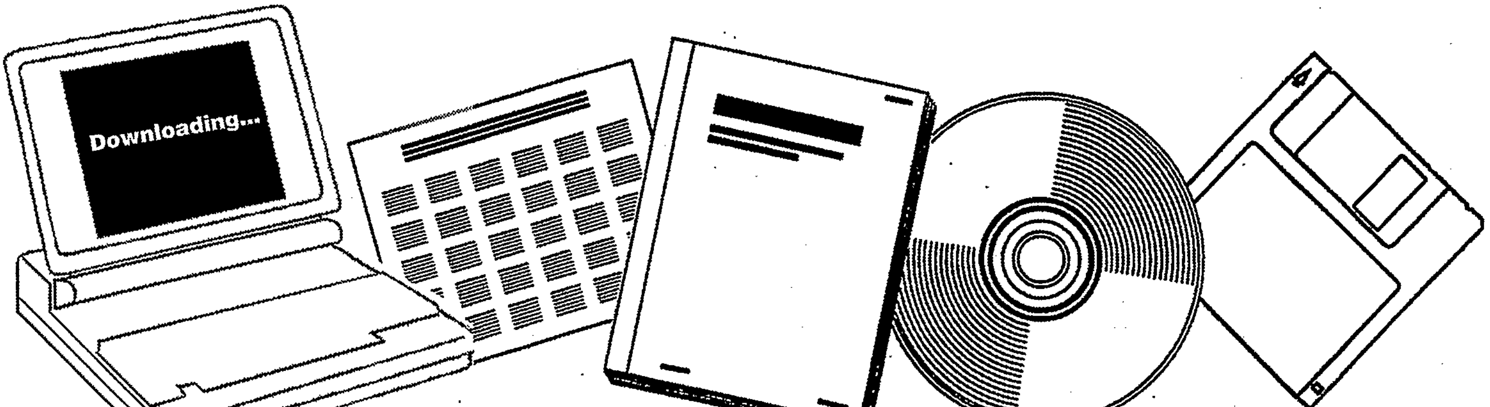
DE86007674

# CHEMICAL TRAPPING AND IR STUDIES OF CO/H EXP 2 SURFACE SPECIES. SECOND QUARTERLY REPORT, DECEMBER 1985-FEBRUARY 1986

PITTSBURGH UNIV., PA. DEPT. OF CHEMICAL
AND PETROLEUM ENGINEERING

MAR 1986



## One Source. One Search. One Solution.

# NTIS

### Providing Permanent, Easy Access to U.S. Government Information

National Technical Information Service is the nation's largest repository and disseminator of government-initiated scientific, technical, engineering, and related business information. The NTIS collection includes almost 3,000,000 information products in a variety of formats: electronic download, online access, CD-ROM, magnetic tape, diskette, multimedia, microfiche and paper.

**Search the NTIS Database from 1990 forward**

NTIS has upgraded its bibliographic database system and has made all entries since 1990 searchable on **www.ntis.gov.** You now have access to information on more than 600,000 government research information products from this web site.

**Link to Full Text Documents at Government Web Sites**

Because many Government agencies have their most recent reports available on their own web site, we have added links directly to these reports. When available, you will see a link on the right side of the bibliographic screen.

**Download Publications (1997 - Present)**

NTIS can now provides the full text of reports as downloadable PDF files. This means that when an agency stops maintaining a report on the web, NTIS will offer a downloadable version. There is a nominal fee for each download for most publications.

For more information visit our website:

**www.ntis.gov**

U.S. DEPARTMENT OF COMMERCE
Technology Administration
National Technical Information Service
Springfield, VA 22161

DOE/PC/80526--T2
DE86 007674

# Chemical Trapping and IR Studies of $CO/H_2$ Surface Species

2nd Quarterly Report
December 1985 - February 1986

Donna G. Blackmond (PI)
Irving Wender (Co-PI)

Department of Chemical and Petroleum Engineering
University of Pittsburgh
Pittsburgh, PA 15261

March 1986

Prepared for the U.S. Department of Energy
under Grant No. DE-FG22-85PC80526

# TABLE OF CONTENTS

1. OBJECTIVE AND SCOPE OF WORK .......... 2

2. SUMMARY OF PROGRESS .......... 2

3. DETAILED DESCRIPTION OF TECHNICAL PROGRESS .......... 3

Personnel .......... 3

Equipment .......... 3

IR Investigations .......... 4

Model Transition Metal Complexes .......... 4

4. FORECAST OF WORK .......... 4

## 1. OBJECTIVE AND SCOPE OF WORK

CO hydrogenation reactions over supported metals can produce a wide spectrum of hydrocarbon and oxygenated compounds depending to a large extent on the type of metal used. Elucidating the nature of the intermediate species formed on metal surfaces may provide the key to understanding the pathways through which these reactions proceed on different metals. Research is proposed to investigate intermediate species formed during CO hydrogenation reactions on silica-supported Pd, Rh, Ru, and Ni catalysts. A novel chemical trapping technique will be used to identify adsorbed intermediates. Coupled with in-situ IR spectroscopic investigations of adsorption and reaction on the catalysts, these trapping studies should provide a framework for understanding differences in CO hydrogenation reaction pathways over different metals. In addition, chemical trapping studies using selected transition metal complexes of known structure will be carried out to help evaluate the technique as well as to help establish relationships between homogeneous and heterogeneous catalytic reactions of CO and $H_2$.

