DESIGN OF A HIGH ACTIVITY AND SELECTIVITY ALCOHOL CATALYST. FIRST QUARTERLY REPORT, AUGUST 7, 1990-NOVEMBER 6, 1990

DELAWARE UNIV., NEWARK. CENTER FOR CATALYTIC SCIENCE AND TECHNOLOGY

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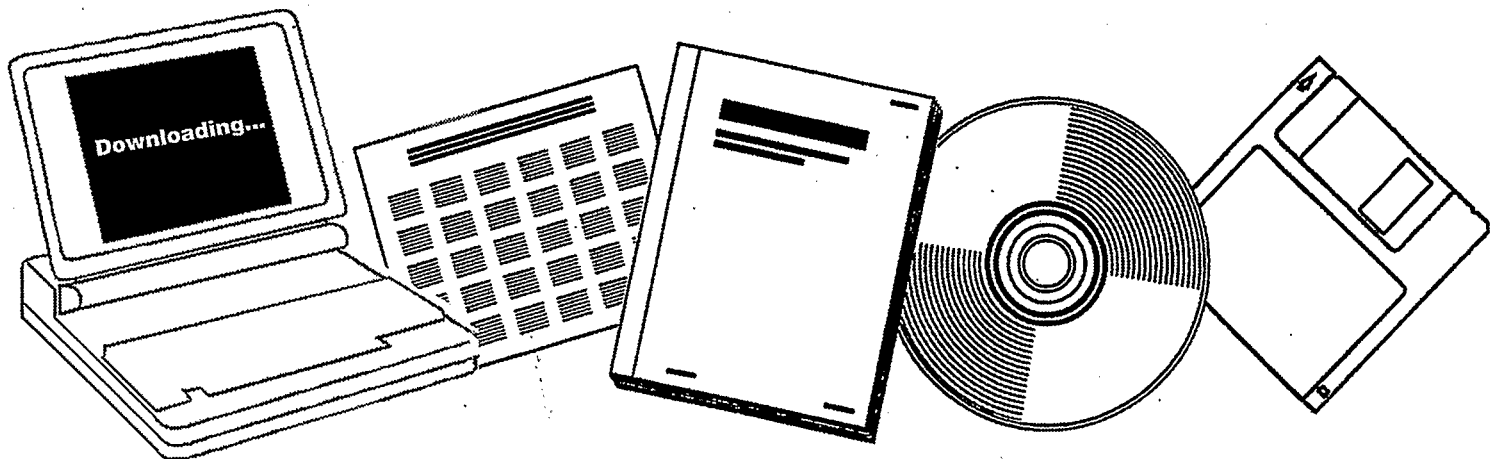

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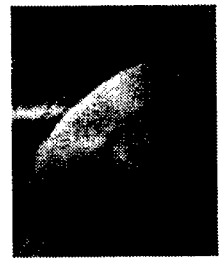

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DESIGN OF A HIGH ACTIVITY AND SELECTIVITY ALCOHOL CATALYST

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First Quarterly Report for Period
August 7, 1990 to November 6, 1990

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Summary

During the first quarter of this project, our goals have been to overhaul key experimental equipment used in the previous project, plan and design new equipment and to identify a person to carry out the research program.

A new, Ph.D. candidate, Mr. Eric Lowenthal, has been selected to carry out this project as the basis for his Ph.D. thesis dissertation in Chemical Engineering.

The flow microreactor, previously assembled, has been reactivated and major improvements have been made both to the reactor and its attendant analytical instrumentation. This equipment is described later in this report.

In addition to this, a state-of-the-art hydrogen chemisorption instrument has been designed and the components for its construction have been ordered.

Additionally, four recent publications by the principal investigators on the subject of this project have appeared.

Foley, H. C., Hong, A. J., Brinen, J. S., Allard, L. F., and Garratt-Reed, A. J., "Bimetallic Catalysts Comprised of Dissimilar Metals for the Reduction of Carbon Monoxide with Hydrogen", Applied Catalysis, 61, 351-375 (1990).

Mills, G. A., "Improving Perspectives for Oxy-Fuels." Proceedings Seventh Annual Pittsburgh Coal Conference, 605, (Sept. 1990) Presented and published.

Hu, X-D; Foley, H. C.; and Stiles, A. B., "Design of Alcohol Synthesis Catalysts Assisted By a Knowledge-Based Expert-System," Industrial and Engineering Chemistry Research, (1990) accepted for publication.

Bhore, N. A., Bischoff, K. B., Manogue, W. H. and Mills, G. A., "Carbon Monoxide Hydrogenation over Rh-Molybdena-Alumina Catalysts: Selectivity Control Using Activation Energy Differences." in Novel Materials in Heterogeneous Catalysis. Baker, T. K. and Murrell, L. L., Eds., ACS Symposium Series, 437, Ch. 23, p. 256-264 (1990).

