



Catalysts for hydrotreating solvent refined lignite
by Nam Kyun Kim

A thesis submitted in partial fulfillment of the requirements for the degree of MASTER OF SCIENCE
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Abstract:

Catalytic hydrogenation of the solvent refined lignite (SRL) made by the University of North Dakota's Process Development Unit was carried out using thirty MSU-fabricated catalysts. The solid SRL was dissolved in tetralin to use as the feed for the trickle bed continuous reactor. A total of 42 tests were performed including three commercial catalysts and four blank carriers.

The liquid products from the reactor were analyzed for nitrogen content. Sulfur content and distillate yield were also analyzed for the selected composite samples.

MSU KN CS catalyst with 12.08% MoO₃ impregnated on Nalco C base showed the best performance in denitrogen-ation. The base comprised of 2%SiO₂ and 98% Al₂O₃ has a pore volume of 0.84 cc/gm base and a surface area of 215 m²/gm base.

The effect of MoO₃ concentration and additional active metal concentration on denitrogenation were studied. Increased MoO₃ concentration of up to 12% by weight improved nitrogen removal. This optimum amount of metal impregnation was also explained by the covalent bond radius and by the visual aid of electron photomicrographs .

The two consecutive thermal regenerations of the best catalyst was completed. The results showed that the catalytic performance can be fully restored.

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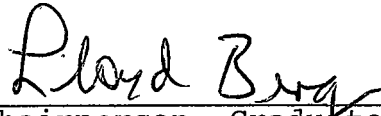
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ABSTRACT

Catalytic hydrogenation of the solvent refined lignite(SRL) made by the University of North Dakota's Process Development Unit was carried out using thirty MSU-fabricated catalysts. The solid SRL was dissolved in tetralin to use as the feed for the trickle bed continuous reactor. A total of 42 tests were performed including three commercial catalysts and four blank carriers.

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The two consecutive thermal regenerations of the best catalyst was completed. The results showed that the catalytic performance can be fully restored.

INTRODUCTION

National recognition of dwindling petroleum resources and increasing dependency of our energy needs on foreign oil accelerated our effort to find alternative energy sources. Although the United States has more energy available in the form of coal than in the combined sources of petroleum, natural gas, oil shale and tar sands, the primary use of this solid fuel has been limited to the electric power generation where coal was pulverized and burned in solid form in boilers.

Our abundant coal reserve is immediately available for commercial production of synthetic fuel, "synfuel," from coal to meet our national demand for clean energy. President Carter has signed the Synthetic Fuel Corp. legislation, a mandate to produce the equivalent of 500,000 barrels per day (bbl/d) of synfuel by 1987 and 2 million bbl/d by 2000 (1,2). The \$20-billion synfuel program, the largest federal energy-assistance effort in history, is designed to encourage the development of oil shale and coal technologies as well as biomass conversion, alcohol fuels production and recovery of heavy oil, tar sands, and gas from unconventional sources.

The coal must be processed into the desirable products that can substitute for those derived from oil and natural gas. These substitute fuels include crude oils, fuel oil, and distillate; chemical feedstocks; pipeline quality and fuel gas; and other solid products such as char and SRL that may be useful in energy production(3).

Numbers of coal conversion processes and technologies have been developed, applied to various domestic coals, and proved to be potential for the commercial production. These conversion processes can be divided into three major categories:

- (1) solid clean fuel (SRL, SRC-I, and char)
- (2) liquid fuel (direct:SRC-II, EDS, H-coal and etc.; indirect:Fischer-Tropsch, methanol, M-gasoline, and etc.)
- (3) gas fuel (CO_2 acceptor, Hygas, Synthane, Bigas, Lurgi process, flash pyrolysis, and etc.) (4)

At present there are many processes that can be put into a commercial demonstration based upon the abundant technological data but the manufacturing cost appears to be yet greater than those for conventional petroleum fuel.

The current war in the Gulf and steadily increasing trend of the oil prices forced us to secure commercial mass production of synfuel from coal today. But the technology is available and all of these coal conversion processes could be viable as the cost of imported crude oil continues to rise.

