

การแปลงซีลิกาเอสบีเอ-15 เป็นซีโอไลต์บีตาสำหรับการแตกตัวของขยะพลาสติกและน้ำมันดิบที่
ได้จากพลาสติก



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วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรดุษฎีบัณฑิต

สาขาวิชาเคมี ภาควิชาเคมี

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ปีการศึกษา 2552

ลิขสิทธิ์ของจุฬาลงกรณ์มหาวิทยาลัย



TRANSFORMATION OF SILICA SBA-15 TO ZEOLITE BETA FOR CRACKING
OF PLASTIC WASTE AND PLASTIC-DERIVED CRUDE OIL

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A Dissertation Submitted in Partial Fulfillment of the Requirements
for the Degree of Doctor of Philosophy Program in Chemistry

Department of Chemistry

Faculty of Science


Chulalongkorn University

Academic year 2009

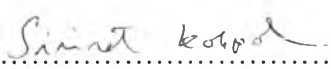
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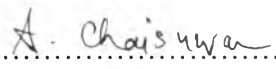
Thesis Title TRANSFORMATION OF SILICA SBA-15 TO ZEOLITE
 BETA FOR CRACKING OF PLASTIC WASTE AND
 PLASTIC-DERIVED CRUDE OIL
By Miss Khanitha Wanchai
Field of Study Chemistry
Thesis Advisor Aticha Chaisuwan, Ph.D.

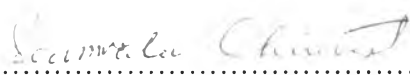
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ขนิษฐา วันชัย : การแปลงซิลิกาเอสบีเอ-15 เป็นซีโอไลต์บีตาสำหรับการแตกตัวของขยะ
พลาสติกและน้ำมันดิบที่ได้จากพลาสติก (TRANSFORMATION OF SILICA SBA-15 TO
ZEOLITE BETA FOR CRACKING OF PLASTIC WASTE AND PLASTIC-DERIVED
CRUDE OIL) อ. ปรีกษาวิทยานิพนธ์หลัก: อ.ดร.อริชา ฉายสุวรรณ, 178 หน้า

