HYDROGEN PRODUCTION FROM OXIDATIVE STEAM REFORMING OF METHANOL OVER Au/CeO₂-ZrO₂ CATALYSTS

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ABSTRACT

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Hydrogen production by oxidative steam reforming of methanol (OSRM) was investigated over Au/CeO₂–ZrO₂ catalysts prepared by a depositionprecipitation (DP) technique. Pure supports (CeO₂ and ZrO₂) and mixed supports (CeO₂–ZrO₂) were prepared by precipitation and co-precipitation techniques, respectively. A series of Au supported on CeO₂–ZrO₂ with various atomic ratios of Ce/(Ce+Zr) (0, 0.25, 0.5, 0.75 and 1) were studied. The activity was investigated in the range of 200 °C to 400 °C under atmospheric pressure. The 3 wt%Au/Ce_{0.75}Zr_{0.25}O₂ calcined at 400 °C exhibited the highest catalytic activity with 91.28% methanol conversion and 61.50% hydrogen yield at an O₂/H₂O/CH₃OH molar ratio of 0.6/2/1. In addition, it was observed that the 3 wt% Au/Ce_{0.75}Zr_{0.25}O₂ catalyst showed stable activities for OSRM at 350 °C for 12 hours.

บทคัดย่อ

วราพรรณ นครานุวัฒนะ : กระบวนการผลิตก๊าซไฮโดรเจนจากปฏิกิริยาเปลี่ยนรูปมทา นอลด้วยไอน้ำและก๊าซออกซิเจนโดยใช้ตัวเร่งปฏิกิริยาทองบนซีเรียออกไซด์และเซอร์โคเนีย ออกไซด์(Hydrogen Production from Oxidative Steam Reforming of Methanol over Au/CeO₂–ZrO₂ Catalysts) อ. ที่ปรึกษา : รศ. ดร. อาภาณี เหลืองนฤมิตชัย และ ศ. ดร. เออร์โด แกน กูลารี่ 122 หน้า

งานวิจัยนี้ศึกษากระบวนการผลิตก๊าซไฮโครเจนด้วยกระบวนการเปลี่ยนรูปเมทานอล ด้วยไอน้ำและก๊าซออกซิเจน โคยใช้ตัวเร่งปฏิกิริยาทองบนตัวรองรับชนิดซีเรียออกไซค์และ เซอร์โคเนียออกไซด์ ที่เตรียมด้วยวิธีการยึดเกาะควบค่กับการตกผลึก (depositionprecipitation) โดยตัวรับรองบริสุทธิ์ (CeO₂ และ ZrO_2) และตัวรองรับผสม (CeO₂- ZrO_2) ถูก เตรียมด้วยวิธีการตกผลึกและการตกผลึกร่วมตามลำดับ สำหรับตัวแปรที่ศึกษากับตัวเร่งปฏิกิริยา ทองบนซีเรียออกไซด์และเซอร์โคเนียออกไซด์ เช่น อัตราส่วนโดยโมลของตัวรองรับ (0, 0.25, 0.5, 0.75 และ 1) อุณหภูมิที่ใช้ในการเผาตัวเร่งปฏิกิริยา (calcination temperature) และ ปริมาณของทองที่ใช้ในการเตรียมตัวเร่งปฏิกิริยา ซึ่งศึกษาความว่องไวในการเกิดปฏิกิริยาในช่วง อุณหภูมิ 200 องศาเซลเซียส ถึง 400 องศาเซลเซียส ภายใต้ความดันบรรยากาศ ผลการศึกษา แสดงให้เห็นว่าตัวเร่งปฏิกิริยา 3 wt%Au/CeO₂–ZrO₂ เตรียมที่อุณหภูมิ 400 °C ให้ผลในการ เกิดปฏิกิริยาสูงที่สุด โดยพบว่าการเปลี่ยนแปลงของเมทานอล (methanol conversion) มีค่าเป็น ร้อยละ 91.28 และผลผลิตไฮโครเจนร้อยละ (hydrogen yield) มีค่าเป็น 61.50 ยิ่งไปกว่านั้น อัตราส่วนโดยโมลของO2/H2O/CH3OH ที่ 0.6/2/1 เป็นสภาวะที่เหมาะสมที่สุดต่อการเกิดปฏิกิริยา ดังกล่าว นอกจากนี้ยังพบว่าตัวเร่งปฏิกิริยา 3 wt%Au/CeO₂–ZrO₂ มีความเสถียรในการ เกิดปฏิกิริยาสำหรับกระบวนการเปลี่ยนรูปเมทานอลด้วยไอน้ำและก๊าซออกซิเจนที่อุณหภูมิ 350 องศาเซลเซียส เป็นเวลา 12 ชั่วโมง

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