

**ETHYLENE EPOXIDATION IN LOW-TEMPERATURE
ALTERNATING CURRENT PLASMA**

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A Dissertation Submitted in Partial Fulfilment of the Requirements
for the Degree of Doctor of Philosophy
The Petroleum and Petrochemical College, Chulalongkorn University
in Academic Partnership with
The University of Michigan, The University of Oklahoma,
and Case Western Reserve University

2012


I 28373820

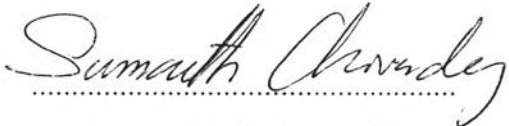
Thesis Title: Ethylene Epoxidation in Low-Temperature Alternating
Current Plasma
By: Thitiporn Suttikul
Program: Petrochemical Technology
Thesis Advisors: Prof. Sumaeth Chavadej
Prof. Hidetoshi Sekiguchi

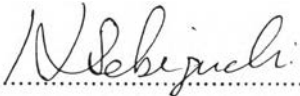
Accepted by the Petroleum and Petrochemical College, Chulalongkorn
University, in partial fulfilment of the requirements for the Degree of Doctor of
Philosophy.

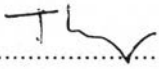

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ABSTRACT

5081007063: Petrochemical Technology

Thitiporn Suttikul: Ethylene Epoxidation in Low-Temperature
Alternating Current Plasma

Thesis Advisors: Prof. Sumaeth Chavadej and
Prof. Hidetoshi Sekiguchi 175 pp.

Keywords: Epoxidation/ Ethylene oxide/ Dielectric barrier discharge/ Plasma-
catalytic reaction / Alumina/ Silica/ Silver/ Feed position/ Corona
Discharge/ Atmospheric plasma jet

In this research, ethylene epoxidation reaction was investigated in low-temperature plasma systems: parallel plate dielectric barrier discharge (DBD), cylindrical DBD, corona discharge, and DBD jet. The combined catalytic and plasma process was initially investigated in the parallel plate DBD system, cooperating with silver catalysts loaded on two different supports (silica and alumina particles) for ethylene oxide production. From the results, the presence of silver catalysts improved the ethylene oxide production performance. The silica support interestingly provided a higher ethylene oxide selectivity than the alumina support. The optimum Ag loading on the silica support was found to be 20 wt.%, exhibiting the highest ethylene oxide selectivity of 30.6%. Next, the separate C₂H₄/O₂ feed was investigated in the cylindrical DBD system in order to improve the epoxidation performance. The C₂H₄ feed position of 0.25 was considered to be an optimum position and other operating conditions, including O₂/C₂H₄ feed molar ratio, applied voltage, input frequency, and total feed flow rate, were subsequently investigated to find out the best conditions. In comparisons with the mixed feed, the separate feed of C₂H₄ and O₂ could provide a superior ethylene epoxidation performance, resulting in higher EO selectivity and yield, and lower power consumption. These results can be explained by the fact that the C₂H₄ separate feed can reduce all undesired reactions of C₂H₄ cracking, dehydrogenation, and oxidation reactions; therefore, the separate C₂H₄/O₂ feed was used to further study in corona discharge and DBD jet. The effects

of C₂H₄ feed position, O₂/C₂H₄ feed molar ratio, applied voltage, input frequency, total feed flow rate, and electrode gap distance on ethylene epoxidation were investigated in the corona discharge reactor. The optimum operating conditions; a distance between plate electrode and C₂H₄ feed position of 0.2 cm, an O₂/C₂H₄ feed molar ratio of 1:2, an applied voltage of 18 kV, an input frequency of 500 Hz, a total feed flow rate of 100 cm³/min, and an electrode gap distance of 10 mm; provided the highest EO yield of 1.8%. The DBD jet, modified from the corona discharge and cylindrical DBD was employed for the ethylene epoxidation. The highest EO selectivity of 55.2% and yield of 27.6%, as well as the lowest power consumption were obtained at a total feed flow rate of 1,625 cm³/min, an O₂/C₂H₄ feed molar ratio of 0.25:1, an applied voltage of 9 kV, an input frequency of 300 Hz, and an inner electrode position of 0.3 mm.

