

**SELECTIVE NO_x REDUCTION OVER METAL SUPPORTED
ON ALUMINA CATALYSTS DERIVED FROM SOL-GEL METHOD**

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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

2010

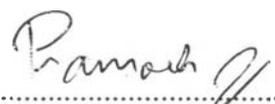
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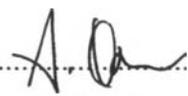
Thesis Title: Selective NO_x Reduction over Metal Supported on Alumina Catalysts Derived from Sol-Gel Method
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ABSTRACT

4291001063: Petrochemical Technology Program

Jiraporn Leerat: Selective NO_x Reduction over Metal Supported on Alumina Catalysts Derived from Sol-gel Method

Thesis Advisors: Prof. Somchai Osuwan and Prof. Erdogan Gulari

113 pp.

Keywords: Sol-gel/ Au/ Pt/ Alumina/ SCR/ Propene/ Urea

The selective catalytic reduction of NO_x by propene and urea were studied over gold and platinum on alumina supported catalysts. These catalysts were prepared from sol-gel technique together with other conventional methods. They were tested over the temperature range of 150-500 °C under various conditions. The presence of water and SO₂ coupled with different oxygen concentrations in reactant gas streams were employed in order to investigate the catalytic activities. It was found that the preparation method had strong influence on the catalytic performance. Among different gold catalysts, the one that prepared from single step sol-gel exhibited the highest activity. However, the impregnated sol-gel alumina was the best catalyst in terms of overall performance that the formation of N₂ was also concerned. In selective reduction of NO_x by propene, almost 100% NO_x conversion was found. In addition, its activity window was enlarged in higher oxygen concentrations. Water was considered to enhance NO_x conversion. Conversely, it suppressed the selectivity towards N₂. Furthermore, the catalysts exhibited good stability under prolonged time on stream in both dry and humid conditions. In contrast, both gold and platinum catalysts showed the average activity in selective NO_x reduction with urea. Increasing in the activity with the temperature was observed on gold catalysts while platinum catalyst reached the maximum NO_x conversion around 250-300 °C. The study of catalytic performance with urea was extended over platinum catalyst. It gave negative NO_x conversion at high temperature region for urea delivery from aqueous solution system. This resulted from the oxidation of derivative nitrogen-containing compounds from urea.

บทคัดย่อ

จิราพร สิริรัตน์ : ปฏิริยาการเลือกเกิดรีดักชันของก๊าซไนโตรเจนออกไซด์ โดยตัวเร่งปฏิริยาโลหะบนตัวรองรับอลูมินาที่สังเคราะห์โดยวิธีโซลเจล (Selective NO_x Reduction over Metal Supported on Alumina Catalysts Derived from Sol-gel Method) อ. ที่ปรึกษา : ศาสตราจารย์กิตติคุณ ดร. สมชาย ไอสุวรรณ และ ศ. ดร. เออร์โดแกน กุลาริ 113 หน้า

