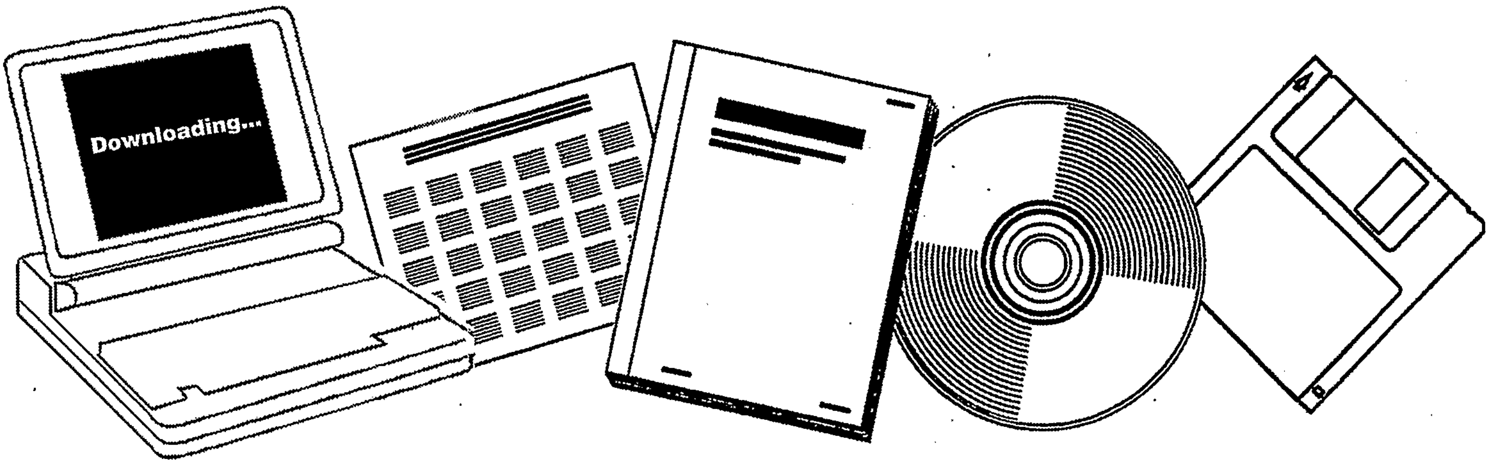
CHEMICAL TRAPPING AND IR STUDIES OF CO/H SUB 2 SURFACE SPECIES. FIRST QUARTERLY REPORT, SEPTEMBER-NOVEMBER 1985

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

DEC 1985



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--T1

DE86 004847

Chemical Trapping and IR Studies of CO/H_2 Surface Species

1st Quarterly Report
September 1985 - November 1985

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

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

December 1985

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

Received by OSTI

JAN 1 3 1986

TABLE OF CONTENTS

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 techniques as well as to help establish relationships between homogeneous and heterogeneous catalytic reactions of CO and H_2.

2. SUMMARY OF PROGRESS

During the first quarter of the project, a research team has been assembled, the high pressure reaction system has been designed and reaction parts and supplies have been ordered. Preliminary IR investigations of CO adsorption on Rh/Al_2O_3 have been performed. Preparation methods of model transition metal complexes has been investigated.

3. DETAILED DESCRIPTION OF TECHNICAL PROGRESS

Personnel

Mr. John A. Williams and Mr Prakash G. Bendale have been recruited 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 equipped with on-line gas chromatographic analysis. The design of the reactor system is schematically shown in Figure 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 will be 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 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 through a thermostated saturator containing the trapping agent. Catalyst sample weights will be chosen so as to maintain differential CO conversions. It is estimated 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 .

All components of this sytems have been ordered, and construction of the system has commenced.

IR Investigations

Preliminary IR investigations of CO adsorption on Rh/Al_2O_3 were performed. Pressing of wafers was investigated and instrument para- meters varied to optimize spectra.