BACKGROUND

Coal is readily combustible rock containing more than 50% by weight and more than 70% by volume of carbonaceous material including inherent moisture, formed from compaction and induration of variously altered plant remains similar to those in peat. The fermentation of vegetable matter under conditions of no air and abundant moisture where volatiles are retained, resulting in formation of bituminous, such as peat and coal, is known as bituminous fermentation.

The peat-to-anthracite theory of coal formation is described as a process in which the progressive ranks of coal are indicative(4).

According to the most recent estimates by the U.S. Geological Survey, the lignite shares a 6% of the demonstrated reserve base (437 billion tons) which is approximately 25% of the identified resources.

The lignite in the Northern Great Plains occurs in relatively thick seams ranging from 5 ft to more than 100 ft and typically with overburden from 50 to 200 ft (5) (Figure 1).

Significant sulfur and nitrogen reduction is obtained and the mineral substances are removed from

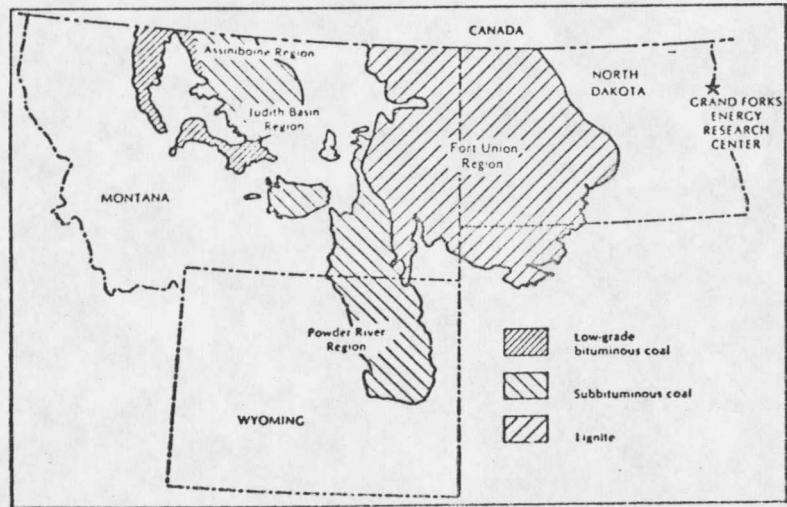


FIGURE 1. NORTHERN GREAT PLAINS COAL PROVINCE

SOURCE: GRAND FORKS ENERGY RESEARCH CENTER,
1978 PROGRAM REPORT

the liquid phase to a low concentration by the SRC-I process.

Project Lignite, equivalent to the SRC-I, was awarded to the University of North Dakota in 1972 for the purpose of determining the appropriate technological approach to the conversion of lignite to clean fuel. The project was expected to extend the two stage conversion of lignite to liquid fuel (equivalent to the SRC-II) with the SRL as an intermediate solid fuel. The first stage to convert lignite to the SRL has been successfully accomplished but the second stage that catalytically hydrotreat the SRL to the distillate fuel was not implemented(6).

Lignite

The classification system adopted by the American Society for Testing and Materials (ASTM) indicates that the lignite is the lowest rank of coals in terms of calorific value and carbonaceous content. These criteria are presented in Figure 2 (7): Lignite coal is dark brown substance that is intermediate in coalification between peat and subbituminous coal. It is consolidated coal with a calorific value less than

8,300 Btu per pound, on a moist, mineral-matter-free basis.

The U.S. Geological Survey established a coal resource base of 3.2 trillion tons, about half of which is considered as recoverable. Of this 1.6 trillion recoverable tons, approximately half (780 billion tons) was determined from actual mapping and exploration to depths of 3,000 ft below ground surface. Approximately 200 billion tons are projected as occurring in bituminous coal and anthracite beds 42 inches or more in thickness and in subbituminous coal and lignite beds of 10 ft or more in thickness. They are generally less than 1000 ft below the surface. Approximately one-half of the 200 billion tons is based on a relative abundance of information(8). A summary of recoverable coal reserves is shown in Table I by rank.

