

**OXIDATIVE STEAM REFORMING OF METHANOL OVER Au-BASED
CATALYSTS: EFFECT OF SUPPORT COMPOSITION AND
BIMETALLIC CATALYST**

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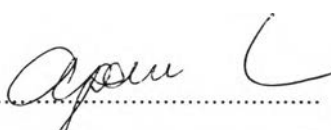
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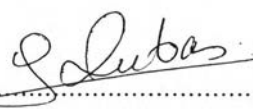
Thesis Title: Oxidative Steam Reforming of Methanol over Au-based Catalysts: Effect of Support Composition and Bimetallic Catalyst
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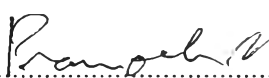
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ABSTRACT

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Achiraya Kumyam: Oxidative Steam Reforming of Methanol over Au-based Catalysts: Effect of Support Composition and Bimetallic Catalyst.

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The catalytic activity of bimetallic Au-Cu/ $\text{CeO}_2\text{-ZrO}_2$ catalysts was studied for oxidative steam reforming of methanol (OSRM). Various variables, such as support composition (Ce/(Ce+Zr) atomic ratio), support preparation, Au/Cu atomic ratio, total metal loading, and calcination temperature were investigated. The catalysts were characterized by using Brunauer-Emmett-Teller (BET) surface area measurement, X-ray diffraction (XRD), temperature-programmed reduction (TPR), atomic absorption spectroscopy (AAS), FT-Raman spectroscopy, UV-visible, and temperature-programmed oxidation (TPO). Catalytic activity of the prepared catalysts was investigated under atmospheric pressure at a reaction temperature ranging from 200 °C to 400 °C. The 5 wt% 3Au1Cu/ $\text{Ce}_{0.75}\text{Zr}_{0.25}\text{O}_2$ calcined at 300°C exhibited the highest catalytic activity with 99.6% methanol conversion and 62.4% hydrogen yield. Moreover, the 5 wt% 3Au1Cu/ $\text{Ce}_{0.75}\text{Zr}_{0.25}\text{O}_2$ showed stable catalytic performance at 350 °C for 21 h. This may be related to the Au-Cu alloy structure, as confirmed by XRD patterns.

บทคัดย่อ

อชิรญา คำเข้ม : กระบวนการผลิตก๊าซไฮโดรเจนจากปฏิกิริยาเปลี่ยนรูปเมทานอลด้วยไอน้ำและก๊าซออกซิเจนโดยใช้ตัวเร่งปฏิกิริยาทอง: ผลขององค์ประกอบของตัวรองรับและตัวเร่งปฏิกิริยาชนิดโลหะผสม (Oxidative Steam Reforming of Methanol over Au-based Catalysts: Effect of Support Composition and Bimetallic Catalyst) อ. ที่ปรึกษา: รศ. ดร. อาภาณี เหลืองนฤมิตชัย และ ดร. สเตฟาน ที. คูบาส 108 หน้า

งานวิจัยนี้ศึกษากระบวนการผลิตก๊าซไฮโดรเจนด้วยกระบวนการเปลี่ยนรูปเมทานอลด้วยไอน้ำและก๊าซออกซิเจน โดยใช้ตัวเร่งปฏิกิริยาทอง ที่เตรียมด้วยวิธีการยัดเกาะควบคู่กับการตกผลึก (deposition-precipitation) โดยตัวรองรับบริสุทธ์ (CeO_2 และ ZrO_2) และตัวรองรับผสม ($\text{CeO}_2\text{-ZrO}_2$) ถูกเตรียมด้วยวิธีการตกผลึกและการตกผลึกร่วมตามลำดับ สำหรับตัวแปรที่ศึกษากับตัวเร่งปฏิกิริยาทอง เช่น อัตราส่วนโดยโมลของตัวรองรับ (0, 0.25, 0.5, 0.75 และ 1), วิธีการเตรียมตัวรองรับ, อัตราส่วนของโลหะทองและคอปเปอร์ (3/1, 1/1 และ 1/3), ปริมาณของทองที่ใช้ในการเตรียมตัวเร่งปฏิกิริยา และอุณหภูมิที่ใช้ในการเผาตัวเร่งปฏิกิริยา (calcination temperature) ซึ่งศึกษาความว่องไวในการเกิดปฏิกิริยาในช่วงอุณหภูมิ 200 องศาเซลเซียส ถึง 400 องศาเซลเซียส ภายใต้ความดันบรรยากาศ ผลการศึกษาแสดงให้เห็นว่าตัวเร่งปฏิกิริยา 5 wt% $3\text{Au}1\text{Cu}/\text{Ce}_{0.75}\text{Zr}_{0.25}\text{O}_2$ เตรียมที่อุณหภูมิ 300 °C ให้ผลในการเกิดปฏิกิริยาสูงที่สุด โดยพบว่าการเปลี่ยนแปลงของเมทานอล (methanol conversion) มีค่าเป็นร้อยละ 99.6 และผลผลิตไฮโดรเจนร้อยละ (hydrogen yield) มีค่าเป็น 62.4 ยิ่งไปกว่านั้นยังพบว่าตัวเร่งปฏิกิริยา 5 wt% $3\text{Au}1\text{Cu}/\text{Ce}_{0.75}\text{Zr}_{0.25}\text{O}_2$ มีความเสถียรในการเกิดปฏิกิริยาสำหรับกระบวนการเปลี่ยนรูปเมทานอลด้วยไอน้ำและก๊าซออกซิเจนที่อุณหภูมิ 350 องศาเซลเซียส เป็นเวลา 21 ชั่วโมง ทั้งนี้อาจเกี่ยวเนื่องมาจากการเกิดอัลลอยระหว่างโลหะทองและคอปเปอร์

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