

**HYDROGEN PRODUCTION FROM THE OXIDATIVE  
STEAM REFORMING OF METHANOL OVER Au/CeO<sub>2</sub> CATALYSTS**



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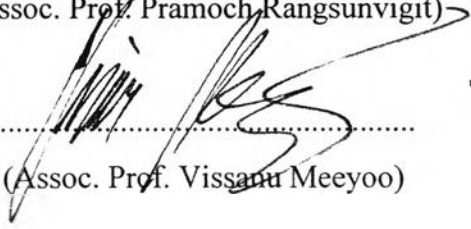
  
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**ABSTRACT**

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The production of hydrogen by the oxidative steam reforming of methanol (OSRM) was investigated on a series of Au/CeO<sub>2</sub> catalysts prepared by deposition-precipitation. The influences of the main parameters considered on the methanol conversion are the H<sub>2</sub>O/CH<sub>3</sub>OH and O<sub>2</sub>/CH<sub>3</sub>OH feed molar ratios, content of Au loading, calcination temperature, and operating reaction temperature. Among all the samples studied, 1%wt Au/CeO<sub>2</sub> exhibited nearly a 100 % methanol conversion and 23.63 % H<sub>2</sub> yield at 300 °C. Optimum operating conditions—GHSV = 30, 000 ml/h-g<sub>cat</sub>, T = 300 °C, H<sub>2</sub>O/CH<sub>3</sub>OH molar ratio = 2/1, and O<sub>2</sub>/CH<sub>3</sub>OH molar ratio = 1.25/1—could be suggested to obtain the high methanol conversion and hydrogen yield. Interestingly, 5 %wt Au/CeO<sub>2</sub> exhibited the highest activity under the optimum conditions with 100 % methanol conversion and 24.5 % H<sub>2</sub> yield since larger Au particle sizes might be more active in OSRM without metal sintering during the reaction. In the stability test, methanol conversion dropped rapidly from 100 % to 88.8 % after 40 h due to a blocking of pores by coke formation; whereas an average H<sub>2</sub> yield at 16.12 % was still steady.

## บทคัดย่อ

จินชนป พจนาวราพันธ์ : กระบวนการผลิตก๊าซไฮโดรเจนจากปฏิกิริยาเปลี่ยนรูปเมทานอลด้วยไอน้ำและก๊าซออกซิเจน โดยใช้ตัวเร่งปฏิกิริยาทองบนซีเรียออกไซด์ (Hydrogen Production from the Oxidative Steam Reforming of Methanol over Au/CeO<sub>2</sub> Catalysts) อ. ที่ปรึกษา : ผศ.ดร. อาภาณี เหลืองนฤมิตชัย ศ.ดร. เออโดแกน กุลารี่

งานวิจัยนี้ศึกษากระบวนการผลิตก๊าซไฮโดรเจนด้วยกระบวนการเปลี่ยนรูปเมทานอลด้วยไอน้ำและก๊าซออกซิเจน โดยใช้ตัวเร่งปฏิกิริยาทองบนตัวรองรับชนิดซีเรียออกไซด์ ที่เตรียมด้วยวิธีการยัดเกาะควบคู่กับการตกผลึก (Deposition-precipitation) ตัวแปรที่ศึกษาที่มีอิทธิพลต่อการเปลี่ยนแปลงของเมทานอล (methanol conversion) เช่น อัตราส่วนโดยโมลของ H<sub>2</sub>O/CH<sub>3</sub>OH และ O<sub>2</sub>/CH<sub>3</sub>OH ปริมาณของทองที่ใช้ในการเตรียมตัวเร่งปฏิกิริยา รวมถึงอุณหภูมิที่ใช้ในการเตรียมตัวเร่งปฏิกิริยา (calcination temperature) และช่วงของอุณหภูมิที่ใช้ในการเกิดปฏิกิริยาในเตาปฏิกรณ์ขนาดเล็ก ผลการศึกษาในสถานะเริ่มต้นพบว่าตัวเร่งปฏิกิริยา 1%wt Au/CeO<sub>2</sub> ให้ผลร้อยละการเปลี่ยนแปลงเมทานอล 100 และร้อยละผลผลิตของไฮโดรเจน 23.63 ที่อุณหภูมิ 300 องศาเซลเซียส ณ สภาวะอัตราส่วนโดยโมลของ H<sub>2</sub>O/CH<sub>3</sub>OH เท่ากับ 2/1 และ O<sub>2</sub>/CH<sub>3</sub>OH เท่ากับ 1.25/1 ซึ่งเป็นสภาวะที่เหมาะสมที่สุดต่อการเกิดปฏิกิริยาดังกล่าว เพื่อให้การเปลี่ยนแปลงเมทานอลและค่าผลิตผลของไฮโดรเจนมากที่สุด อย่างไรก็ตามในส่วนของการศึกษาผลของตัวเร่งปฏิกิริยาพบว่า 5%wt Au/CeO<sub>2</sub> ให้ร้อยละการเปลี่ยนแปลงเมทานอลและผลิตผลของไฮโดรเจนสูงสุดเท่ากับ 100 และ 24.5 ตามลำดับ ในสภาวะการเกิดปฏิกิริยาเดียวกัน เนื่องจากมีขนาดอนุภาคของโลหะทองที่ใหญ่ที่สุด ส่งผลให้มีความว่องไวต่อการเกิดปฏิกิริยาสูงสุด โดยปราศจากภาวะการรวมตัวเป็นกลุ่มก้อน (sintering) นอกจากนี้ในการทดสอบความเสื่อมสภาพของตัวเร่งปฏิกิริยา พบว่าร้อยละการเปลี่ยนแปลงของเมทานอลลดลงอย่างรวดเร็วจาก 100 เป็น 88.8 หลังจากทำการทดสอบเป็นเวลา 40 ชั่วโมง สาเหตุที่เป็นเช่นนั้นมาจากการที่โค้ก (coke) ไปอุดรูพรุน (blocking) ที่ใช้ในการเกิดปฏิกิริยาของตัวเร่งปฏิกิริยา อย่างไรก็ตามค่าของร้อยละผลิตผลไฮโดรเจนยังคงมีค่าเฉลี่ยอยู่ที่ 16.12 ตลอดช่วงเวลาที่ทำการทดสอบการเสื่อมสภาพ

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