

**ETHYLENE EPOXIDATION OVER AG CATALYSTS: EFFECTS OF
SUPPORT, SECOND METAL, PROMOTER, AND DILUENT GAS**

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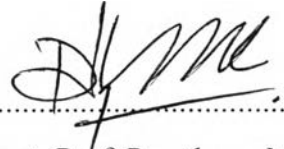
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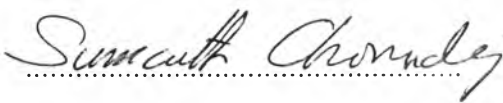
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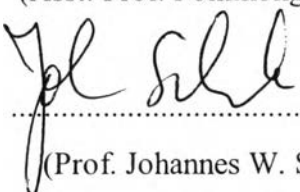
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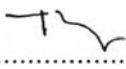
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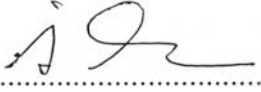

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ABSTRACT

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The ethylene epoxidation reaction was conducted in a packed-bed tubular reactor to produce ethylene oxide in this work. The effects of oxide supports (α -Al₂O₃, Al₂O₃ C, Al₂O_{3,Acid}, SiO₂ 90, TiO₂, SrTiO₃, MgTiO₃, CaTiO₃, and BaTiO₃), second metals (Au, Cu, Ba, Pd and Sn), and the diluent gases in the reactant feed (He, Ar, N₂, CH₄, and CH₄ balanced with He) were investigated in order to find the optimum reaction conditions for the epoxidation of ethylene, leading to an improvement of the catalytic activity in terms of both EO selectivity and EO yield. Among the investigated supports, SrTiO₃ was found to be the best support, relating to Sr atoms that are incorporated in the SrTiO₃ support that made this support more selective in EO formation. The most effective catalyst was the bimetallic 1.39 wt.% Cu-17.16 wt.% Ag/SrTiO₃ catalyst with 0.32 wt.% Sn promoter. The superior performance of this catalyst might be due to its high oxygen and ethylene uptakes. Moreover, the tin promoter enhanced the long-term stability of the catalyst, resulting from the decrease in the rate of carbonaceous species formation. Under the optimum experimental conditions, the EO selectivity was found to be extremely high up to 99.5 % at 6 h and could maintain at 96 % after 7 days of time on stream, together with the maximum EO yield of 5.5 % at 6 h and 4.9 % at 7 days of time on stream.

บทคัดย่อ

อชิพร จงเทิดทูนสกุล : ปฏิกริยาอีพอกซิเดชันของเอทิลีนบนตัวเร่งปฏิกริยาโลหะเงิน: ผลของวัสดุรองรับ, โลหะที่สอง, ตัวเสริม และ ก๊าซเจือจาง (Ethylene Epoxidation over Ag Catalysts: Effects of Support, Second Metal, Promoter, and Diluent Gas)
 อ. ที่ปริกษา : ศ. ดร. สุเมธ ชวเดช และ ศ. ดร. โจฮานเนส ดับบลิว ชวังก์ 169 หน้า

ในงานวิจัยนี้ สารเอทิลีนออกไซด์ (EO) ถูกผลิตโดยปฏิกริยาอีพอกซิเดชันของเอทิลีนในระบบถังปฏิกริการแบบแพคเบด ผลกระทบของวัสดุรองรับออกไซด์ (แอลฟาอลูมินา, อลูมินาซี. อลูมินาเอไซด์, ซิลิกาแก้วสับ, ไททานีย, สตรอนเทียมไททานต, แมกนีเซียมไททานต, แคลเซียมไททานต และ แบเรียมไททานต), ผลของโลหะที่สอง (ทอง, ทองแดง, แบเรียม, แพลเลเดียม และ ดีบุก) และ ผลของก๊าซเจือจาง (ฮีเลียม, อาร์กอน, ไนโตรเจน, มีเทน และ มีเทนผสมกับฮีเลียม) ได้ถูกทำการศึกษาเพื่อหาสภาวะทำปฏิกริยาที่ดีที่สุดสำหรับการผลิตสารเอทิลีนออกไซด์เพื่อนำไปสู่การปรับปรุงประสิทธิภาพการเลือกเกิด (Selectivity) และประสิทธิผล (Yield) ของ EO จากการศึกษา พบว่าสตรอนเทียมไททานตเป็นวัสดุรองรับที่ดีที่สุด เป็นผลมาจากอะตอมของสตรอนเทียมซึ่งอยู่ในวัสดุรองรับทำให้เกิดสารเอทิลีนออกไซด์ได้มากกว่า นอกจากนั้น ตัวเร่งปฏิกริยาที่มีประสิทธิภาพที่สุดคือ ตัวเร่งปฏิกริยา 1.39 เปอร์เซ็นต์โดยน้ำหนักทองแดง-17.16 เปอร์เซ็นต์โดยน้ำหนักโลหะเงิน บนวัสดุรองรับสตรอนเทียมไททานต โดยเติม 0.32 เปอร์เซ็นต์โดยน้ำหนักดีบุก เป็นตัวเสริม (Promoter) การดูดซับของออกซิเจนและเอทิลีนที่สูงของตัวเร่งปฏิกริยานี้ส่งผลต่อความสามารถที่เหนือกว่า อีกทั้งยังพบว่าดีบุกเป็นตัวเสริมที่ช่วยทำให้ความเสถียร ในการทำปฏิกริยาของตัวเร่งปฏิกริยาให้ยาวนานขึ้น โดยลดอัตราการเกิดสารประกอบคาร์บอน ภายใต้สภาวะการทำงานที่เหมาะสมที่สุด ประสิทธิภาพการเลือกเกิดของสารเอทิลีนออกไซด์ (EO selectivity) เท่ากับ 99.5 เปอร์เซ็นต์ ที่ 6 ชม. และ 96 เปอร์เซ็นต์ หลังจากทำการทดลองเป็นเวลา 7 วัน พร้อมกับ ประสิทธิภาพ (EO yield) สูงสุดเท่ากับ 5.5 เปอร์เซ็นต์ ที่ 6 ชม. และ 4.9 เปอร์เซ็นต์ หลังจากทำการทดลองเป็นเวลา 7 วัน

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