

**CATALYTIC CONVERSION OF METHYLESTER OVER ZEOLITE  
CATALYSTS FOR BIODIESEL UPGRADING**

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for the Degree of Doctor of Philosophy  
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in Academic Partnership with  
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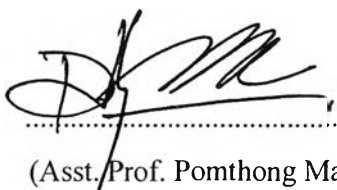
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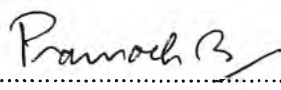
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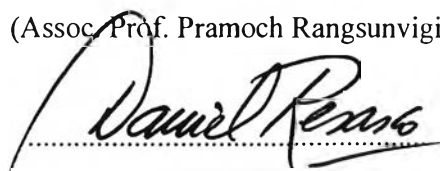
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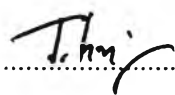
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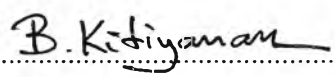
  
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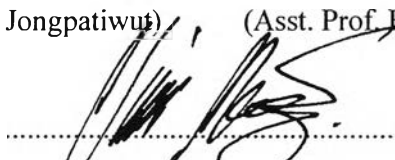
  
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## ABSTRACT

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Tanate Danuthai: Catalytic Conversion of Methyl ester over Zeolite Catalysts for Biodiesel Upgrading

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Due to the thermal and oxidative instability of the biodiesel, the elimination of the oxygen content has been proposed to improve the stability of the fuel and its utilization potential. In this work, the catalytic conversion of methyl octanoate, a model biodiesel fuel, to hydrocarbon fuels and chemicals over the H-ZSM5, Zn/H-ZSM5 and CsNaX zeolite catalysts was investigated in a gas phase reaction. It was found that the deoxygenation of methyl octanoate over the H-ZSM5 yielded a variety of hydrocarbons ( $C_1-C_7$ ), with significant amounts of aromatics. Octanoic acid and 8-pentadecanone were primary products. Aromatics are formed through a series of reactions, cracking, oligomerization, cyclization, and direct dehydrocyclization. The addition of the Zn species does not provide any alternative route for aromatization, but only improve the dehydrogenation activity. Moreover, the deoxygenation of 10 wt% methyl octanoate in methanol over the CsNaX catalysts revealed that the decarbonylation/deacetylation activity can occur at high rate and stability. Heptenes and hexenes as main products are produced via primary decomposition of the methyl ester, forming octanoate-like species as an intermediate. Moreover, the methanol decomposition provides the hydrogen for hydrogenation/dehydration. The content of excess cesium on the CsNaX catalysts alters the basic strength, leading to the difference in the product distributions. The basicity and the highly polar character of the zeolite play an important role in the decarbonylation.

## บทคัดย่อ

รณรงค์ คนุไทย : การเปลี่ยนแปลงของเมทิลเอสเตอร์บนตัวเร่งปฏิกิริยาประเภทซีโอไลต์เพื่อเพิ่มคุณภาพของน้ำมันไบโอดีเซล (Catalytic Conversion of Methyl ester over Zeolite Catalysts for Biodiesel Upgrading) อ. ที่ปรึกษา: ศ.ดร. สมชาย โอสุวรรณ รศ. ดร. วีรศักดิ์ ฤกษ์สมบูรณ์ ผศ.ดร. ศิริพร จงผาคิวุฒิ และ ศ.ดร. แคนเน็ล อี ริชส์โก 121 หน้า

เนื่องด้วยน้ำมันไบโอดีเซลที่ได้จากกระบวนการทรานเอสเตอริฟิเคชันมีองค์ประกอบของออกซิเจนในโมเลกุล จึงทำให้ไม่เสถียรต่อความร้อนและปฏิกิริยาออกซิเดชัน ดังนั้นการกำจัดออกซิเจนในโมเลกุลเพื่อเพิ่มความเสถียรและศักยภาพในการใช้น้ำมันไบโอดีเซลจึงเป็นงานที่น่าสนใจ งานวิจัยชิ้นนี้ได้ศึกษาการเปลี่ยนแปลงของเมทิลออกทานโอเอทซึ่งเป็นสารต้นแบบบนตัวเร่งปฏิกิริยาซีโอไลต์ประเภท แซคเอสเอ็มไฟว์ (H-ZSM5) สังกะสีบนแซคเอสเอ็มไฟว์ (Zn/H-ZSM5) และซีเซียมโซเดียมเอ็กซ์ (CsNaX) จากการศึกษาพบว่า ปฏิกิริยาคีออกซิเจนชันของเมทิลออกทานโอเอทบน H-ZSM5 ให้ผลิตภัณฑ์ประเภทไฮโดรคาร์บอนชนิดต่าง ๆ ( $C_1-C_7$ ) รวมทั้งสารประเภทอะโรมาติกส์ โดยมีกรดออกทานอิก และ 8-เพนทาเดคคาโนนเป็นผลิตภัณฑ์ปฐมภูมิ สารประเภทอะโรมาติกส์ถูกสร้างขึ้นจากหลายปฏิกิริยา ได้แก่ ปฏิกิริยาแตกตัวของโมเลกุล ปฏิกิริยารวมตัวของโอเลฟินส์ ปฏิกิริยาปิดวงแหวนอะโรมาติกส์ และปฏิกิริยาคีไฮโดรจิเนชันพร้อมปิดวงแหวนในขั้นตอนเดียว ยังพบอีกว่า การเติมสังกะสีบนตัวเร่งปฏิกิริยาไม่ทำให้เกิดกลไกปฏิกิริยาใหม่ เพียงแต่ช่วยเพิ่มสมบัติการคีไฮโดรจิเนชัน นอกจากนี้การศึกษาปฏิกิริยาคีออกซิเจนชันของสารละลาย 10 เปอร์เซ็นต์ โดยน้ำหนักของเมทิลออกทานโอเอทในเมทานอลบนตัวเร่งปฏิกิริยา CsNaX พบว่า ปฏิกิริยาคีคาร์บอนิลเลชันและปฏิกิริยาคีอะเซทิลเลชันสามารถเกิดขึ้นด้วยอัตราการเกิดที่สูงและมีความเสถียรสูง ผลิตภัณฑ์ที่ได้จากปฏิกิริยาส่วนใหญ่เป็นสารจำพวกเฮปทีนและเฮกซีน โดยเกิดผ่านปฏิกิริยาการย่อยสลายปฐมภูมิของสารเมทิลเอสเตอร์ ซึ่งมีสารที่มีโครงสร้างคล้ายเมทิลออกทานโอเอทเป็นสารตัวกลาง นอกจากนี้การสลายตัวของเมทานอลยังผลิตไฮโดรเจนสำหรับปฏิกิริยาไฮโดรจิเนชันและปฏิกิริยาคีไฮโดรเจชัน ปริมาณของซีเซียม (Cs) ส่วนเกินบนตัวเร่งปฏิกิริยา CsNaX ส่งผลต่อความแข็งแรงเชิงเบสของตัวเร่งปฏิกิริยา ซึ่งทำให้เกิดผลิตภัณฑ์ที่แตกต่างกัน ทั้งนี้ความเป็นเบสและสมบัติความมีขั้วที่สูงของซีโอไลต์มีบทบาทสำคัญมากในการเกิดปฏิกิริยาคีคาร์บอนิลเลชัน

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