งานวิจัยนี้ ได้ศึกษาปฏิริยาการเลือกเกิดรีดักชันของก๊าซไนโตรเจนออกไซด์บนตัวเร่งปฏิริยาทองและแพลทตินัมบนตัวรองรับอลูมินาโดยใช้สารโพรฟินและยูเรีย ซึ่งตัวเร่งปฏิริยาต่างๆ นี้ ถูกเตรียมจากวิธีโซลเจลและวิธีการสังเคราะห์แบบดั้งเดิมอื่นๆ ในการทดสอบตัวเร่งปฏิริยาเหล่านี้ได้ศึกษาในช่วงอุณหภูมิ 150-500 °C ภายใต้สภาวะการทำปฏิริยาต่างๆ กัน โดยใช้ก๊าซตั้งต้นในการทำปฏิริยาที่ประกอบด้วยไอน้ำและก๊าซซัลเฟอร์ไดออกไซด์ ร่วมกับก๊าซออกซิเจนที่ความเข้มข้นต่างๆ เพื่อสำรวจความว่องไวในการทำปฏิริยา จากการศึกษาแสดงให้เห็นว่า วิธีการเตรียมตัวเร่งปฏิริยามีอิทธิพลต่อประสิทธิภาพในการทำปฏิริยาอย่างมาก เมื่อเปรียบเทียบกันระหว่างตัวเร่งปฏิริยาต่างๆ พบว่าตัวเร่งปฏิริยาที่เตรียมจากวิธีโซลเจล 1 ขั้นตอน แสดงความว่องไวในการทำปฏิริยาสูงที่สุด อย่างไรก็ตาม หากพิจารณาถึงความสามารถโดยรวมของตัวเร่งปฏิริยาซึ่งเกี่ยวข้องกับการเกิดก๊าซผลิตภัณฑ์ไนโตรเจน พบว่าตัวเร่งปฏิริยาโซลเจลอลูมินาชนิดที่ถูกทำให้ชุ่ม เป็นตัวเร่งปฏิริยาที่มีประสิทธิภาพสูงสุดในปฏิริยาการเลือกเกิดรีดักชันด้วยสารโพรฟิน พบว่าตัวเร่งปฏิริยาทองสามารถเปลี่ยนก๊าซไนโตรเจนออกไซด์ได้สูงเกือบ 100% นอกจากนี้ ก๊าซออกซิเจนที่มีความเข้มข้นสูงสามารถขยายช่วงอุณหภูมิความว่องไวของปฏิริยาให้กว้างขึ้น ส่วนไอน้ำนั้นช่วยเสริมประสิทธิภาพในการเปลี่ยนก๊าซไนโตรเจนไดออกไซด์ แต่ส่งผลต่อการเลือกเกิดผลิตภัณฑ์ก๊าซไนโตรเจนให้ลดลง นอกเหนือจากนี้ ตัวเร่งปฏิริยาทองแสดงความเสถียรเมื่อขยายระยะเวลาการใช้งานทั้งในสภาวะแห้งและชื้น เมื่อเปรียบเทียบกับปฏิริยาการเลือกเกิดรีดักชันด้วยสารยูเรีย พบว่าตัวเร่งปฏิริยาทองและแพลทตินัมมีความว่องไวปานกลาง โดยตัวเร่งปฏิริยาทองมีความว่องไวในการเกิดปฏิริยาสูงขึ้นเมื่อเพิ่มอุณหภูมิ ในขณะที่ตัวเร่งปฏิริยาแพลทตินัมมีประสิทธิภาพในการเปลี่ยนก๊าซไนโตรเจนไดออกไซด์สูงสุดในช่วงอุณหภูมิ 250-300 °C จากการศึกษาเพิ่มเติมในการเกิดปฏิริยารีดักชันด้วยสารยูเรียบนตัวเร่งปฏิริยาแพลทตินัม โดยการใช้ยูเรียในรูปของสารละลายพบว่าตัวเร่งปฏิริยาแพลทตินัมให้ผลการเปลี่ยนก๊าซไนโตรเจนไดออกไซด์เป็นลบในช่วงอุณหภูมิสูง เนื่องจากการเกิดปฏิริยาออกซิเดชันของสารประกอบไนโตรเจนที่เกิดจากยูเรีย

ACKNOWLEDGEMENTS

First of all, I would like to express my deepest gratitude to my advisor, Prof. Somchai Osuwan to give me the endless opportunities throughout my study. He always provides consistent supports not only on academia but also on other issues which help me to go through the difficulties. He took attention enthusiastically from the beginning to the end of this work. His boundless generosity is overwhelmingly appreciated.

I am very grateful to Prof. Erdogan Gulari for partial financial support. He took me a very good care during the experimental work in his research group at The University of Michigan. He always gave innovative guidance and valuable advices on the research.

The Thailand Research Fund (TRF) is gratefully acknowledged for providing financial support through two programs of the Royal Golden Jubilee Ph.D. and the Basic Research Grant. This thesis work was also funded by the Petroleum and Petrochemical College, and by the National Center of Excellence for Petroleum, Petrochemicals, and Advanced Materials, Thailand.

The graduated work could not be successful without the assistance of my bosses, in particular Khun Thanyathorn Torai. She put very much effort throughout the final part of this work. Her truly goodwill is highly appreciated. Additionally, Bangkok Synthetics Co., Ltd. and BST Elastomers Co., Ltd. are gratefully acknowledged.

I also would like to give my appreciation to faculty and staff of the Petroleum and Petrochemical College together with friends for their supportive suggestions, kind helps, cheerfulness and sincere friendships.

This work is solely dedicated to my beloved father and mother for their wholehearted love and forever supports. I am most obliged to my husband for his understanding, constant encouragement and sharing the difficult times. I also would like to deliver special thanks to Pongsirisatorn family for their hospitalities.

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ABBREVIATIONS

BET	Brunauer-Eemmett-Teller
BJH	Barrett-Joyner-Halenda
DP	Deposition-precipitation
FWHM	Full width at half maximum
FTIR	Fourier-transform infrared spectroscopy
GC	Gas chromatography
HC	Hydrocarbon
HID	Helium ionization detector
HPGe	High purity germanium
HRTEM	High resolution transmission electron microscopy
ICP	Inductively coupled plasma
ID	Inside diameter
IMP	Impregnation
LDD	Light duty diesel
NAA	Neutron activation analysis
NO_x	Nitric oxide and nitrogen dioxide
NSC	NO _x storage compound
NSCR	Non-selective catalytic reduction
NSR	NO _x storage and reduction
NTP	Non-thermal plasma
MCT	Mercury cadmium telluride
PID	Proportional integral derivative
SNCR	Selective non-catalytic reduction
PGM	Platinum group metal
PMT	Photomultiplier tube
SCR	Selective catalytic reduction
SG	Sol-gel
SV	Space velocity

TCD	Thermal conductivity detector
TWC	Three-way catalyst
UAMR	Ultrasound-assisted membrane reduction
UHP	Ultra high purity
XRD	X-ray diffraction