MONTANA STATE UNIVERSITY

Catalytic hydrotreating of solvent refined coal (SRC-II)
by An-Gong Yeh

A thesis submitted in partial fulfillment of the requirements for the degree of MASTER OF SCIENCE in Chemical Engineering
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Abstract:
Solvent Refined Coal (SRC-II) from Pittsburg and Midway Coal Mining Company's pilot plant was hydrotreated with commercial and Montana State University developed catalysts. Twenty-two batch autoclave runs and twenty-five continuous trickle bed reactor runs were performed.

The liquid products were analyzed for sulfur and nitrogen content, and the extent of hydrocracking was determined by ASTM-D86 distillation test.

Nitrogen and sulfur content was decreased to meet the requirements, 0.3wt%. The catalyst lasted three hours before carbon laid down on the preheat section caused shut-down.

The study of catalyst- base properties and metal loading was performed to determine the effects of pore diameter, pore volume, surface area, and MoO3 concentration on the nitrogen removal. The higher surface area gave the better nitrogen removal. However, the smallest surface area with a large median pore diameter was not the poorest performer. An optimum combination of proper surface area and pore diameter seems important. The effect of MoO3 concentration on nitrogen removal is dependent on the catalyst base used, but it is insignificant compared with the effect of catalyst base. The larger pore volume base gave the higher liquid product yield.

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Date *November 28, 1979*

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by

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A thesis submitted in partial fulfillment
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of

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# ABSTRACT

Solvent Refined Coal (SRC-II) from Pittsburg and Midway Coal Mining Company's pilot plant was hydrotreated with commercial and Montana State University developed catalysts. Twenty-two batch autoclave runs and twenty-five continuous trickle bed reactor runs were performed.

The liquid products were analyzed for sulfur and nitrogen content, and the extent of hydrocracking was determined by ASTM-D86 distillation test.

Nitrogen and sulfur content was decreased to meet the requirements, 0.3wt%. The catalyst lasted three hours before carbon laid down on the preheat section caused shut-down.

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# INTRODUCTION

In view of energy crisis and national energy policy, it seems clear that sooner or later the United States will come to rely much more on coal as a resource of energy than it has over the past few decades. It is estimated that coal accounts for 80 percent of the fossil-fuel resources in the U.S.(1). In contrast, for the past decade or so the sources of energy in the U.S. have been predominantly oil and gas (44 and 31 percent respectively), with coal accounting for 21 percent(2). Coal is not the ideal fuel both because it is not a fluid and causes air pollution. Therefore, the development of a technology that will convert the U.S.'s abundant reserves of coal to clean fluid fuels is needed.

Coal conversion processes include gasifications and liquefactions. Since the shortage of domestic liquid hydrocarbons has caused the balance-of-payments problem in the U.S., coal liquefaction schemes are being examined closely. There are three major ways to turn coal into liquid fuels : pyrolysis, indirect liquefaction and direct hydroliquefaction. So far, most pyrolysis processes haven't been too suitable for making liquid fuels. Although indirect coal technology is in a more advanced state of development, direct hydroliquefaction offers, at least in theory, better economics and higher efficiency in terms of liquids per ton of coal. Therefore, most federal support is going to the direct processes.

Several direct hydroliquefaction processes have been developed such as the Solvent Refined Coal (SRC) process, the Exxon Donor Solvent (EDS) process and the H-Coal process. SRC process is the oldest of these modern processes dating back to 1962. Its original process is known as SRC-I, a later modified version is SRC-II process. Conceivably, its commercial scale plant could be in operation by 1989 or 1990(1).

The product of SRC-II process still cannot be used as a clean fuel at present costs, it must be catalytically upgraded or hydrorefined(3). This research is the second step of SRC-II process. The SRC-II product must be catalytically hydrotreated in a trickle bed reactor to remove unfavorable hetroatom molecules, sulfur and especially nitrogen, and to improve the overall product. This research is expected to provide the technology for the rapid commercialization of the SRC-II process and give the SRC-II process greater advantages over other processing schemes.

## BACKGROUND

### SRC-II Process

Of major concern to this research is the SRC-II process operated by Pittsburg and Midway Coal Mining Company. A fifty ton per day pilot plant is being operated at Fort Lewis, Washington. Pulverized raw coal is mixed with a process-derived slurry product and hydrogen at high temperature and pressure. The coal dissolves; most of its ash and much of its sulfur settle out and can be removed by filtration. Most of the coal is converted to liquids; naphtha, boiler fuel and vacuum residue. This residue contains heavy oil, ash, and undissolved organic material from coal(4). A schematic diagram of the SRC-II process is shown in Figure 1(5).

