# OXIDATIVE BROMINATION OF METHANE BY BARIUM OXIDE OR TUNGSTEN OXIDE ON SILICA CATALYSTS: EFFECT OF CATALYSTS PREPARATION

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#### ABSTRACT

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Oxidative bromination of methane (OBM) has been proposed for methane activation. In this work, methane was brominated by HBr/H<sub>2</sub>O solution with oxygen as an oxidizing agent to produce methyl bromide. Barium oxide or tungsten oxide on silica were chosen as the catalysts to improve the activity and selectivity of the reaction. The catalysts were prepared by 2 techniques, incipient wetness impregnation and sol-gel method. The effects of catalyst preparations on oxidative bromination of methane were investigated. The reaction was carried out in a fixed-bed continuous-flow reactor at atmospheric pressure. The catalysts were characterized by BET and XRD techniques. The results showed that, under the same condition (20 ml/min of CH<sub>4</sub>, 5 ml/min of O<sub>2</sub>, 5 ml/min of N<sub>2</sub>, 6.5 ml/h of 48 wt% HBr/H<sub>2</sub>O and temperature at 660 °C), the sol-gel catalysts exhibited higher methane conversion and methyl bromide selectivity than impregnated catalysts.

## บทคัดย่อ

กรณ์ สมจิตร : ออกซิเคทีฟโบรมิเนชันของมีเทนโคยใช้ตัวเร่งปฏิกิริยาแบเรียมออกไซต์ หรือทั้งสเตนออกไซต์บนตัวรองรับซิลิกา : ผลของการเตรียมตัวเร่งปฏิกิริยา (Oxidative Bromination of Methane by Barium Oxide or Tungsten Oxide on Silica Catalysts: Effect of Catalysts Preparation) อาจารย์ที่ปรึกษา : ผศ. คร. บุนยรัชต์ กิติยานันท์ 71 หน้า

ปฏิกิริยาออกซิเดทีฟโบรมิเนชันเป็นปฏิกิริยาสำหรับกระตุ้นความว่องไวของมีเทน ใน งานวิจัยนี้มีเทนจะถูก โบรมิเนทโดยสารละลายกรดไฮโดร โบรมิกและมีออกซิเจนเป็นตัวกระตุ้น ให้เกิดปฏิกิริยาออกซิเดชันเพื่อผลิตเมทิลโบรไมด์ โดยได้ใช้แบเรียมออกไซต์หรือทังสเตนออก ไซต์ บนตัวรองรับซิลิกาเป็นตัวเร่งปฏิกิริยาเพื่อปรับปรุงการเลือกเกิดของเมทิลโบรไมด์ ซึ่งตัวเร่ง ปฏิกิริยาจะเตรียมด้วยวิธีที่แตกต่างกัน 2 วิธี คือ วิธีการเอิบชุ่ม และ วิธีการโซลเจล โดยผลของการ เตรียมตัวเร่งต่อปฏิกิริยาออกซิเดทีฟโบรมิเนชันของมีเทนจะถูกศึกษา สารตั้งต้นทั้งหมดจะถูก ไปอนเข้าสู่เตาปฏิกรณ์แบบต่อเนื่องและทำปฏิกิริยาที่ความดันบรรยากาศ ตัวเร่งปฏิกิริยาจะถูก วิเคราะห์ด้วยเทคนิกต่างๆ เพื่อหาพื้นที่ผิวและความเป็นผลึก จากผลการทดลองพบว่าที่สภาวะการ ทดลองเดียวกัน (20 มิลลิลิตรต่อนาทีของมีเทน, 5 มิลลิลิตรต่อนาทีของออกซิเจน, 5 มิลลิลิตรต่อ นาทีของไนโตรเจน และ6.5 มิลลิลิตรต่อชั่วโมงของกรดไฮโครโบรมิก และ อุณหภูมิการทำ ปฏิกิริยาเท่ากับ 660 องศาเซลเซียส) ตัวเร่งปฏิกิริยาที่เตรียมด้วยวิธีโซลเจลจะให้ก่าการแปรผัน ของมีเทนและค่าการเลือกเกิดของเมทิลโบรไมด์สูงกว่าตัวเร่งปฏิกิริยาฏิกิริยาจากวิธีการเอิบชุ่ม ซึ่งอาจจะเป็นเหตุผลมาจากโลหะออกไซต์ของตัวเร่งปฏิกิริยาแบบโซลเจลมีขนาดเล็กว่าตัวเร่ง ปฏิกิริยาแบบเอิบชุ่ม

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