Flow Microreactor

Figures 1 and 2 show the flow diagram of the reactor. The details of all the pieces of equipment used in this reactor assembly are given in Table 1. There are four feed gases. Three of them are high pressure lines while the fourth is a low pressure gas line for test reactions. The design pressure is 1000 psia. The gases are connected to flow limit valves to shut off the gases if the instantaneous flow rate exceeded a set limit. The hydrogen gas is passed through a filter, a line regulator, and a catalytic oxygen remover followed by a molecular sieve trap for removing water. Hydrogen is then passed through a filter, digital mass flow controller, and a check valve before feeding it to the mixing manifold. The line filter before the mass flow controller prevents fine dust from entering the mass flow controller, and the check flow valve prevents back flow from the manifold.

The connection between the hydrogen line and manifold is controlled by a diaphragm valve, which gives good service for repeated use at high pressures. The helium line is similar to the hydrogen line except that the oxygen and the water are simultaneously removed in a water trap. The CO manifold has a carbonyl trap that cracks the Fe and Ni carbonyls formed in the cylinder. Initially, a heated charcoal trap was used; however, it formed a lot of carbon dioxide. Later, a heated alkalized alumina trap was successfully used. This is followed by a water trap. The carbon monoxide line after the trap is Teflon® coated to prevent formation of carbonyls in the line. The gas manifold has ten openings. The pressure transducer and the rupture disc are located on the manifold. The low pressure gas line is also hooked up to the manifold. The entire reactor assembly and the analytical system is placed within a hood.

Equipment	Model	Purchased from
Pressure Transducer	PLC, 0-1000 psia	Test Equipment
Back Pressure Regulator	Mity-Mite, SD-91W	Test Equipment
Mass Flow Controller	High Pressure - 5850	Brooks
Line Filters	6184-p4FF	Matheson
Flow Limit Valves	Model LV	Veriflo
Hydrogen Purifier	Model B8087	Engelhard
Oxygen Water Remover	Model 98	AIRCO
Rupture Disc	Model 10-61AF4	High Pres. Equip.
Diaphragm Valves	SS-4D4S	Swagelok
Check Valves	SS-2C-10	Swagelok
Water Remover	SG-6140-2	Linde
Aluminized Reactors	Custom-Made	Alloy Surfaces

The reactor is made of internally aluminized stainless steel. The details are given in Table 1. Empty reactor runs under similar reaction conditions do not show any reaction of carbon monoxide and hydrogen. However, the presence of secondary reactions cannot be ruled out. The reactor is periodically checked for disruption of the aluminized layer, and replaced whenever necessary. The reactor effluent is then passed through a high temperature valve and the back pressure regulator. All reactor effluent lines are heated to 120°C to prevent condensation of products. A simple vapor pressure calculation of expected product distribution shows that under the low conversions used there is no appreciable condensation. The pressure of the gas manifold is controlled by the pressure on the nitrogen on the other side of the diaphragm. A separate air line is connected to an air-actuated sampling valve in the analytical system. Here again, the interconnecting lines are heated.

The reactor is connected to the manifold through a Teflon® coated stainless steel line. The reactor consists of two sections (Figure 3). The top section has an internal diameter of 1/4 inch, while the internal diameter of the bottom section is 1/8 inch. The glass wool was seated at the cone shape section connecting the top and the bottom section. Typically, the height of the glass wool would range from 1/4-1/2 inch. The catalyst is then seated over the glass wool. Typical catalyst loading is 0.5-1.0 gm and typical height of the catalyst bed is 1-2 cm.

The reactor has been completely overhauled since the beginning of this project. A major change is the addition of a set of high pressure liquid traps that can be held at low temperatures to collect liquid alcohol, hydrocarbon and other products. The traps are arranged with three vessels (500 cm^3) in series. Furthermore, there are two sets of traps aligned in parallel so that for long run times, the product stream can be swung to a clean set of traps, and the first set can be emptied of product. We believe this will be a major benefit to completing our carbon balances in an integral analysis as well as to identify minor as well as major components of reaction.

In addition, a Hewlett-Packard g.c.- m.s. with a capillary column will be used for analysis of products both in real time and after the products are down loaded from the traps. This capability is a major improvement in our analytical capabilities. Along these lines two rebuilt gas chromatographs (one TCD and one FID) have been installed in the system. Calibration of the TCD is completed, while that for the FID is underway. This will provide us with a means to measure hydrogen consumption more accurately.

Our goals during the next quarter include testing the new reactor system for synthesis gas conversion and to begin construction of the hydrogen chemisorption apparatus. In the course of this effort the new Ph.D. student will be trained to carry out this research program.

