

**ADIPONITRILE HYDROGENATION OVER NICKEL SUPPORTED
ON CERIA-ZIRCONIA AND CERIA-ZIRCONIA-MAGNESIA
MIXED OXIDE CATALYSTS**

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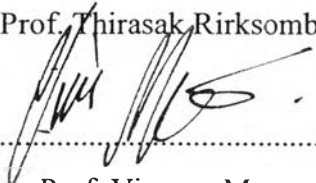
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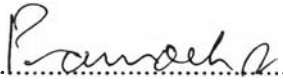
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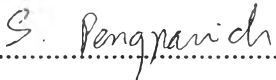

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ABSTRACT

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The gas phase hydrogenation of adiponitrile (ADN) to 1,6-hexamethylenediamine (HMDA) over nickel supported on ceria-zirconia and ceria-zirconia-magnesia mixed oxide catalysts was studied at atmospheric pressure. The catalysts, $\text{Ni/Ce}_{0.75}\text{Zr}_{0.25}\text{O}_2$ and $\text{Ni/Ce}_{0.75}\text{Zr}_{0.15}\text{Mg}_{0.20}\text{O}_2$, were prepared via coprecipitation, followed by 15 %wt. Ni impregnation. The catalysts were characterized by several techniques, i.e., N_2 -physisorption, H_2 -TPR, XRD, and NH_3 -TPD. The reaction was carried out using a continuous flow fixed-bed reactor by varying temperature, hydrogen-to-adiponitrile mol ratio (H_2/ADN), gas hourly space velocity (GHSV), and acidity of the supports. The main product produced via partial hydrogenation was observed to be 6-aminohexanenitrile. The catalyst with less support acidity can enhance the production of HMDA.

บทคัดย่อ

กัณฑ์พงศ์ มงคลหัตถี : ปฏิริยาไฮโดรจิเนชันของอะไดโพไนไตรล์โดยใช้ตัวเร่งปฏิริยานิกเกิลบนตัวรองรับออกไซด์ผสมของซีเรียเซอร์โคเนียและของซีเรียเซอร์โคเนียแมกนีเซียม (Adiponitrile Hydrogenation over Nickel Supported on Ceria-Zirconia and Ceria-Zirconia-Magnesia Mixed Oxide Catalysts) อ. ที่ปริกษา : - รศ.ดร. ชีรศักดิ์ ฤกษ์สมบูรณ์ และ รศ.ดร. วิษณุ มีอยู่ 48 หน้า

การศึกษาปฏิริยาไฮโดรจิเนชันของอะไดโพไนไตรล์ในสถานะก๊าซโดยใช้ตัวเร่งปฏิริยานิกเกิลบนตัวรองรับออกไซด์ผสมของซีเรียเซอร์โคเนียและของซีเรียเซอร์โคเนียแมกนีเซียมที่ความดันบรรยากาศ ตัวเร่งปฏิริยาเตรียมถูกโดยวิธีการตกตะกอนร่วมของตัวรองรับตามด้วยการเติมนิกเกิลลงบนตัวรองรับโดยวิธีการทำให้ชุ่ม โดยปริมาณโลหะนิกเกิลคงที่ร้อยละ 15 ของน้ำหนักตัวเร่งปฏิริยา จากนั้นได้ศึกษาคุณลักษณะสมบัติของตัวเร่งปฏิริยาที่เตรียมได้โดยวิธี XRF, BET surface area, H₂-TPR, XRD, และ NH₃-TPD รวมทั้งศึกษาถึงสภาวะต่างๆ ที่มีผลต่อความสามารถในการเกิดปฏิริยา ทั้งอุณหภูมิ อัตราส่วนโดยโมลระหว่างไฮโดรเจนต่ออะไดโพไนไตรล์ ความเร็วในการทำปฏิริยา และความเป็นกรดของตัวเร่งปฏิริยา ซึ่งจากการทดลองพบว่าผลิตภัณฑ์หลักของปฏิริยา คือ AHN ซึ่งเกิดจากการไฮโดรจิเนชันเพียงบางส่วนของอะไดโพไนไตรล์ และสภาวะทุกสภาวะมีผลต่อ ADN conversion โดย ADN conversion มีผลอย่างมากต่อปริมาณของผลิตภัณฑ์ที่เกิดขึ้น สำหรับตัวเร่งปฏิริยาที่มีการเติมแมกนีเซียม พบว่าความเป็นกรดของตัวเร่งปฏิริยาลดลง ซึ่งสามารถยับยั้งการเกิดปฏิริยาการกำจัดแอมโมเนีย จึงทำให้สามารถผลิต HMDA ซึ่งเป็นผลิตภัณฑ์ที่ต้องการได้มากขึ้น

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