ได้ใช้วิธีใหม่สังเคราะห์ซีโอไลต์บีตาที่มีขนาดอนุภาคระดับนาโนโดยการแปลงเอสบีเอ-15ในสภาพซึ่งมี
เทระเอทิลแอมโมเนียมไฮดรอกไซด์ และอะลูมิเนียมไอโซโพรพอกไซด์ที่อุณหภูมิคงที่ 135 องศาเซลเซียส ได้
ศึกษาผลของตัวแปรต่างๆ เช่น ระยะเวลาในการแปลง อัตราส่วนของเทระเอทิลแอมโมเนียมไฮดรอกไซด์ต่อ
ซิลิกา และอัตราส่วนของเอสบีเอ-15ต่ออะลูมิเนียมไอโซโพรพอกไซด์ ตรวจสอบลักษณะเฉพาะของซีโอไลต์
บีตาด้วยเทคนิคการเลี้ยวเบนรังสีเอ็กซ์ กล้องจุลทรรศน์แบบส่องกราด อะลูมิเนียม เมจิกแองเกิลสปีนนิวเคลียร์
แมกเนติกเรโซแนนซ์ การดูดซับไนโตรเจน การคายแอมโมเนียด้วยการเพิ่มอุณหภูมิแบบตั้งโปรแกรม และ
เอ็กซ์เรย์ฟลูออเรสเซนซ์ ที่อัตราส่วนของเทระเอทิลแอมโมเนียมไฮดรอกไซด์ต่อซิลิกาเท่ากับ 0.39 เอสบีเอ-15
สลายตัวเป็นแอลฟา-ควอทซ์ และค่อยๆ เปลี่ยนเป็นซีโอไลต์บีตาโดยมีความเป็นผลึกสูงสุดภายในระยะเวลา
24 ชั่วโมง อย่างไรก็ตามได้ซีโอไลต์ที่มีปริมาณสูงสุดร้อยละ 42.4 หลังจากการแปลงโครงสร้างเป็นเวลา
48 ชั่วโมง ตัวอย่างส่วนใหญ่ที่มีซีโอไลต์บีตามีขนาดอนุภาคในช่วง 116-147 นาโนเมตร ขึ้นกับเวลาที่ใช้ในการ
แปลงโครงสร้าง แต่ไม่มีผลมากนักต่อขนาดผลึกและพื้นที่ผิวจำเพาะชนิดบีอีที โดยการแปรค่าอัตราส่วนเทระ
เอทิลแอมโมเนียมไฮดรอกไซด์ต่อซิลิกาจาก 0.10 ถึง 0.39 พบว่าที่อัตราส่วนของเทระเอทิลแอมโมเนียม
ไฮดรอกไซด์ต่อซิลิกาเท่ากับ 0.26 ให้ปริมาณซีโอไลต์สูงสุดถึงร้อยละ 71.8 โดยมีขนาดอนุภาคเฉลี่ย
215 นาโนเมตร ซีโอไลต์บีตาที่เตรียมได้ถูกนำไปทดสอบความว่องไวของการเร่งปฏิกิริยาในการแตกตัวของ
พอลิโพรพิลีน ขยะพอลิเอทิลีนชนิดความหนาแน่นสูง และน้ำมันที่ได้จากการแตกย่อยพอลิโพรพิลีนใน
รีแอกเตอร์ชนิดไมโครเนื่องภายใต้ภาวะต่างๆ พบว่า ซีโอไลต์บีตามีความว่องไวของการเร่งปฏิกิริยามากถึงร้อยละ
95.8 การเปลี่ยนค่าในการแตกตัวของพอลิโพรพิลีน เห็นได้ชัดเจนว่าการเปลี่ยนพลาสติกและปริมาณของผลิตภัณฑ์
ขึ้นกับอุณหภูมิที่ใช้ในการแตกย่อย อัตราส่วนของเอสบีเอ-15 ต่ออะลูมิเนียมไอโซโพรพอกไซด์ และอัตราส่วน
ของพลาสติกต่อตัวเร่งปฏิกิริยา ความเลือกจำเพาะต่อชนิดของผลิตภัณฑ์ได้รับผลจากตัวแปรเหล่านี้ ผลิตภัณฑ์
ชนิดแก๊สซึ่งได้จากการแตกย่อยพอลิโพรพิลีน และพอลิเอทิลีนชนิดความหนาแน่นสูงส่วนใหญ่ประกอบด้วย
โพรพิน ไอโซบิวทีน และสารที่มีจุดเดือดสูงกว่านอมัลเพนเทน ขณะที่แก๊สที่ได้จากการแตกย่อยน้ำมันที่ได้จาก
การแตกย่อยพอลิโพรพิลีน คือ โพรพิน นอมัลบิวเทน ไอโซบิวทีน และสารที่มีจุดเดือดสูงกว่านอมัลเพนเทน
ผลิตภัณฑ์ส่วนที่เป็นของเหลวที่ได้จากการแตกย่อยพลาสติกทั้งสองชนิดส่วนใหญ่อยู่ในช่วงจุดเดือดจาก เฮกเซน
ถึงโนเนน และสำหรับน้ำมันที่ได้จากการแตกย่อยพอลิโพรพิลีนผลิตภัณฑ์ชนิดของเหลวที่ได้คือ โนเนน เป็น
ส่วนใหญ่ ตัวเร่งปฏิกิริยาที่ผ่านการใช้งานแล้วสามารถทำให้กลับคืนสภาพเดิมได้ง่ายโดยการเผาที่อุณหภูมิสูง
และความว่องไวของมันยังใกล้เคียงกับตัวเร่งปฏิกิริยาที่ยังไม่ได้ผ่านการใช้งาน

ภาควิชาเคมี.....ลายมือชื่อนิสิต.....
สาขาวิชาเคมี.....ลายมือชื่อ อ. ปรีกษาวิทยานิพนธ์หลัก.....
ปีการศึกษา2552.....

4873809323: MAJOR CHEMISTRY

KEYWORDS: BEA / CATALYTIC CRACKING / HDPE/ MATERIAL / MESOPOROUS /
MICROPOROUS / PP / SBA-15 / TRANSFORMATION / ZEOLITE BETA

KHANITHA WANCHAI: TRANSFORMATION OF SILICA SBA-15 TO
ZEOLITE BETA FOR CRACKING OF PLASTIC WASTE AND PLASTIC-
DERIVED CRUDE OIL. THESIS ADVISOR: ATICHA CHAISUWAN, Ph.D.,
178 pp.