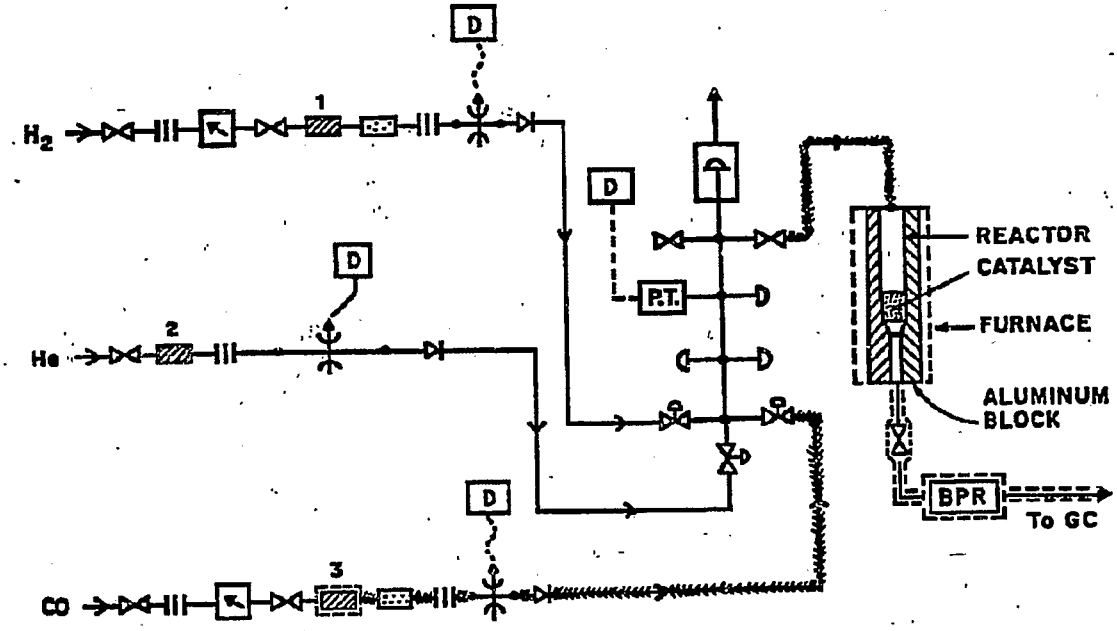

Figure 1: Flow diagram of the high pressure reactor

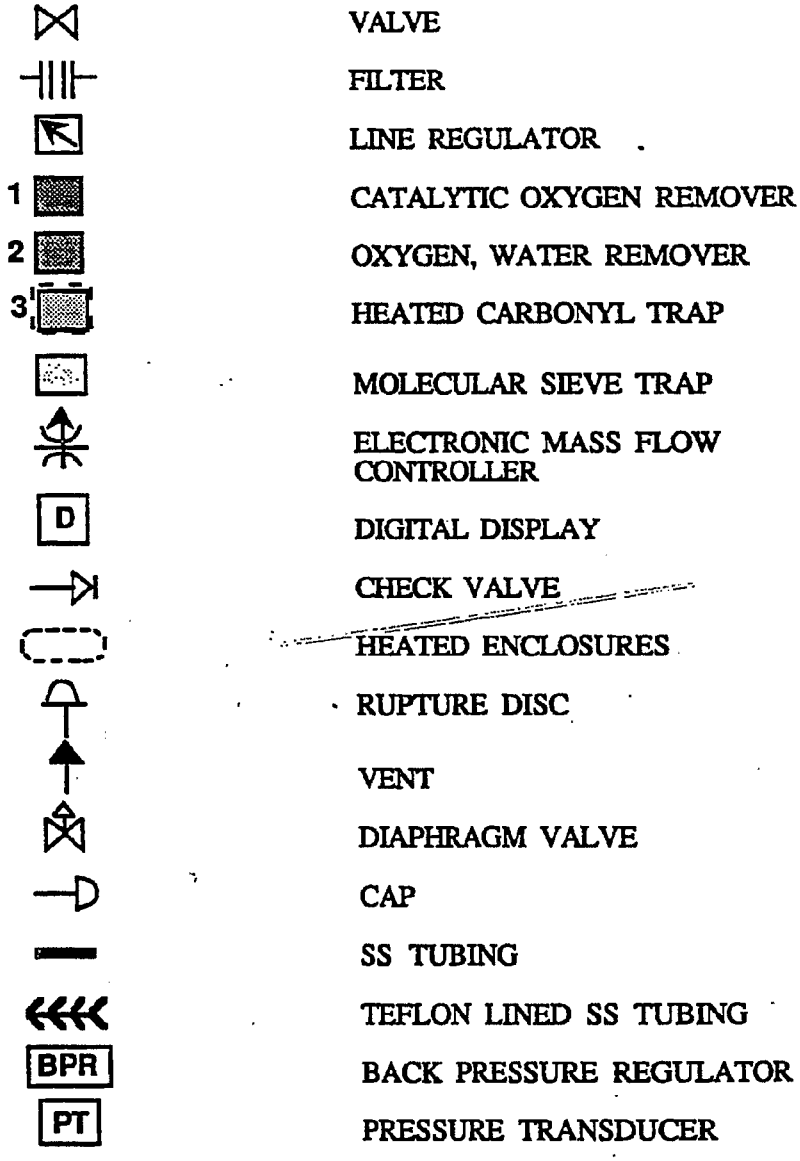

Figure 2: Key to the flow diagram of the high pressure reactor given in Figure 1

