

**HYDROGEN PRODUCTION VIA METHANE PARTIAL OXIDATION
OVER CERIA-NICKEL AND CERIA-ZIRCONIA-NICKEL
MIXED-OXIDE CATALYSTS**



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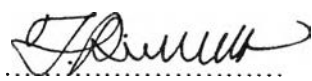
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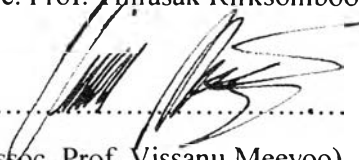
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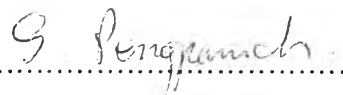
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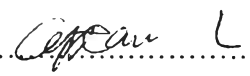

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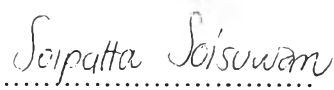
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ABSTRACT

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Mr. Adinun Khamnetr: Hydrogen Production via Methane Partial Oxidation over Ceria-Nickel and Ceria-Zirconia-Nickel Mixed-Oxide Catalysts

Thesis Advisors: Assoc. Prof. Thirasak Rirksomboon, Assoc. Prof. Vissanu Meeyoo, Dr. Sitthipong Pengpanich

Keywords: Hydrogen production/ Methane/ Partial oxidation/ Ni/ Ceria/ Zirconia/ Carbon deposition

Hydrogen production via the partial oxidation of light hydrocarbons has received much attention in recent years due to the high demand for clean energy sources alleviating environmental problems. An alternative means of producing hydrogen from methane partial oxidation (MPO) over different types of catalysts has been investigated by a number of research groups. However, carbon deposition on a catalyst bed, resulting in deteriorating its kinetics, is observed as a major cause of catalyst deactivation. In order to find alternative catalyst that can be more tolerant to carbon deposition, $(\text{Ce}_{0.75}\text{Zr}_{0.25})_{2.14}\text{Ni}_{0.86}\text{O}_3$ and $\text{Ce}_{2.14}\text{Ni}_{0.86}\text{O}_3$ mixed oxide catalysts were investigated for MPO over the temperature range of 400–800°C at atmospheric pressure. The catalysts were prepared by the co-impregnation method and characterized by X-ray diffraction (XRD) analysis, BET surface area measurement, H_2 chemisorption, temperature programmed reduction (TPR) and oxidation (TPO), and transmission electron microscopy (TEM). The XRD results showed that the reduction of the oxides proceeds through the formation of intermediate species to obtain Ni^0 and CeO_2 . Under the reaction conditions, both $(\text{Ce}_{0.75}\text{Zr}_{0.25})_{2.14}\text{Ni}_{0.86}\text{O}_3$ and $\text{Ce}_{2.14}\text{Ni}_{0.86}\text{O}_3$ catalysts were active at temperatures above 650°C. Among the catalysts tested, the $(\text{Ce}_{0.75}\text{Zr}_{0.25})_{2.14}\text{Ni}_{0.86}\text{O}_3$ catalyst calcined at 700°C exhibited better catalytic activity and stability for methane partial oxidation.

บทคัดย่อ

อดิพนธ์ คำเนตร : การผลิตไฮโดรเจนจากปฏิกิริยาออกซิเดชันบางส่วนของมีเทนโดยใช้ตัวเร่งปฏิกิริยานิกเกิลบนซีเรียและนิกเกิลบนซีเรีย-เซอร์โคเนีย (Hydrogen Production via Methane Partial Oxidation over Ceria-Nickel and Ceria-Zirconia-Nickel Mixed-Oxide Catalysts)

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กระบวนการผลิตไฮโดรเจนจากปฏิกิริยาออกซิเดชันบางส่วนของสารไฮโดรคาร์บอนเบาได้รับความนิยมอย่างมากในปัจจุบันเนื่องจากมีความต้องการพลังงานสะอาดที่สามารถลดปัญหามลพิษที่ปล่อยสู่สิ่งแวดล้อม กระบวนการผลิตไฮโดรเจนจากปฏิกิริยาออกซิเดชันบางส่วนของมีเทนโดยใช้ตัวเร่งปฏิกิริยาได้รับความนิยมศึกษาและพัฒนาจากหลากหลายกลุ่มงานวิจัย อย่างไรก็ตามปัญหาสำคัญของกระบวนการนี้คือ เกิดการสะสมของคาร์บอนบนตัวเร่งปฏิกิริยาซึ่งเป็นผลทำให้ความสามารถในการทำปฏิกิริยาของตัวเร่งปฏิกิริยาลดลง ดังนั้นในการวิจัยจึงทำการศึกษาและปรับปรุงตัวเร่งปฏิกิริยาที่สามารถลดการสะสมของคาร์บอนเพื่อใช้ในการผลิตไฮโดรเจนจากปฏิกิริยาออกซิเดชันบางส่วนของมีเทนโดยใช้ตัวเร่งปฏิกิริยานิกเกิลบนซีเรียและนิกเกิลบนซีเรีย-เซอร์โคเนียในช่วงอุณหภูมิ 400 ถึง 800 องศาเซลเซียส ที่ความดันบรรยากาศ โดยใช้วิธีตกตะกอนร่วม (co-impregnation method) เตรียมตัวเร่งปฏิกิริยาและศึกษาลักษณะเฉพาะ (characterization) ของตัวเร่งปฏิกิริยาโดยวิธี XRD, BET, H₂ chemisorption, TPR, TPO และ TEM จากการศึกษาพบว่าแก๊สไฮโดรเจนและคาร์บอนมอนอกไซด์เกิดที่อุณหภูมิ 650 องศาเซลเซียสขึ้นไป อีกทั้งพบว่าตัวเร่งปฏิกิริยานิกเกิลบนซีเรีย-เซอร์โคเนียเผา (calcination) ที่อุณหภูมิ 700 องศาเซลเซียสแสดงกัมมันตภาพ (activity) และเสถียรภาพ (stability) ได้ดีในปฏิกิริยาออกซิเดชันบางส่วนของมีเทน

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