## 2. SUMMARY OF PROGRESS

During the second quarter of the project, emphasis has been placed on the construction of the chemical trapping reaction system and installation of the gas chromatograph. Two of the four catalyst sytems were prepared, those being $Ni/SiO_2$ and $Rh/SiO_2$. Preliminary IR investigations of CO adsorption on $Rh/SiO_2$ have been performed.

## 3. DETAILED DESCRIPTION OF TECHNICAL PROGRESS

### Personnel

Mr. John A. Williams and Mr. Prakosh G. Bendale continued to work as Ph.D. candidates in Chemical Engineering to assist the P.I.'s (Professors Blackmond and Wender).

### Equipment

The reaction selectivity and in-situ chemical trapping studies during CO hydrogenation will be performed in single-pass glass or stainless steel microreactors equiped with on-line gas chromatographic analysis. The design of the reactor system was schematically shown in the last quarterly report (DOE/PC/85026-1). Three ultra high purity feed streams ($H_2$/CO, $H_2$, He) will be available to the reaction system. Flow rates will be accurately controlled via separate Brooks Micro Mass flow controllers. Valving is arranged for the selection of one or more of the feed streams and switching therein. CO hydrogenation will be performed using a gaseous feed stream of a 2:1 $H_2$/CO mixture of at a flow rate of 10 to 200 cc/min (STP). Reaction conditions will be varied depending on catalyst selectivity. Anticipated reaction conditions are temperatures of 200-300°C and 0-10 atmospheres pressure. Trapping will be performed by redirection of a gaseous feed stream and passing it through a thermostated saturator containing the trapping agent. Catalyst sample weights will be chosen so as to maintain differential CO conversions. It is estimated that 100 to 1000 mg of catalyst sample will be required in the microreactor to obtain accurate analysis. Steady state gaseous reactor effluent samples will be taken periodically through an automatic sampling valve.

The reactor controls have been mounted and wired. The remainder of the reactor system has been constructed. The sampling valve is presently being installed. Standardization of the gas chromatograph has begun.

## IR Investigations

Preliminary IR investigations of CO adsorption on Rh/$SiO_2$ were performed.

## Model Transition Metal Complexes

The selection of model transition metal complexes were previously investigated and two complexes were chosen. The complexes are acetylene dicobalt hexacarbonyl ($C_2H_2(Co(CO)_3)_2$) and ethyl tricobalt enneacarbonyl ($Co_3(CO)_9CCH_3$). Preparation methods were previously determined for both complexes and the acetylene dicobalt hexacarbonyl complex was prepared for us commercially. The ethyl tricobalt enneacarbonyl model transition metal complex is still under preparation. These complexes will be contacted with the trapping reagents under various conditions to determine whether the organic ligands will interact with the reagent in the same manner as it is conjectured the hydrocarbon species on a metal catalyst surface do. These studies will enable us to test and evaluate the trapping technique for identification of surface species.

## 4. FORECAST OF WORK

During the third quarter, final testing of the reaction system and gas chromatograph is planned. Preliminary chemical trapping reaction work will commence with Ni/$SiO_2$ and then work will be focused on Rh/$SiO_2$. Preliminary IR work will continue with Ni/$SiO_2$ and Rh/$SiO_2$. Reactivity of the two model transition metal complexes will be investigated as described above.

# Reproduced by NTIS

National Technical Information Service
Springfield, VA 22161

***This report was printed specifically for your order from nearly 3 million titles available in our collection.***

For economy and efficiency, NTIS does not maintain stock of its vast collection of technical reports. Rather, most documents are custom reproduced for each order. Documents that are not in electronic format are reproduced from master archival copies and are the best possible reproductions available.

Occasionally, older master materials may reproduce portions of documents that are not fully legible. If you have questions concerning this document or any order you have placed with NTIS, please call our Customer Service Department at (703) 605-6050.

## About NTIS

NTIS collects scientific, technical, engineering, and related business information – then organizes, maintains, and disseminates that information in a variety of formats – including electronic download, online access, CD-ROM, magnetic tape, diskette, multimedia, microfiche and paper.

The NTIS collection of nearly 3 million titles includes reports describing research conducted or sponsored by federal agencies and their contractors; statistical and business information; U.S. military publications; multimedia training products; computer software and electronic databases developed by federal agencies; and technical reports prepared by research organizations worldwide.

For more information about NTIS, visit our Web site at http://www.ntis.gov.

NTIS

**Ensuring Permanent, Easy Access to U.S. Government Information Assets**

**SATISFACTION GUARANTEED**

**NTIS strives to provide quality products, reliable service, and fast delivery. Please contact us for a replacement within 30 days if the item you receive is defective or if we have made an error in filling your order.**

**▶E-mail: info@ntis.gov**
**▶Phone: 1-888-584-8332 or (703)605-6050**

U.S. DEPARTMENT OF COMMERCE
Technology Administration
National Technical Information Service
Springfield, VA 22161 (703) 605-6000