



The effect of hydrogenated creosote oil as a hydrogen donor agent on upgrading of solvent refined coal (SRC-II)  
by Chia-Ren Jack Pan

A thesis submitted in partial fulfillment of the requirements for the degree of Master of Science in  
Chemical Engineering  
Montana State University  
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**Abstract:**

The effect of partially hydrogenated creosote oil as a hydrogen donor agent on catalytic upgrading of SRC-II was investigated. Creosote oil is a coal-derived solvent used as a start-up solvent in the Solvent Refined Coal process.

The hydrogenated creosote oil was added in three different ratios to SRC-II Light End Column Feed product: 25%:75%, 50 % : 50%, 75%:25%. MSU-C-49 catalyst which was developed at Montana State University, and two commercial catalysts, Shell NM324 and Harshaw HT-400, were evaluated in a small scale trickle bed reactor.

The effect of high concentration of creosote oil on catalytic upgrading of SRC-II is nonbeneficial. As the percentage of creosote oil is increased the activity in denitrogenation and desulfurization decreased. Harshaw HT-400 is more active for denitrogenation and desulfurization than either Shell NM324 or MSU-C-49.

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of a thesis submitted by

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## ABSTRACT

The effect of partially hydrogenated creosote oil as a hydrogen donor agent on catalytic upgrading of SRC-II was investigated. Creosote oil is a coal-derived solvent used as a start-up solvent in the Solvent Refined Coal process.

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## CHAPTER I

## INTRODUCTION

The thirty years of success in coal liquefaction technology has demonstrated coal's feasibility as a new energy source. However, no commercial plant has been constructed since the technology has become a reality. Politics, economics and environmental considerations have influenced its progress.

The most efficient use of coal is when it is burned directly in furnaces or under boilers. The problem of combustion is the emission of noxious gases such as sulfur oxides and nitrogen oxides. Indeed, this concern was one reason for initiating the shift from coal to oil for electrical power generation.

Liquifying coal has been proved to be the most efficient way(1) to provide liquid fuel from coal. However, liquified coal has too high a sulfur and nitrogen content to be accepted either by the EPA(2,3) or in conventional petrochemical refining processes. The petroleum refining industries do not accept liquified coal because of equipment and pollution problems. Nitrogen contained in the feedstock for Fluid Catalytic Cracking(FCC) would be a major source of pollution and equipment damage.

Hydrogen blistering is an example of the extensive damage possible in equipment. Another important reason for the upgrading of coal liquids is that high nitrogen and sulfur content in organic compounds cause undesirable poisoning of the acidic functions of catalysts in refining processes (5). Poisoning shortens catalyst life and increases the cost of investment. From economic considerations, price determines the feasibility of substituting coal liquid for petroleum. The upgrading of coal liquids became a necessary procedure if the liquids are to be used as substitutes for petroleum.

The purpose of this research is to investigate the effect of different catalysts on the upgrading of Solvent Refined Coal(SRC-II) by the addition of hydrogenated creosote oil. The creosote oil acts as a hydrogen donor agent.

## CHAPTER II

## COAL CONVERSION

Coal has been used in many different ways for more than a century. In the beginning it was used primarily for heat by combustion. Direct combustion is the most straightforward and convenient use of coal. Coal gasification was introduced as a substitute for natural gas. However, the energy supply and the resource structure has changed over the years. It was not until World War II when people needed much more energy. Because of the petroleum shortage, coal again became an important energy resource. It was during World War II that the non-petroleum producing countries were forced to develop a substitute for petroleum. However, consumers currently use about 75% of their petroleum products in the form of distillates(6). The enormous consumption of distillates in transportation and heating fuel has become a major reason to the liquify coal.

## Coal Liquefaction

Coal is converted to a liquid product through either direct or indirect liquefaction techniques. Indirect liquefaction is the most developed. First, coal is gasi-

fied to produce a mixture of hydrogen and carbon monoxide which is then combined by catalysts to form liquid compounds. Currently, the only commercial process is the Fisher-Tropsch being used in South Africa in conjunction with Koppers-Totzek, Texaco, Winkler-Lurgi and Shell-Koppers gasifiers.

Two different processes of direct liquefaction are catalytic hydrogenation and the use of a donor solvent. The latter was characterized as the Solvent Refined Coal (SRC) process developed by Pittsburg and Midway Coal Co. and the Exxon Donor Solvent(EDS) process. Both processes produce liquified coal by solvent extraction.

Direct catalytic hydrogenation essentially reduces molecular weight and increases the H/C ratio of the parent coal molecule to a ratio similar to that of a petroleum crude. The process used catalysts comprising cobalt and molybdenum oxide on alumina or molten  $ZnCl_2$ (5). The Synthoil and H-coal processes are examples of direct catalytic hydrogenation.