### Chemical Properties of SRC-II Products

The SRC process is not defined as a single product process. The gas and liquid yields per ton of Solvent Refined Coal is shown in Table I(6). Table II(7) presents the analysis of SRC-II product obtained in this research. The SRC-II product shown was made from

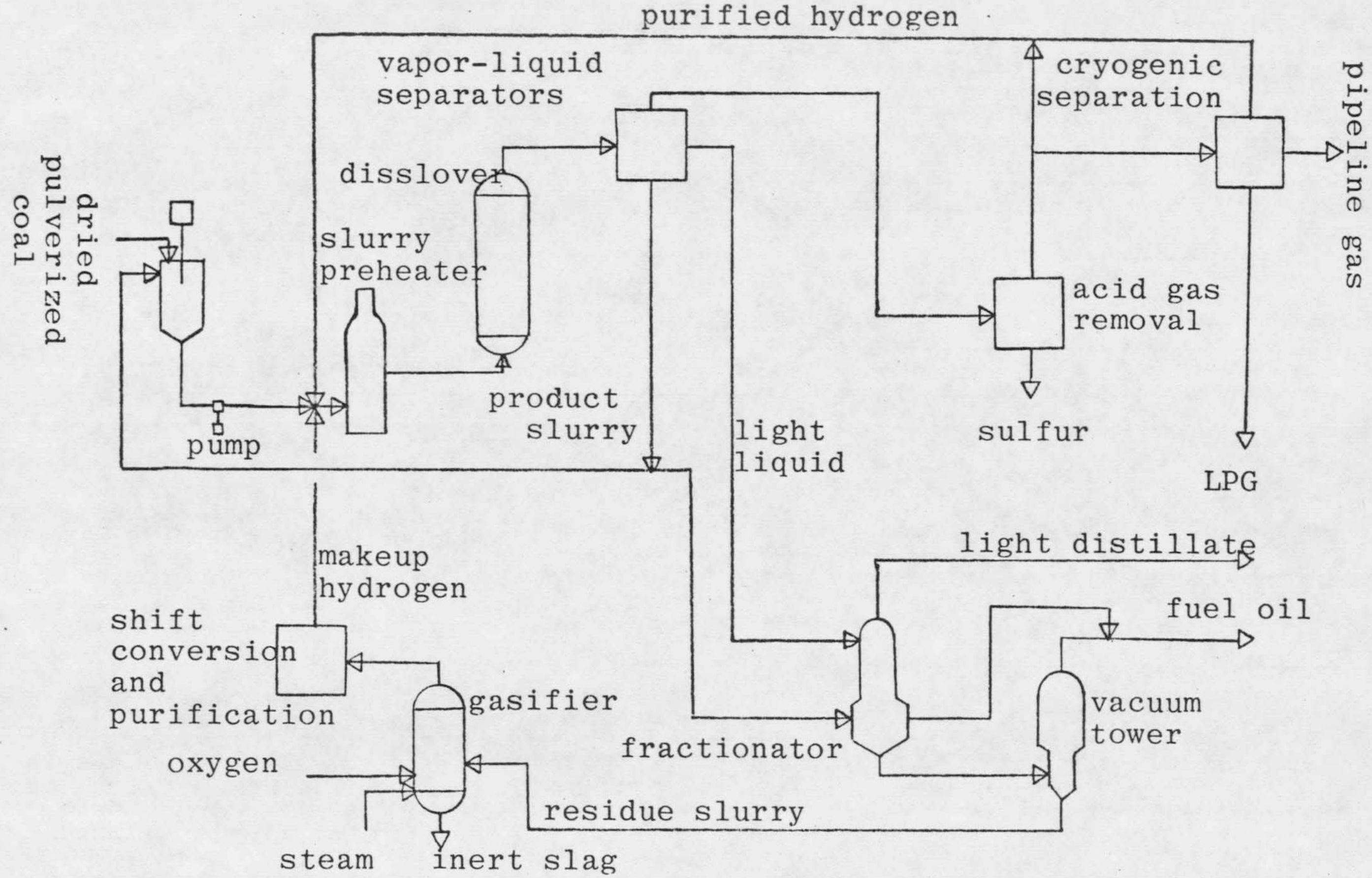


FIGURE 1. SRC-II PROCESS SCHEMATIC

TABLE I

SRC Process Gas and Liquid Yields*

| | | |
|---|---|---|
| $C_1 - C_4$ gas, scf | | 3130 |
| $CH_4$ gas | | 2100 |
| $C_5$ - 350°F gal | | 32 |
| bbl | | 0.762 |
| 350-750°F distillate, gal | | 38 |
| bbl | | 0.904 |
| Total liquid, gal | | 70 |
| bbl | | 1.666 |

Approximate analysis of $C_1 - C_4$ gas cut:

| | Vol.% | BTU value/ft$^3$ |
|---|---|---|
| $CH_4$ | 67.0 | 680 |
| $C_2H_6$ | 19.3 | 340 |
| $C_3H_8$ | 10.0 | 260 |
| $C_4H_{10}$ | 3.7 | 120 |
| | 100.0 | 1400 |

* Per ton solvent refined coal