A novel method for the synthesis of nanoparticle zeolite beta was performed by transformation of SBA-15 in the presence of tetraethylammonium hydroxide and aluminum isopropoxide under autogenous pressure at a constant temperature of 135°C. The effects of various parameters such as transformation periods, TEAOH/SiO₂ ratio and SBA-15/AIP ratio on the properties of the zeolite product were studied. The zeolite samples were characterized by XRD, SEM, ²⁷Al-MAS-NMR, nitrogen adsorption, NH₃-TPD and XRF techniques. With a TEAOH/SiO₂ ratio of 0.39, SBA-15 is completely decomposed to α-quartz and gradually to the zeolite beta with the maximum crystallinity within a period of 24 h. However, the zeolite is obtained at the highest yield of 42.4 % after transformation for 48 h. Most samples containing zeolite beta have the particle sizes ranging from 116 to 147 nm depending on the transformation period, but the crystallite size and BET specific surface area are not significantly affected. By varying TEAOH/SiO₂ ratios from 0.10 to 0.39, the TEAOH/SiO₂ ratio of 0.26 is found to give the highest yield of zeolite beta up to 71.8 % with larger average particle size of 215 nm. The zeolite beta samples obtained were tested for their catalytic activities in the cracking of PP, HDPE waste and PP-derived crude oil in a batch reactor under various conditions. The nanoparticle zeolite beta is found very active up to 95.8 % conversion in cracking of PP waste. The plastic conversions and the yields of products fractions obviously depend on the reaction temperatures, the SBA-15/AIP ratios and the plastic to catalyst ratios. The product selectivity is affected by those factors. The gas products obtained by PP and HDPE cracking are mainly propene, *i*-butene and C₅⁺ while those from catalytic cracking of PP-derived crude oil are propene, *n*-butane, *i*-butene and C₅⁺. The liquid products obtained by cracking of both types of plastic are mainly in the boiling point ranging from C₆ to C₉ and for PP-derived crude oil C₉ is mainly obtained. The used catalyst can be regenerated easily by calcination and its activity remains comparable to the fresh catalyst.

Department..... Chemistry..... Student's Signature *Khanitha Wanchai*
Field of Study..... Chemistry..... Advisor's Signature *A. Chaisuwan*
Academic year..... 2009.....

ACKNOWLEDGEMENTS

The success of this thesis can be attributed to the extensive support and assistance from Dr. Aticha Chaisuwan, my thesis advisor. I deeply thank her for the valuable advice and guidance in this research and her kindness throughout this study.

I would like to give my gratitude to Associate Professor Dr. Sirirat Kokpol, Assistant Professor Dr. Soamwadee Chaiansutcharit, Assistant Professor Dr. Warinthorn Chavasiri, and Dr. Suchada Butnark, from PTT Public Company Limited as the chairman and member of this thesis committee, respectively, for all of their kindness and useful advice in the research.

I would like to gratefully thank PTT Chemical Public Company Limited for supporting the standard mixtures for GC analysis. Moreover, I would like to thank Department of Chemistry, Faculty of Science, Chulalongkorn University for the valuable knowledge and experience. I would like to thank the Office of The High Education Commission for supporting a research fund. In addition, Thailand Japan Technology Transfer Project supported a loan by Japan Banks for International Cooperation (TJTTP-JBIC) for instrument support. Furthermore, I would like to thank the members of Materials Chemistry and Catalysis Research Unit for their help and encouragement throughout the course of my research.

Finally, I would like to express my gratitude to beloved parents and my family for encouragement, assistance during my study and research. The usefulness of this thesis, I dedicated this work to my father, my mother and all the teachers who have taught me since my childhood.

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LIST OF ABBREVIATIONS

TEAOH	Tetraethylammonium hydroxide
PEO ₂₀ -PPO ₇₀ -PEO ₂₀	Poly(ethylene oxide) ₂₀ -poly(propylene oxide) ₇₀ -poly(ethylene oxide) ₂₀
BEA	Zeolite beta
SBA-15	Santa Barbara Amorphous-15
TEOS	Tetraethyl orthosilicate
BET	Brunauer- Emmett-Teller
BJH	Barret, Joyner, and Halenda
XRD	X-ray Diffraction
SEM	Scanning Electron Microscopy
TPD	Temperature-Programmed Desorption
GC	Gas Chromatography
MAS-NMR	Magic-angle-spinning-nuclear magnetic resonance
PP	Polypropylene
HDPE	High density polyethylene
°C	degree Celsius
g	gram (s)
h	hour (s)
mg	milligram (s)
min	minute (s)
ppm	part per million or milligram per liter
M	molar
cps	counts per second
%	percentage