

**EPOXIDATION OF ETHYLENE OVER SILVER CATALYSTS  
IN LOW-TEMPERATURE CORONA DISCHARGE**

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**ABSTRACT**

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The production of ethylene oxide via partial ethylene oxidation, so-called ethylene epoxidation, is currently indispensable for obtaining valuable chemical feedstock or intermediate to be used for manufacturing several kinds of chemicals, such as solvents, adhesive, surfactant, and foam polyurethane. In this study, the epoxidation of ethylene in a low-temperature corona discharge system in the presence of different catalysts, namely Ag/(low-surface-area, LSA) $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, Ag/(high-surface-area, HSA) $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, Au-Ag/(HSA) $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, and Au/TiO<sub>2</sub>, was studied. In a comparison among the studied catalysts, Ag/(LSA) $\alpha$ -Al<sub>2</sub>O<sub>3</sub> catalyst was found to offer the highest selectivity of ethylene oxide, as well as the lowest selectivities of carbon dioxide and carbon monoxide. The selectivity of ethylene oxide increased with increasing applied voltage, while the selectivity of ethylene oxide remained unchanged when the frequency was varied in the range of 300-500 Hz. Nevertheless, the selectivity of ethylene oxide decreased with increasing frequency beyond 500 Hz. The optimum Ag loading on (LSA) $\alpha$ -Al<sub>2</sub>O<sub>3</sub> was found to be 12.5 wt%, at which a maximum ethylene oxide selectivity of 12.98% was obtained at the optimum applied voltage and input frequency of 15 kV and 500 Hz, respectively. Under these optimum conditions, the power consumption was found to be  $12.6 \times 10^{-16}$  W's/molecule of ethylene oxide produced. In addition, low oxygen to ethylene molar ratio and low feed gas flow rate were also experimentally found to be beneficial for the ethylene epoxidation.

## บทคัดย่อ

อโนทัย ดันสุวรรณ : การอีพอกซิเดชันของเอธิลีนโดยใช้ตัวเร่งปฏิกิริยาซิลเวอร์ภายใต้ระบบพลาสมาอุณหภูมิต่ำ (Epoxidation of Ethylene over Silver Catalysts in Low-Temperature Corona Discharge) อ. ที่ปรึกษา: ดร. ชรรณบุญ ศรีทะวงศ์ และรศ. ดร. สุเมธ ชวเวช 70 หน้า

กระบวนการอีพอกซิเดชันของเอธิลีนไปเป็นเอธิลีนออกไซด์เป็นกระบวนการออกซิเดชันที่ไม่สมบูรณ์ของสารอินทรีย์ที่มีบทบาทสำคัญอย่างยิ่งในการผลิตผลิตภัณฑ์อุตสาหกรรมที่จำเป็นหลากหลายชนิด เช่น ตัวทำละลาย ตัวเชื่อมประสาน สารลดแรงตึงผิว และโพลีโพลียูรีเทน เป็นต้น ในงานวิจัยนี้ กระบวนการอีพอกซิเดชันของเอธิลีนไปเป็นเอธิลีนออกไซด์ถูกทำการทดลองในเครื่องปฏิกรณ์พลาสมาอุณหภูมิต่ำแบบโคโรนาร่วมกับตัวเร่งปฏิกิริยา 4 ชนิด ได้แก่ โลหะเงินบนอลูมินาเฟสแอลฟาชนิดพื้นที่ผิวดำ โลหะเงินบนอลูมินาเฟสแอลฟาชนิดพื้นที่ผิวสูง โลหะเงิน-โลหะทองบนอลูมินาเฟสแอลฟาชนิดพื้นที่ผิวสูง และโลหะทองบนไทเทเนีย จากผลการทดลองพบว่า ตัวเร่งปฏิกิริยาโลหะเงินบนอลูมินาเฟสแอลฟาชนิดพื้นที่ผิวดำให้ความเฉพาะเจาะจงในการเกิดเอธิลีนออกไซด์สูงที่สุด ขณะเดียวกัน ให้ความเฉพาะเจาะจงในการเกิดคาร์บอนไดออกไซด์และคาร์บอนมอนอกไซด์ต่ำที่สุด การเพิ่มความต่างศักย์ช่วยเพิ่มความเฉพาะเจาะจงในการเกิดเอธิลีนออกไซด์ ขณะที่ความเฉพาะเจาะจงในการเกิดเอธิลีนออกไซด์ค่อนข้างคงที่เมื่อใช้ความถี่ในช่วง 300-500 เฮิร์ตซ์ อย่างไรก็ตาม ที่ความถี่มากกว่า 500 เฮิร์ตซ์ ความเฉพาะเจาะจงในการเกิดเอธิลีนออกไซด์ลดลง ปริมาณโลหะเงินบนอลูมินาเฟสแอลฟาชนิดพื้นที่ผิวดำที่ให้ความเฉพาะเจาะจงในการเกิดเอธิลีนออกไซด์มากที่สุด (12.98%) คือ 12.5 เปอร์เซ็นต์โดยน้ำหนัก ณ สภาวะที่มีความต่างศักย์และความถี่เป็น 15 กิโลโวลต์ และ 500 เฮิร์ตซ์ ตามลำดับ สำหรับพลังงานที่ใช้ในสภาวะดังกล่าวเท่ากับ  $12.6 \times 10^{-16}$  วัตต์·วินาทีต่อโมเลกุลของเอธิลีนออกไซด์ที่เกิดขึ้น นอกจากนี้ ยังพบว่า เมื่ออัตราส่วนระหว่างออกซิเจนต่อเอธิลีนในก๊าซขาเข้าและอัตราการไหลขาเข้าของก๊าซลดลง จะส่งผลดีต่อกระบวนการอีพอกซิเดชันของเอธิลีน

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