

**HYDROGEN PRODUCTION BY OXIDATIVE STEAM REFORMING OF
METHANOL OVER Au-CuO/CeO₂ CATALYSTS**


Phatchanon Pipatpratanporn

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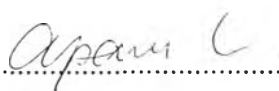
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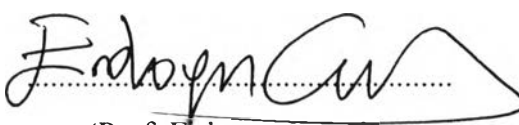
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By: Phatchanon Pipatpratanporn
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Thesis Advisors: Asst. Prof. Apanee Luengnaruemitchai
Prof. Erdogan Gulari

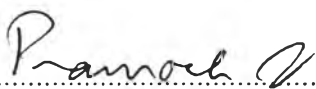
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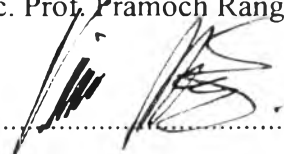

..... College Dean
(Asst. Prof. Pomthong Malakul)

Thesis Committee:


.....
(Asst. Prof. Apanee Luengnaruemitchai)


.....
(Prof. Erdogan Gulari)


.....
(Assoc. Prof. Pramoch Rangsunvigit)


.....
(Assoc. Prof. Vissanu Meeyoo)

ABSTRACT

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Keywords: Oxidative Steam Reforming of Methanol/ Gold-Copper oxide Based
Catalyst/ Hydrogen Production/Au-CuO/CeO₂

Oxidative steam reforming of methanol has been performed on a variety of supported Au catalysts. In this work, the catalysts were prepared by co-precipitation with various (Cu/Au) (wt/wt) ratio and calcinations temperature. Their catalytic activity were studied in a micro-reactor with various reaction temperatures ranging from 200 to 400°C under atmospheric pressure. A feed of 1.5 ml/hour of the mixture of distilled water, oxygen, and methanol over 3% Au-CuO/CeO₂ catalyst was used. Among the catalysts tested, the 3%(5:1), (Cu/Au) Au-CuO/CeO₂ gave methanol conversion and hydrogen yield of 95% and 59% 400°C, respectively. Moreover, the catalysts were characterized for their morphology, size of the Au particle, and reduction behavior by XRD, TEM, and TPR, respectively. The TPR result of the 3% Au-CuO/CeO₂ showed that there is some changing in reduction peaks when Cu was doped. Shifting of reduction peaks referred to the metal-metal interaction between Cu and Au particle which reduced the interaction between Au and CeO₂ support. Additionally, in the UV-visible spectra showed revealed that 3% (5:1), (Cu/Au) Au-CuO/CeO₂ calcined at 200°C exhibited the biggest size of Au particle with highest Au metallic particle. Moreover, from TEM image, it was showed that gold particle size is between 5 to 10 nm. Finally, 3%(5:1), (Cu/Au) Au-CuO/CeO₂ catalyst calcined at 200°C was used to catalyze DCM and SRM. The result showed that catalyst yield the low methanol conversion for DCM but suitable for SRM.

บทคัดย่อ

ภัทรชนน พิพัฒน์ประทานพร : กระบวนการผลิตก๊าซไฮโดรเจนจากปฏิกิริยาเปลี่ยนรูปเมทานอลด้วยไอน้ำและออกซิเจนโดยใช้ตัวเร่งปฏิกิริยาทองและคอปเปอร์ออกไซด์บนซีเรียออกไซด์ (Hydrogen Production from the Oxidative Steam Reforming of Methanol over Au/Fe₂O₃-CeO₂) อ. ที่ปรึกษา : ผศ.ดร. อาภาณี เหลืองนฤมิตชัย และ ศ.ดร. เออโดแกน กุลารี่ 92 หน้า

ในงานวิจัยนี้ศึกษาปฏิกิริยาการเปลี่ยนรูปเมทานอลโดยใช้ไอน้ำและออกซิเจนด้วยตัวเร่งปฏิกิริยาทองและคอปเปอร์ออกไซด์ที่อัตราส่วนต่างๆบนตัวรองรับซีเรียออกไซด์ โดยที่เปลี่ยนแปลงอุณหภูมิในการเผาตัวเร่งปฏิกิริยาระหว่าง 200 ถึง 500 องศาเซลเซียส โดยตัวเร่งปฏิกิริยาจะเตรียมโดยใช้วิธี Co-precipitation และนำไปศึกษาความสามารถในการเร่งปฏิกิริยาในเตาปฏิกรณ์ขนาดเล็ก อุณหภูมิที่ใช้ในการศึกษาอยู่ในช่วง 200 ถึง 400 องศาเซลเซียส ภายใต้สภาวะความดันปกติ โดยใช้สารตั้งต้นเป็นน้ำกลั่นผสมเมทานอลทำปฏิกิริยาบนตัวเร่งปฏิกิริยาทอง ผลการศึกษาพบว่าตัวเร่งปฏิกิริยา 3% (5:1), (Cu/Au) Au-CuO/CeO₂ เผาที่ 200 องศาเซลเซียสให้ผลการเปลี่ยนแปลงเมทานอลและให้ค่าผลผลิตเท่ากับ 95% และ 59% ตามลำดับ ที่อุณหภูมิ 400 องศาเซลเซียส ซึ่งให้ค่าสูงที่สุด นอกจากนั้นทำการวิเคราะห์คุณลักษณะของตัวเร่งปฏิกิริยา โดยใช้เทคนิค XRD, TEM, และ TPR จากผลการวิเคราะห์โดยใช้ TPR พบว่าตัวเร่งปฏิกิริยา เมื่อเพิ่มปริมาณของคอปเปอร์ออกไซด์ในตัวเร่งปฏิกิริยานั้นจะเป็นการลดแรงกระทำระหว่างทองและตัวรองรับซีเรียออกไซด์โดยการสร้างแรงกระทำระหว่างทองและคอปเปอร์ออกไซด์ซึ่งส่งผลต่ออุณหภูมิการรีดักชันของตัวเร่งปฏิกิริยา ในขณะเดียวกัน จากผลการวิเคราะห์ด้วย UV-visible พบว่าขนาดของอนุภาคทองบนตัวเร่งปฏิกิริยาดังกล่าวมีขนาดใหญ่ที่สุดและมีปริมาณทองสูงมากซึ่งว่องไวต่อปฏิกิริยาอย่างมากที่สุด จากการศึกษาโดยเครื่อง TEM พบว่าอนุภาคทองมีขนาด 5 ถึง 10 นาโนเมตร นอกจากนั้นตัวเร่งปฏิกิริยายังแสดงให้เห็นว่าความสามารถในการเร่งปฏิกิริยาในปฏิกิริยาข้างเคียงอย่างการสลายตัวของเมทานอลไม่ดีเท่าที่ควรเมื่อเปรียบเทียบกับการนำตัวเร่งปฏิกิริยาไปใช้ในการเร่งปฏิกิริยาของการเปลี่ยนรูปเมทานอลโดยใช้ไอน้ำ

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