

A Two-Step, Temperature-Staged, Direct Coal Liquefaction Process

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Abstract : The world crude oil demand is projected to rise to 108.5 million bbl/d by the year 2035. With reserves estimated at 869 billion tonnes worldwide, coal is an abundant resource. This work was aimed at producing a high value hydrocarbon liquid product from the Direct Coal Liquefaction (DCL) process at, comparatively, mild operating conditions. Via hydrogenation, the temperature-staged approach was investigated. In a two reactor lab-scale pilot plant facility, the objectives included maximising thermal dissolution of the coal in the presence of a hydrogen donor solvent in the first stage, subsequently promoting hydrogen saturation and hydrodesulphurization (HDS) performance in the second. The feed slurry consisted of high grade, pulverized bituminous coal on a moisture-free basis with a size fraction of < 100µm; and Tetralin mixed in 2:1 and 3:1 solvent/coal ratios. Magnetite (Fe₃O₄) at 0.25wt% of the dry coal feed was added for the catalysed runs. For both stages, hydrogen gas was used to maintain a system pressure of 100barg. In the first stage, temperatures of 250°C and 300°C, reaction times of 30 and 60 minutes were investigated in an agitated batch reactor. The first stage liquid product was pumped into the second stage vertical reactor, which was designed to counter-currently contact the hydrogen rich gas stream and incoming liquid flow in the fixed catalyst bed. Two commercial hydrotreating catalysts; Cobalt-Molybdenum (CoMo) and Nickel-Molybdenum (NiMo); were compared in terms of their conversion, selectivity and HDS performance at temperatures 50°C higher than the respective first stage tests. The catalysts were activated at 300°C with a hydrogen flowrate of approximately 10 ml/min prior to the testing. A gas-liquid separator at the outlet of the reactor ensured that the gas was exhausted to the online VARIOplus gas analyser. The liquid was collected and sampled for analysis using Gas Chromatography-Mass Spectrometry (GC-MS). Internal standard quantification methods for the sulphur content, the BTX (benzene, toluene, and xylene) and alkene quality; alkanes and polycyclic aromatic hydrocarbon (PAH) compounds in the liquid products were guided by ASTM standards of practice for hydrocarbon analysis. In the first stage, using a 2:1 solvent/coal ratio, an increased coal to liquid conversion was favoured by a lower operating temperature of 250°C, 60 minutes and a system catalysed by magnetite. Tetralin functioned effectively as the hydrogen donor solvent. A 3:1 ratio favoured increased concentrations of the long chain alkanes undecane and dodecane, unsaturated alkenes octene and nonene and PAH compounds such as indene. The second stage product distribution showed an increase in the BTX quality of the liquid product, branched chain alkanes and a reduction in the sulphur concentration. As an HDS performer and selectivity to the production of long and branched chain alkanes, NiMo performed better than CoMo. CoMo is selective to a higher concentration of cyclohexane. For 16 days on stream each, NiMo had a higher activity than CoMo. The potential to cover the demand for low-sulphur, crude diesel and solvents from the production of high value hydrocarbon liquid in the said process, is thus demonstrated.

Keywords : catalyst, coal, liquefaction, temperature-staged

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