บทคัดย่อ

จิติพร สุทธิกุล : ปฏิกริยาอีพอกซิเดชันโดยใช้พลาสมาอุณหภูมิต่ำแบบกระแสสลับ (Ethylene Epoxidation in Low-Temperature Alternating Current Plasma) อ.ที่ปรึกษา : ศ. ดร. สุเมธ ชวเดช และ ศ. ดร. ฮิเดโตะชิ เซคิกุจิ 175 หน้า

ในงานวิจัยนี้ ปฏิกริยาอีพอกซิเดชันได้ถูกสำรวจการภายใต้ระบบพลาสมาอุณหภูมิต่ำแบบกระแสสลับหลายระบบ ได้แก่ ระบบพลาสมาชนิดไดอิเล็กทริกแบริเออร์ดีสชาร์จ(ดีบีดี)แบบทรงสี่เหลี่ยมด้านขนานและแบบทรงกระบอก ระบบพลาสมาชนิดโคโรนา และระบบพลาสมาชนิดไดอิเล็กทริกแบริเออร์ดีสชาร์จแบบไอพ่น กระบวนการร่วมพลาสมาและการเร่งปฏิกิริยาถูกศึกษาเริ่มแรกโดยใช้พลาสมาชนิดดีบีดีแบบแผ่นคู่ขนานบนตัวเร่งปฏิกิริยาเงินบนฐานรองรับซิลิกา กับอลูมินาสำหรับปฏิกริยาอีพอกซิเดชัน จากผลการทดลองพบว่า ตัวเร่งปฏิกิริยาเงินเพิ่มสมรรถนะของการผลิตเอธีลีนออกไซด์ ฐานรองรับซิลิกาให้ค่าการเลือกเกิดผลิตภัณฑ์เอธีลีนออกไซด์สูงกว่าฐานรองรับอลูมินา ปริมาณโลหะเงินที่เหมาะสมบนฐานรองรับซิลิกาพบว่าเป็น 20 เปอร์เซ็นต์โดยน้ำหนัก ซึ่งให้ค่าการเลือกเกิดเอธีลีนออกไซด์สูงสุดเป็น 30.6% จากนั้นการป้อนแยกของก๊าซเอธีลีนกับออกซิเจนถูกใช้ในการทดลองภายใต้ระบบพลาสมาชนิดไดอิเล็กทริกแบริเออร์ดีสชาร์จแบบทรงกระบอกเพื่อเพิ่มประสิทธิภาพของปฏิกริยาอีพอกซิเดชัน ตำแหน่งในการป้อนเอธีลีน 0.25 ถูกพิจารณาว่าเป็นตำแหน่งที่เหมาะสม และได้ทำการศึกษาตัวแปรต่างๆเช่น อัตราส่วนโดยโมลของออกซิเจนต่อเอธีลีน ความต่างศักย์ไฟฟ้า ความถี่ไฟฟ้า และอัตราการไหลของสารตั้งต้นเพื่อค้นหาสภาวะที่ดีที่สุด ในการเปรียบเทียบกับระบบการป้อนรวม ระบบการป้อนแยกของเอธีลีนกับออกซิเจนสามารถให้สมรรถนะปฏิกริยาอีพอกซิเดชันที่สูงกว่า ซึ่งส่งผลให้ค่าการเลือกเกิดเอธีลีนออกไซด์และผลได้ของเอธีลีนออกไซด์สูงขึ้น ในขณะที่พลังงานที่ใช้ในการผลิตเอธีลีนออกไซด์ต่ำลง ผลการทดลองนี้สามารถอธิบายได้ว่า การป้อนแยกของเอธีลีนสามารถลดปฏิกิริยาที่ไม่ต้องการทั้งหมดของปฏิกริยาการแตกตัวของเอธีลีน ปฏิกริยาการดึงไฮโดรเจนออกจากโมเลกุล และปฏิกริยาออกซิเดชัน ดังนั้นการป้อนแยกระหว่างเอธีลีนกับออกซิเจนจึงถูกนำไปใช้ศึกษาต่อในระบบพลาสมาชนิดโคโรนา และไดอิเล็กทริกแบริเออร์ดีสชาร์จแบบไอพ่น ผลของระยะห่างระหว่างขั้วไฟฟ้าแบบแผ่นและตำแหน่งป้อนก๊าซเอธีลีน อัตราส่วนโดยโมลของออกซิเจนต่อเอธีลีน ความต่างศักย์ไฟฟ้า ความถี่ไฟฟ้า อัตราการไหลของสารตั้งต้น และระยะห่างระหว่างขั้วไฟฟ้า ต่อปฏิกริยาเอธีลีนอีพอกซิเดชันถูกทดลองในพลาสมาชนิดโคโรนา จากการทดลองพบว่าสภาวะที่เหมาะสมดังต่อไปนี้ ระยะห่างระหว่างขั้วไฟฟ้าแบบแผ่นและตำแหน่งป้อน

