunauer, Emmett, and Teller
ESR	=	Electron Spin Resonance
Fe-MFI	=	Iron-containing Mobil Five
GC	=	Gas Chromatograph
GC-MS	=	Gas Chromatograph- Mass Spectrometer
GHSV	=	Gas-Hourly Space Velocity
MTO	=	Methanol to Olefins
SEM	=	Scanning Electron Microscope
TOS	=	Time on Stream
TPABr	=	Tetrapropyl ammonium bromide
XRD	=	X-ray diffraction or diffractometer
ZSM-5	=	Zeolite Socony Mobil Five