#### Solvent Refined Coal (SRC)

In 1974, Pittsburg & Midway Coal Mining Company owned by the Gulf Oil Company was awarded a contract by the Department of Energy(DOE) for the SRC-I pilot plant start-up operation. Figure 1 shows the flow scheme of the SRC-I process. SRC-I was designed to produce low ash and low

sulfur solid boiler fuel(7) to meet the sulfur emission requirement. The pulverized coal and anthracene oil was introduced into a slurry tank and pumped to the slurry preheater, where slurry was preheated to above 800°F at 100 atm. The preheated slurry was fed to a dissolver and the solvent in the slurry from the dissolver was recovered as a recycle stream which went back to the slurry tank. The SRC-II mode was connected to SRC-I in 1977(8). Figure 2 shows the SRC-II flow scheme. The final product in SRC-II is liquid at room temperature. This makes SRC-II considerably different from SRC-I. The overall yield and hydrogen consumption of the SRC-II process are given in Table I. The feed for this research is Light End Column Feed(LECF), a light liquid from the vapor-liquid separators of the SRC-II process.



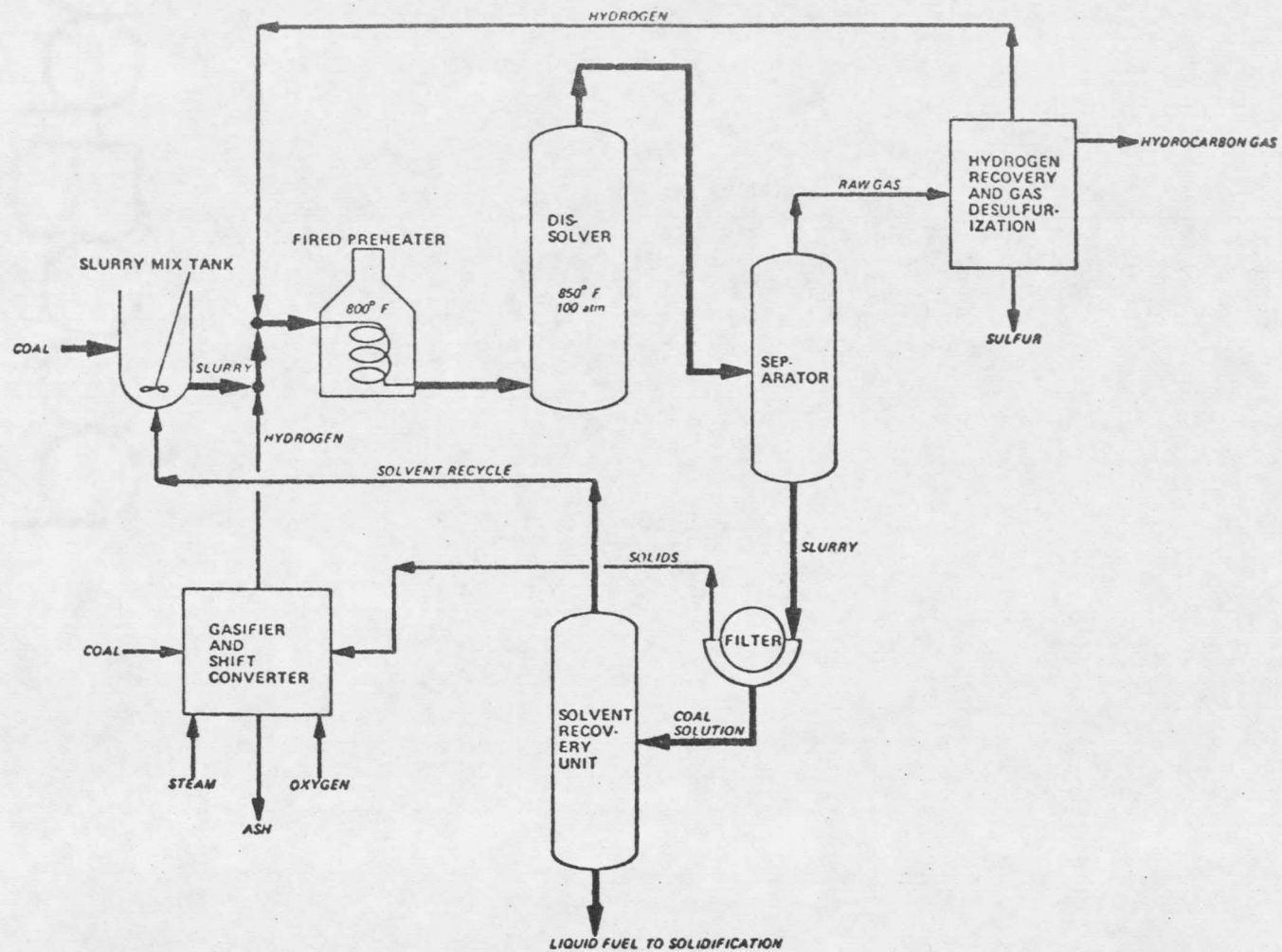


Figure 1. Flow Scheme of SRC-I Process.

































































































