ก๊าซเอธิลีน 0.2 เซนติเมตร อัตราส่วน โดยโมลของออกซิเจนต่อเอธิลีน 0.25:1 ความต่างศักย์ไฟฟ้า 18 กิโลโวลต์ ความถี่ไฟฟ้า 500 เฮิร์ตซ์ อัตราการไหลของสารตั้งต้น 100 ลูกบาศก์เซนติเมตรต่อ นาที และระยะห่างระหว่างขั้วไฟฟ้า 1 เซนติเมตร ให้ผลได้ของเอธิลีนสูงสุด พลาสมาชนิดได อิเล็กทริกแบรีเออร์ดีสแบบไอพ่นซึ่งถูกปรับเปลี่ยนมาจากพลาสมาชนิดโคโรนาและไดอิเล็กทริก แบรีเออร์ดีสซาร์จ ถูกใช้สำหรับปฏิกิริยาอีพอกซิเคชันพบว่า ค่าการเลือกสรรในการเกิดเอธิลีนออก ไซด์และผล ได้ของเอธิลีนออก ไซด์สูงสุด ตลอดจนพลังงานที่ใช้ในการผลิตเอธิลีนออก ไซด์ต่ำสุด ได้รับจากสภาวะดังต่อไปนี้ อัตราการไหลของสารตั้งต้น 1,625 ลูกบาศก์เซนติเมตรต่อ นาที อัตราส่วน โดยโมลของออกซิเจนต่อเอธิลีน 0.25:1 ความต่างศักย์ไฟฟ้า 9 กิโลโวลต์ ความถี่ไฟฟ้า 300 เฮิร์ตซ์ ตำแหน่งอิเล็กโทรดตัวใน 0.3 มิลลิเมตร

ACKNOWLEDGEMENTS

This work could not be completed successfully without the contribution of the following individuals and organizations.

First of all, I would like to express my sincere gratitude to my thesis advisors, Prof. Sumaeth Chavadej and Prof. Hidetoshi Sekiguchi, for all of their direction, valuable guidance and assistance throughout my research. Prof. Hidetoshi Sekiguchi taught me about the technical knowledge of plasma devices with valuable suggestions while I was conducting my work at Tokyo Institute of Technology (TIT). I was indebted unforgettably to him for giving me a great opportunity to visit his plasma laboratory at TIT.

I would also like to specially thank Dr. Thammanoon Sreethawong, for all helpfulness, fruitful suggestions, and discussions. I would like to express my gratitude to Mrs. Barbara Best for her valuable comments and corrections in writing papers and thesis.

It is a pleasure to acknowledge the Petroleum and Petrochemical College, Chulalongkorn University; the Center of Excellence on Petrochemical and Materials Technology, Thailand; the Department of Chemical Engineering, Tokyo Institute of Technology, Japan, for providing research facilities; and the Dudsadeepipat Scholarship, Chulalongkorn University, Thailand, for providing me a Ph.D. scholarship.

I also especially extend many thanks to Mr. Matsumura Yasutaka (my tutor), Mr. Takashi Nakashima, and everyone in Prof. Sekiguchi's plasma group as well as Thai friends for their kindness who help during my stay in Tokyo. At PPC, I would like to sincerely thank Mr. Sanit Prinakorn, Mr. Chaturong Tiamsiri, and Mr. Poon Arj-proo for helping me to make all electrical and mechanical pieceworks.

Last but not least, the great thankfulness is forwarded to my God and my family for their encouragement, strong belief, and understanding in me.

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