HYDROGEN PRODUCTION BY OXIDATIVE STEAM REFORMING OF METHANOL OVER Au-CuO/CeO_2 CATALYSTS

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ABSTRACT

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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, oxyhen, and methanol over 3% Au-CuO/CeO_2 catalyst was used. Among the catalysts tested, the 3%(5:1), (Cu/Au) Au-CuO/CeO_2 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/CeO_2 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 CeO_2 support. Additionally, in the UV-visible spectra showed revealed that 3% (5:1), (Cu/Au) Au-CuO/CeO_2 calcined at 200°C exhibited the biggest size of Au particle with highest Au metallic particle. Moreover, from TEM image, it was shoed that gold particle size is between 5 to 10 nm. Finally, 3%(5:1), (Cu/Au) Au-CuO/CeO_2 catalyst calcined at 200°C was used to catalyze DCM and SRM. The result showed that catalyst yield the low methanol conversison for DCM but suitable for SRM.

บทคัดย่อ

ภัทรชนน พิพัฒน์ประทานพร : กระบวนการผลิตก๊าซไฮโดรเจนจากปฏิกิริยาเปลี่ยนรูปเมทานอลด้วยไอน้ำและออกซิเจนโดยใช้ตัวเร่งปฏิกิริยาทองและคอปเปอร์ออกไซด์บนซีเรียออกไซด์ (Hydrogen Production from the Oxidative Steam Reforming of Methanol over Au/Fe_2O_3-CeO_2) อ. ที่ปรึกษา : ผศ.ดร. อาภาณี เหลืองนฤมิตชัย และ ศ.ดร. เออโดแกน กูลารี่ 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/CeO_2 เผาที่ 200 องศาเซลเซียสให้ผลลการเปลี่ยนแปลงเมทานอลและให้ค่าผลผลิตเท่ากับ 95% และ 59% ตามลำดับ ที่อุณหภูมิ 400 องศาเซลเซียส ซึ่งให้ค่าสูงที่สุด นอกจากนั้นทำการวิเคราะห์คุณลักษณะของตัวเร่งปฏิกิริยา โดยใช้เทคนิค XRD, TEM, และ TPR จากผลการวิเคราะห์โดยใช้ TPR พบว่าตัวเร่งปฏิกิริยา เมื่อเพิ่มปริมาณของคอปเปอร์ออกไซด์ในตัวเร่งปฏิกิริยานั้นจะเป็นการลดแรงกระทำระหว่างทองและตัวรองรอบซีเรียออกไซด์โดยการสร้างแรงกระทำระหว่างทองและคอปเปอร์ออกไซด์ซึ่งส่งผลต่ออุณหภูมิการรีดักชั่นของตัวเร่งปฏิกิริยา ในขณะเดียวกัน จากผลการวิเคราะห์ด้วย UV-visible พบว่า ขนาดของอนุภาคทองบนตัวเร่งปฏิกิรยาตัวดังกล่าวมีขนาดใหญ่ที่สุดและมีปริมาณทองศูนย์มากซึ่งว่องไวต่อปฏิกิริยาอยู่มากที่สุด จากการศึกษาโดยเครื่อง TEM พบว่าอนุภาคทองมีขนาด 5 ถึง 10 นาโนเมตร นอกจากนั้นตัวเร่งปฏิกิริยายังแสดงให้เห็นว่าความสามารถในการเร่งปฏิกิริยาในปฏิกิริยาข้างเคียงอย่างการสลายตัวของเมทานอลไม่ดีเท่าที่ควรเมื่อเปรียบเทียบกับการนำตัวเร่งปฏิกิรยาไปใช้ในการเร่งปฏิกิริยาของการเปลี่ยนรูปเมทานอลโดยใช้ไอน้ำ

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