

**CATALYST DEVELOPMENT  
FOR METHANE REFORMING WITH CO<sub>2</sub>**



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A Thesis Submitted in Partial Fulfillment of the Requirements  
for the Degree of Master of Science  
The Petroleum and Petrochemical College, Chulalongkorn University  
in Academic Partnership with  
The University of Michigan, The University of Oklahoma  
and Case Western Reserve University

1999

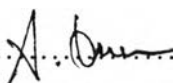
ISBN 974-331-897-6

I19584830

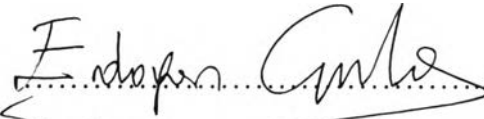
**Thesis Title** : Catalyst Development for Methane Reforming with CO<sub>2</sub>  
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**Program** : Petrochemical Technology  
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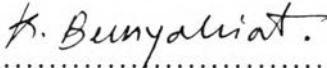
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Accepted by the Petroleum and Petrochemical College, Chulalongkorn University, in partial fulfillment of the requirements for the Degree of Master of Science.


  
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## ABSTRACT

##971009 : PETROCHEMICAL TECHNOLOGY PROGRAM

KEY WORDS : Methane Reforming/Nickel/Sol-Gel

Krit Punburananon : Catalyst Development for Methane Reforming with CO<sub>2</sub>. Thesis Advisors : Prof. Erdogan Gulari, Assoc. Prof. Kunchana Bunyakiat, and Dr. Thirasak Rirksomboon 51 pp ISBN 974-331-897-6

The reforming of methane with CO<sub>2</sub> produces synthesis gas with high CO/H<sub>2</sub> ratio, which is suitable to produce higher hydrocarbons and oxygenated compounds by Fischer-Tropsch synthesis. In this study, the sol-gel technique, which has several advantages over conventional technique, was applied to prepare 5% Ni/Al<sub>2</sub>O<sub>3</sub> catalyst and alumina support. The performance of 5% sol-gel catalyst on CH<sub>4</sub> reforming with CO<sub>2</sub> was compared to that of 5% impregnation catalysts supported on commercial alumina and on sol-gel alumina. It was found that all three catalysts deactivated with time on stream, because of carbon deposition on the catalysts resulting in total loss of catalytic activity. In addition, the reverse water gas shift reaction, the side reaction, uses H<sub>2</sub> to produce CO. Therefore, CO selectivity is higher than H<sub>2</sub> selectivity. Temperature programmed oxidation (TPO) on thermogravimetric analyzer (TGA) was used to determine the amount of carbon on the three prepared catalysts used for 20 hours. It was found that the carbon deposition on the catalyst can be oxidized at high temperatures in the range of 670-700 °C and % carbon on Ni/sol-gel Al<sub>2</sub>O<sub>3</sub> is 17.61%, whereas that on sol-gel Ni/Al<sub>2</sub>O<sub>3</sub> is 25.15% and on Ni/commercial Al<sub>2</sub>O<sub>3</sub> is 17.89%.

## บทคัดย่อ

กฤษฎี พันธุ์บูรณานนท์ : การพัฒนาตัวเร่งปฏิกิริยาการรีฟอร์มก๊าซมีเทนด้วยก๊าซคาร์บอนไดออกไซด์ (Catalyst Development for Methane Reforming with CO<sub>2</sub>)  
 อ. ที่ปรึกษา : ศ.ดร. เออโดแกน กุลาริ (Prof. Erdogan Gulari) รศ. กัญจนา บุญเกียรติ และ  
 ดร. ชีรศักดิ์ ฤกษ์สมบูรณ์ 51 หน้า ISBN 974-331-897-6

การรีฟอร์มก๊าซคาร์บอนไดออกไซด์ (CO<sub>2</sub>) ด้วยก๊าซมีเทน (CH<sub>4</sub>) เพื่อผลิตก๊าซสังเคราะห์ (synthesis gas) ที่มีสัดส่วนของก๊าซไฮโดรเจน (H<sub>2</sub>) ต่อก๊าซคาร์บอนมอนอกไซด์ที่เหมาะสมกับการผลิตไฮโดรคาร์บอนโมเลกุลใหญ่โดยปฏิกิริยาฟิชเชอร์ทรอป (Fischer-Tropsch) งานวิจัยนี้ใช้ 5% Ni/Al<sub>2</sub>O<sub>3</sub> เป็นตัวเร่งปฏิกิริยา เตรียมโดยวิธีโซลเจล (sol-gel technique) และวิธีอิมเพกเนชัน (impregnation technique) บนอลูมินาที่ใช้ในอุตสาหกรรมและอลูมินาที่เตรียมโดยวิธีโซลเจล จากการศึกษาพบว่าตัวเร่งปฏิกิริยาเสียความว่องไวเนื่องจากการเกาะตัวของคาร์บอนบนตัวเร่งปฏิกิริยา นอกจากนี้ปฏิกิริยาริเวอร์ส วอเตอร์ก๊าซ ชิฟท์ (reverse water gas shift reaction) มีผลให้ ค่าการเลือกของคาร์บอนมอนอกไซด์ (CO selectivity) สูงกว่าค่าการเลือกของไฮโดรเจน (H<sub>2</sub> selectivity) ส่วนการหาปริมาณคาร์บอนที่เกาะตัวบนตัวเร่งปฏิกิริยากระทำในเครื่องวิเคราะห์ทางความร้อนชนิดโปรแกรม อุณหภูมิได้ภายใต้บรรยากาศที่มีก๊าซออกซิเจน (temperature programmed oxidation on thermogravimetric analyzer) พบว่า คาร์บอนสามารถทำปฏิกิริยากับก๊าซออกซิเจนเป็น ก๊าซคาร์บอนไดออกไซด์ (CO<sub>2</sub>) ในช่วงอุณหภูมิ 670-700 °C และปริมาณคาร์บอนบน sol-gel Ni/Al<sub>2</sub>O<sub>3</sub>, Ni/commercial Al<sub>2</sub>O<sub>3</sub> และ Ni/sol-gel Al<sub>2</sub>O<sub>3</sub> เป็น 25.15%, 17.89% และ 17.61% ตามลำดับ

## ACKNOWLEDGMENTS

This work cannot be successful without the participation of the following individuals and organizations.

First of all, I wish to express my utmost appreciation and deepest gratitude to my thesis advisors, Professor Erdogan Gulari, Associate Professor Kunchana Bunyakiat and Dr. Thirasak Rirksomboon, for their guidance, discussion, strong encouragement and assistance devotedly throughout my thesis work.

I would like to extend my sincere thanks to all of the Professors who guided me through their courses, establishing the knowledge needed for this thesis. I also would like to thank Assoc. Prof. Sumaeth Chavadej for serving on my thesis committee.

It is a pleasure to acknowledge the PTT Research and Technology Institute for financial support, the Petroleum and Petrochemical College for the support in laboratory facilities and all of the staffs of the college for their helpful assistance.

Very special thanks are forwarded to all of my friends for making my two years of study at the college most memorable and enjoyable.

Finally, I would like to express whole-hearted gratitude to my family for their love, encouragement and measureless support.

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