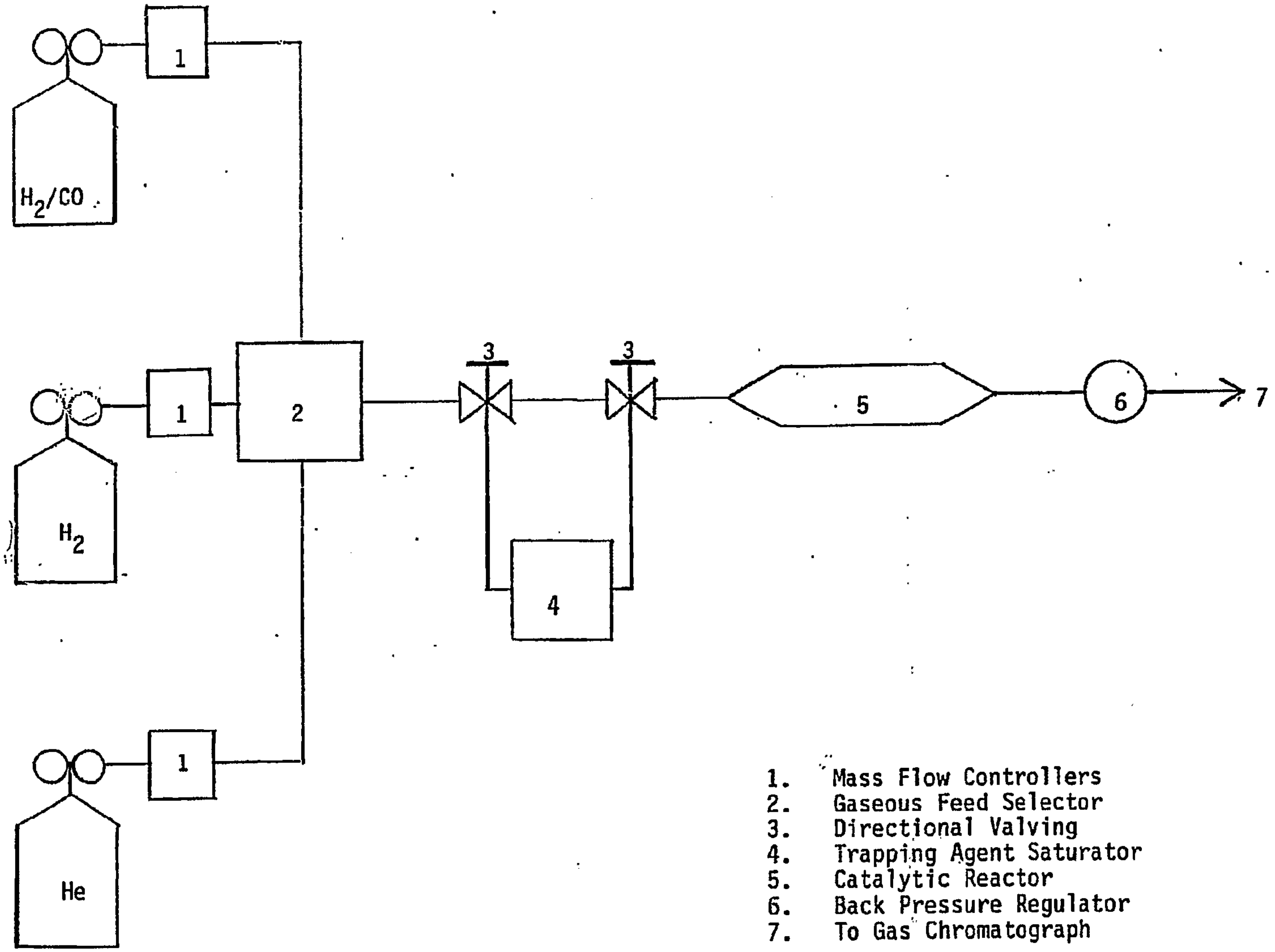
Model Transition Metal Complexes

The selection of model transition metal complexes was investigated and two complexes were chosen. The complexes are acetylene dicobalt hexacarbonyl ($C_2H_2(Co(CO)_3)_2$) and ethylidyne tricobalt nonacarbonyl ($Co_3(CO)_9CCH_3$). Preparation methods were determined for both complexes. The first of the two has been prepared commercially for us and the second is under preparation. These complexes will be contacted with 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 second quarter, completion of construction and testing of the reaction system is planned. Preliminary IR work will continue (Rh/SiO_2 and Ni/SiO_2). Model transition metal complexes will be studied as described above.

Figure 1: Schematic Diagram of Trapping Reactor System



1. Mass Flow Controllers
2. Gaseous Feed Selector
3. Directional Valving
4. Trapping Agent Saturator
5. Catalytic Reactor
6. Back Pressure Regulator
7. To Gas Chromatograph

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