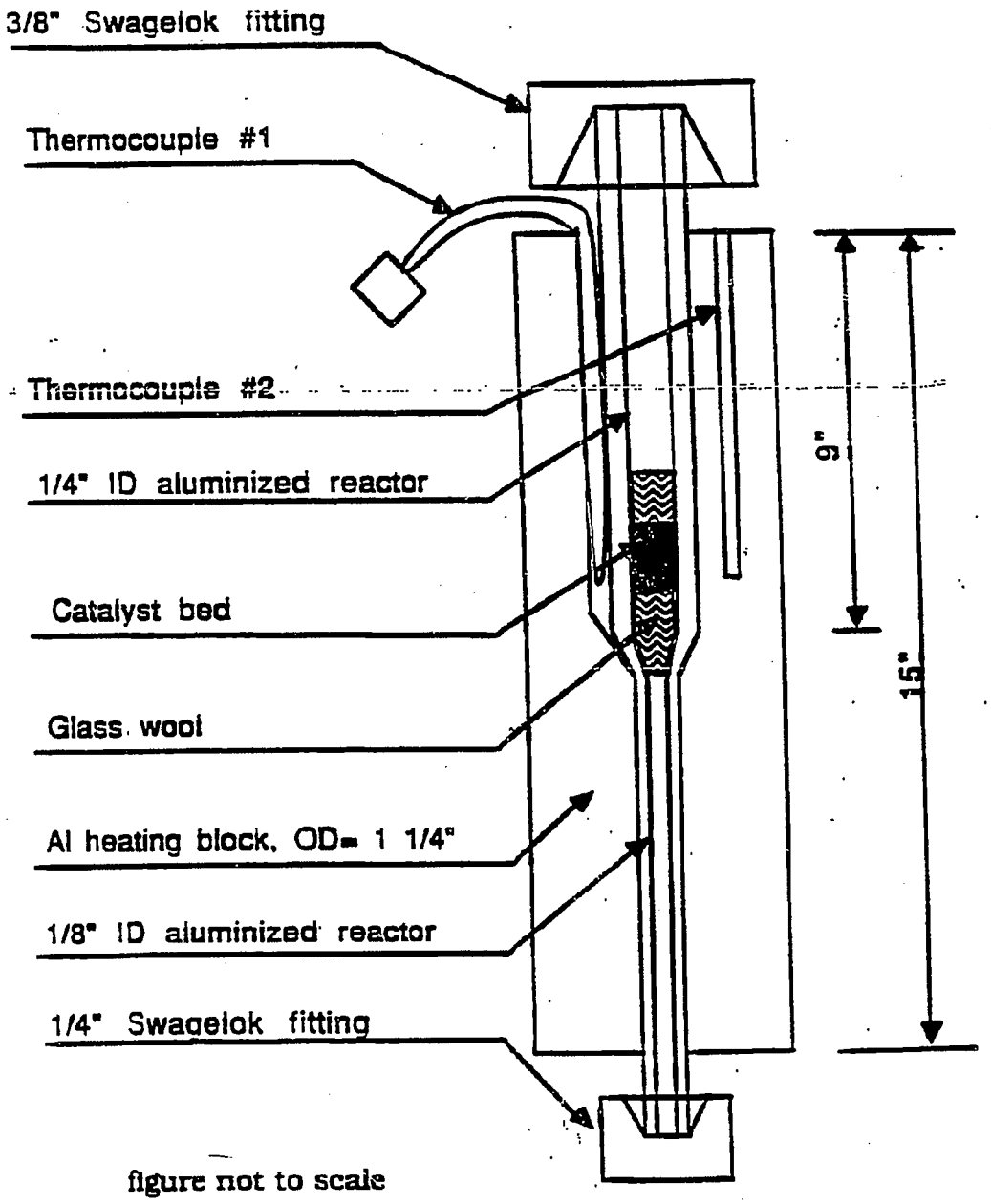

Figure 3: Cross-sectional view of the high pressure reactor assembly

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