CO OXIDATION ON CeO_2-ZrO_2 SOLID SOLUTION PREPARED BY SOL-GEL TECHNIQUE UNDER OXIDIZING AND REDUCING CONDITIONS

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บทคัดย่อ

นางสาวมทินา ธรรมชาติ : การศึกษาปฏิกิริยาออกซิเดชันของก๊าซคาร์บอนมอนอกไซด์โดยใช้ออกไซด์ผสมระหว่างซีเรียกับเซอร์โคเนียที่เตรียมโดยวิธีโซล-เจลเป็นตัวเร่งปฏิกิริยาภายใต้สภาวะออกซิไดซ์และรีดิวซ์ (CO Oxidation on CeO_2-ZrO_2 Solid Solution Prepared by Sol-Gel Technique under Oxidizing and Reducing Conditions) อ. ที่ปรึกษา : ศ. ดร. โจฮานส์ ชแวงค์ ดร. วิษณุ มีอยู่ และ ผศ. ดร. ธีรศักดิ์ ฤกษ์สมบูรณ์ 83 หน้า ISBN 974-334-132-3

การศึกษาผลของการใช้ออกไซด์ผสมระหว่างซีเรียกับเซอร์โคเนียในตัวเร่งปฏิกิริยาสามทางเพื่อลดมลพิษในไอเสียจากเครื่องยนต์เป็นที่น่าสนใจมากในช่วงไม่กีปีที่ผ่านมา การวิจัยนี้เป็นการศึกษาผลของการใช้ออกไซด์ผสมระหว่างซีเรียกับเซอร์โคเนียที่เตรียมโดยผ่านปฏิกิริยายูเรียไฮโดรไลซิสต่อปฏิกิริยาออกซิเดชันของก๊าซคาร์บอนมอนอกไซด์ภายใต้สภาวะออกซิไดซ์และรีดิวซ์ โดยแปรปริมาณของเซอร์โคเนียตั้งแต่ 25 ถึง 100 โมลเปอร์เซ็นต์ การใช้วิธีการโซล-เจลในการเตรียมตัวเร่งปฏิกิริยาสามารถควบคุมให้เกิดอนุภาคที่มีขนาดสม่ำเสมอและมีพื้นที่ผิวของอนุภาคสูงกว่าเมื่อเตรียมโดยวิธีเก่า นอกจากนี้เมื่อนำตัวเร่งปฏิกิริยาที่เตรียมได้มาทดสอบดูลักษณะของอนุภาค พบว่าอนุภาคที่เกิดมีขนาดที่อยู่ในช่วงนาโนมิเตอร์ซึ่งมีขนาดเล็กมาก สิ่งที่น่าสนใจคือผลที่ได้จากเครื่องเอ็กซเรย์ดิฟแฟรกชัน และ เครื่องรามานสเป็กโตรสโคปี แสดงให้เห็นว่าในแลตติซท์ของซีเรีย เกิดการแทนที่ของอนุภาคซีเรียบางส่วนด้วยอนุภาคเซอร์โคเนียได้แม้จะเตรียมที่อุณหภูมิต่ำคือที่ 100 องศาเซลเซียส และเมื่อทำการวิเคราะห์โดยเครื่องเทมเพอเรเจอร์โปรแกรมรีดักชัน พบว่าการเติมเซอร์โคเนียเข้าไปในแลททิตซ์ของซีเรียจะช่วยสนับสนุนการเกิดบัลค์รีดักชันทำให้อุณหภูมิในการรีดักชันลดลงมากเมื่อเทียบกับซีเรีย และจากปฏิกิริยาออกซิเดชันของก๊าซคาร์บอนมอนอกไซด์พบว่าสามารถเกิดปฏิกิริยาไปเป็นก๊าซคาร์บอนไดอกไซด์ได้สูงขึ้น

ABSTRACT

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The promotive effects of ceria (CeO_2)/zirconia (ZrO_2) mixed oxides on commercial three-way catalysts (TWC) for purification of motor exhaust gases have been investigated in recent years. This work presented the cooperative effects of CeO_2-ZrO_2 mixed oxides prepared by urea hydrolysis on the CO conversion under oxidizing and reducing conditions. The ZrO_2 contents investigated were varied between 25 and 100 %mol. Noteworthy is that the versatility of the sol-gel technique allows for control of the composition, homogeneity particles and higher BET surface areas compared to conventional methods. The homogeneous nanometer-sized particles were observed using a scanning electron microscopy (SEM). From the results of an X-ray diffraction (XRD) and FT-Raman spectroscopy, the solid solutions were observed even though the catalysts were prepared at a low temperature (ca 100°C). Temperature-programmed reduction (TPR) in a H_2/N_2 mixture of the solid solutions revealed that the incorporation of ZrO_2 into CeO_2 lattice strongly promoted bulk reduction of the solid solutions, resulting in a CO conversion at low temperatures. It has been proposed that CeO_2-ZrO_2 mixed oxides would be one of the best supports for the TWC applications.

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LIST OF SYMBOLS

W	Weight of gas adsorbed at relative pressure P_o
W_m	Weight of adsorbate constituting a monolayer of surface coverage
C	Constant related to the energy of adsorption in the first adsorbed layer and magnitude of adsorbate/adsorbent interaction
$A_{nitrogen}$	Cross-sectional area of one molecule nitrogen = 0.162 nm^2 (at 77 K)
$M_{w,\ nitrogen}$	Molecular weight of nitrogen (28)
λ	Wave length of the monochromatic X-ray diffraction (Å)
K	Scherrer constant depended on the shape of the particle
B_d	Angular width of the peak in the terms of $\Delta(2\theta)$ (radian)
θ	Glancing angle (degree)
d_b	Mean crystallite diameter (Å)