

**n-OCTANE AROMATIZATION ON Pt SUPPORTED ON NOVEL
ZEOLITES**



Supak Trakarnroek

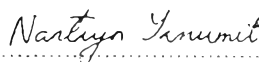
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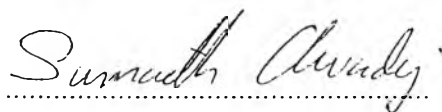
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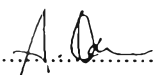
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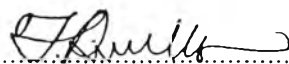
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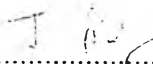

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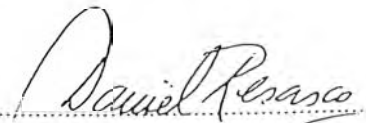
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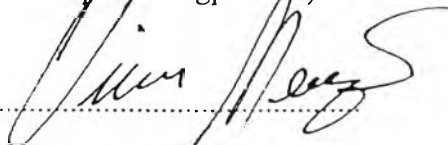

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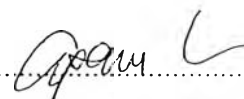

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ABSTRACT

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Supak Trakarnroek: n-Octane Aromatization on Pt Supported on Novel Zeolites

Thesis Advisors: Prof. Somchai Osuwan, Prof. Daniel E. Resasco, Assoc. Prof. Thirasak Rirksomboon, and Dr. Siriporn Jongpatiwut
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KL zeolites with cylindrical shape are effective catalyst supports for n-octane aromatization, but the effectiveness strongly depends on the channel length of the zeolite crystallite as well as Pt dispersion and location of the Pt cluster. The different Pt/KL catalysts in the series were compared in the aromatization of n-octane at 500°C and atmospheric pressure. It was found that the catalysts with shorter channel length exhibited improved activity, selectivity, and catalyst life. In addition, the catalysts with shorter channels had a much lower extent of secondary hydrogenolysis. Consequently, more C₈ aromatics are preserved in the product and less benzene and toluene are produced compared to the catalysts with longer channels. In the case that the inactive metal was added into Pt/KL catalysts, it was found that the addition of tin improved the stability of the Pt/KL catalyst by inhibiting the adsorption of dehydrogenated species which is the intermediate for formation of coke. Furthermore, the selectivity to C₈-aromatics products is increased but the secondary hydrogenolysis reaction, which occurred inside the pore of KL zeolite, is decreased. In addition, it was found that Co₁Pt₁Sn prepared by vapor phase co-impregnation method yielded a high fraction of alloy phase compared to Se₁Pt₁Sn and Se₁Sn₁Pt which were prepared by vapor phase sequential impregnation method. As a result, Co₁Pt₁Sn gave the highest activity and C₈-aromatics selectivity among the Pt-Sn catalysts investigated. Moreover, it was observed that OX molecules were produced more compared to without addition of tin because of electronic effect. As a result, the EB/OX ratio was lower than unity.

บทคัดย่อ

สุภัท ตระการฤกษ์ : ปฏิกริยาอะโรมาไทเซชันของนอร์มัลออกเทนบนโลหะแพลททินัมที่อยู่บนซีโอไลต์ชนิดใหม่ (n-Octane Aromatization on Pt Supported on Novel Zeolites) อ. ที่ปรึกษา : ศ. สมชาย โอสุวรรณ ศ. แคนเน็ล อี ริชส์โก รศ. ดร. ชีรศักดิ์ ฤกษ์ สมบูรณ์ และ ดร. ศิริพร จงผาคิวติ 112 หน้า

