LOW-TEMPERATURE COAL
DEPOLYMERIZATION AND
LIQUEFACTION

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LOW-TEMPERATURE COAL DEPOLYMERIZATION AND LIQUEFACTION

by

Tanakom Skulthai

A dissertation submitted to the faculty of
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Department of Fuels Engineering

The University of Utah

December 1987

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ABSTRACT

A new approach to low-temperature (≤ 290°C) coal depolymerization was developed. It involves the application of several consecutive reaction steps in which different types of intercluster linkages in the coal structure are subjected to selective or preferential cleavage. The sequential depolymerization-liquefaction procedure consists of the following three steps: (1) deep-seated impregnation of the coal sample with catalytic amounts (5-20 %) of a metal halide, in particular $FeCl_3$, followed by mild hydrotreatment (HT) of the coal-metal halide impregnate at 250-275°C and 1500 psig H_2 initial pressure; (2) base-catalyzed depolymerization (BCD) of the product from step 1, under supercritical conditions, using a methanolic KOH or NaOH solution as catalyst-solvent depolymerizing agent; and (3) hydroprocessing (HPR) of the depolymerized product from the two preceding steps with a sulfided CoMo catalyst.

Four U.S. coals of various rank, e.g., a subbituminous Wyodak (Wyoming) coal, and three different HVB coals from Fruitland (New Mexico), Helper (Utah), Burning Star (Illinois #6) were subjected to systematic studies using the above depolymerization-liquefaction procedure. The optimal conditions for each step of the procedure for the above coal feeds were determined. Products obtained were analyzed by a combination of FTIR, ^{13}C NMR, PMR, elemental analysis, and simulated distillation.

With the Wyodak coal as feed, suitable HT conditions were found

to be; 20 % $FeCl_3$ concentration, 275°C HT temperature, 1500 psig H_2 pressure, and 3 hr reaction time. BCD of the above mildly hydrotreated coal at 275°C, using a 10 % methanolic KOH solution, 1:1 feed to catalyst weight ratio, 1000 psig N_2 pressure, and 1 hr reaction time, resulted in nearly complete solubilization of the product in THF (90.2 % by wt on a MAF basis). Hydroprocessing (HPR) of the solubilized (depolymerized) coal with a sulfided 6Co8Mo catalyst for 8 hr at 370°C, using 5:1 feed to catalyst weight ratio, and 2700 psig H_2 pressure, yielded a light hydrocarbon oil (C,87.57; H,11.60; O,0.59; and N,0.24 % by wt), which contained 71.3 % by wt of low-boiling fractions. Similar results were obtained with the three HVB coals as feeds.

Parallel to the above investigation, relevant model compound studies were performed under conditions simulating the HT-BCD-HPR procedure, using diphenyl ether, 1- and 2-methoxynaphthalene, benzyl phenyl ether, dibenzofuran, and phenanthrene as reactants.

On the basis of the analysis of coal-derived products and the model compound studies, it was concluded that step 1 (HT step), results in partial depolymerization of the coal by preferential hydrogenolytic cleavage of methylene, benzyletheric and activated aryletheric linkages in the coal framework, while step 2 (BCD step) completes the depolymerization of the product from step 1 by base-catalyzed hydrolysis (or alcoholysis) of diaryl etheric, and other bridging groups. In the HPR step 3 the depolymerized product undergoes exhaustive heteroatom removal and attendant partial hydrogenation and C-C hydrogenolysis reactions to yield a light hydrocarbon oil.

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