ซีโอไลต์แอลที่มีรูปร่างทรงกระบอกเป็นตัวรองรับที่มีประสิทธิภาพในการเป็นตัวเร่งปฏิกิริยาสำหรับปฏิกิริยาอะโรมาไทเซชันของนอร์มัลออกเทน อย่างไรก็ตามประสิทธิภาพของตัวเร่งปฏิกิริยาขึ้นอยู่กับความยาวของท่อซีโอไลต์ การกระจายตัวของโลหะแพลททินัม และตำแหน่งของโลหะแพลททินัม ในงานนี้ได้ใช้ตัวเร่งปฏิกิริยา Pt/KL ชนิดต่างๆมาเปรียบเทียบความเป็นตัวเร่งปฏิกิริยาของปฏิกิริยาอะโรมาไทเซชันของนอร์มัลออกเทน ที่อุณหภูมิ 500°C ณ ความดันบรรยากาศ ผลการทดลองพบว่า ตัวเร่งปฏิกิริยาที่มีความยาวของท่อสั้นให้ความว่องไวในการเป็นตัวเร่งปฏิกิริยาดี มีความเลือกเฉพาะกับผลิตภัณฑ์ที่ต้องการเป็นอย่างดี และมีอายุการใช้งานนาน นอกจากนี้ยังลดการเกิดปฏิกิริยาไฮโดรจีโนไลซิส ส่งผลให้ได้ผลิตภัณฑ์ที่เป็น C8 อะโรมาติกมาก พร้อมทั้งลดการเกิดของเบนซินและโทลูอินเมื่อเทียบกับตัวเร่งปฏิกิริยาที่มีท่อยาวกว่า ในกรณีที่เติมโลหะที่ไม่ว่องไวลงบนตัวเร่งปฏิกิริยา Pt/KL ผลการทดลองพบว่าการเติมโลหะดีบุกช่วยปรับปรุงความเสถียรของตัวเร่งปฏิกิริยาเนื่องจากโลหะดีบุกช่วยยับยั้งการดูดซับของโมเลกุลจำพวกดีไฮโรจีนที่ซึ่งเป็นตัวกลางในการเกิดโค้ก และมีความเลือกเฉพาะกับผลิตภัณฑ์ที่เป็น C8 อะโรมาติกมากขึ้น รวมทั้งลดการเกิดปฏิกิริยาไฮโดรจีโนไลซิสซึ่งเกิดขึ้นในรูพรุนของซีโอไลต์ อีกทั้งพบว่าตัวเร่งปฏิกิริยา Pt-Sn/KL ที่เตรียมจากการเติมโลหะแพลททินัมและดีบุกพร้อมกันมีเฟสที่เป็น PtSn อัดลดยึดมากกว่าตัวเร่งปฏิกิริยา Pt-Sn/KL ที่เตรียมจากการเติมโลหะแพลททินัมและดีบุกแบบลำดับ ส่งผลให้ตัวเร่งปฏิกิริยา Pt-Sn/KL ชนิดนี้มีความว่องไวและมีความเลือกเฉพาะกับผลิตภัณฑ์ที่เป็น C8 อะโรมาติกมากกว่าตัวเร่งปฏิกิริยาชนิดอื่นๆ นอกจากนี้ยังพบว่า ออร์โทไซลีน (OX) เกิดได้ง่ายกว่าการเกิดเอทิลเบนซิน (EB) อันเป็นผลเนื่องมาจากการถ่ายโอนอิเล็กตรอนจากโลหะดีบุกไปยังโลหะแพลททินัม ส่งผลให้ อัตราส่วน EB/OX มีค่าน้อยกว่า 1

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6.12 The variation of benzene selectivity with n-octane conversion of various Sn/Pt ratios of the bimetallic Pt-Sn/KL catalysts prepared by vapor phase co-impregnation; reaction conditions: temperature = 500°C, pressure = 1 atm, WHSV	

FIGURE	PAGE
$= 0.5-5 \text{ h}^{-1}$, and $\text{H}_2:\text{HC} = 6:1$.	100
6.13 The variation of EB/OX ratio with Sn/Pt ratios of the bimetallic Pt-Sn/KL catalysts prepared by vapor phase co-impregnation; reaction conditions: temperature = 500°C , pressure = 1 atm, $\text{WHSV} = 5 \text{ h}^{-1}$, and $\text{H}_2:\text{HC} = 6:1$.	100
6.14 TPO profiles of various Sn/Pt ratios of the bimetallic Pt-Sn/KL catalysts prepared by vapor phase co-impregnation method.	101