PROCESS DESIGN OF A NOVEL CONTINUOUS-MODE MINI-PILOT PLANT, FOR DIRECT LIQUEFACTION OF COAL AND UPGRADING OF COAL LIQUIDS

P. Vijay, Chunshan Song*, and Harold H. Schobert

Fuel Science Program
Dept. of Materials Science and Engineering
209 Academic Projects Bldg.
The Pennsylvania State University
University Park, PA 16802-2303

ABSTRACT

On the basis of existing coal liquefaction process units, and our own laboratory research efforts on microautoclave reactor systems, a continuous flow process reactor system for the direct liquefaction of coal and upgrading of coal liquids was designed. In our mini-pilot plant scale process system, two novel technologies have been coupled to improve the economics over existing coal liquefaction processes. The "technologies" incorporated are temperature programmed liquefaction (TPL) of coal and the co-use of water and dispersed metal catalyst in the low-temperature catalytic coal conversion. Details of this continuous-mode process reactor scheme along with its peripheral units will be presented. The progress of this ongoing project will be updated and presented.

Keywords: Direct Liquefaction of Coal, Upgrading of Coal Liquids, and Continuous Flow Process Reactor System.

INTRODUCTION

Extensive research studies on various aspects of the liquefaction of coal including temperature programmed liquefaction (TPL), and effect of water and dispersed molybdenum catalyst for promoting low-severity liquefaction, in the presence of a solvent were conducted earlier [1, 2]. All these batch-mode experiments were successfully conducted in a 25 mL tubing bomb microautoclave reactor. Coal liquefaction experiments in the presence of dispersed catalyst, water, and solvent are currently being performed in a 300 mL batch-mode reactor system.

In order to simulate conditions prevalent in an industrial-type reaction atmosphere, it is necessary to conduct liquefaction of coal and upgrading of coal liquids in a continuous-mode reactor system. Besides, in this system the dispersed catalyst moves with the products rather than staying in the reactor for extended periods, thus enhancing the once-through reactor productivity. The reactor system can be operated in a continuous-mode for a longer time, leading to an increase in the quantity of the products obtained. If the coal liquefaction products stay in the reactor for a prolonged period of time, retrograde reaction occurs that converts desirable small product molecules into less desirable larger macromolecules, by the recombination of small product molecules or by addition of these product molecules back onto the coal [3]. Since the coal liquefaction products that are formed are continuously removed from the flow reactor system, there would be an alleviation in the effect of retrograde reactions thereby increasing the yield and quality of coal liquids, that can be upgraded to produce transportation fuels, particularly jet fuels.

BRIEF OVERVIEW OF EXISTING COAL LIQUEFACTION UNITS

Three of the following well-documented liquefaction processes were studied, before embarking on the process design and assembly of our novel mini-pilot plant reactor system for liquefaction of coal and upgrading of coal liquids.

Advanced Coal Liquefaction Facility at Wilsonville, Alabama. The liquefaction facility at Wilsonville initially operated with two ebullated-bed reactors in series, with supported catalysts in both the reactors [4]. Due to the several well documented advantages of using dispersed catalysts in the 1st reactor [4], a few experiments using dispersed catalysts in the 1st reactor and supported catalysts in the 2nd reactor were conducted. This hybrid catalyst system improved the distillate production by 30-60 %, and increased the coal & resid conversions compared to using dispersed and supported catalysts separately. This Wilsonville Facility was closed down in 1992.

Hydrocarbon Research Institute (HRI) Inc. Unit, New Jersey. The reactor configuration in the HRI unit is similar to that in Wilsonville [5]. Since very few runs were performed in the Wilsonville facility in the hybrid catalyst mode, HRI, Inc. decided to conduct a few more runs with this reactor scheme. An interstage separator was added between the 2 reactors. Experiments were performed using both pure H_2 feed and syngas (H_2 + CO) feed to the 1st reactor, and pure H_2 feed to the 2nd reactor [5]. A higher distillate yield, coal, and residuum conversion were obtained when syngas was used in the 1st stage instead of H_2.

For a few runs an on-line hydrotreater was in service to further remove heteroatoms from the separator and atmospheric still overhead products. The coal conversions were the highest values obtained for this type of coal. The light distillates (IBF-650 °F) contained ten times less nitrogen

* To whom all correspondence should be addressed.

and 17 wt% more hydrogen than that of the 2 ebullated-bed reactors in series configuration. Other studies in the HRI Inc. unit included an addition of a fixed-bed finishing reactor (second or third stage) after the ebullated bed reactors, a three stage CSTR system with an addition of a third ebullated bed reactor in series along with 2 ebullated bed reactors (plug flow simulation), and lowering the solvent/coal ratio to 0.9 in two ebullated bed reactors in series [6].

Pittsburgh Energy Technology Center (PETC) Coal Liquefaction Unit. In PETC, coal liquefaction research studies are being conducted in a computer-controlled bench-scale continuous unit. In this process, coal-catalyst-solvent slurry continuously flows through a 1-liter reactor, where it reacts in the presence of hydrogen to generate coal liquids [7].

PROCESS DESIGN OF A NOVEL CONTINUOUS MODE MINI-PILOT PLANT FOR LIQUEFACTION OF COAL AND UPGRADING OF COAL LIQUIDS

Mini-Pilot Plant for Coal Liquefaction Process

On the basis of existing coal liquefaction units, and our laboratory research efforts in the 25 mL microautoclave reactor system and the 300 mL batch-mode reactor system, a novel continuous flow process scheme for the liquefaction of coal and upgrading of coal liquids was designed. The process flow diagram of this continuous-mode reactor scheme for the liquefaction of coal is presented in Figure 1. The typical operating conditions of the system are shown in Table 1.

The size of the coal particles is small (≤ 60-200 mesh) in order to reduce mass transfer limitations. The solvent used will either be a process solvent (e.g., Wilsonville middle distillates) or petroleum resids. The advantages of employing dispersed catalysts instead of supported catalysts in the coal liquefaction reactors include, a lesser amount of catalyst, good control of retrogressive reactions at preconversion conditions and hence reducing char formation and catalyst deactivation, reduced reaction severity, faster hydrogen transfer rate, improved economics due to enhanced yields of desired products, and higher coal conversion.

In our process, two novel technologies have been coupled in an attempt to improve the economics over existing coal liquefaction processes. The technologies incorporated are temperature programmed liquefaction (TPL) of coal [1] and the co-use of water and dispersed catalysts in the coal liquefaction process [2]. Studies conducted in the 25 mL microautoclave reactor and in the 300 mL batch-mode reactor scheme, will help to provide some basic information in guiding the selection of the various operating conditions for the continuous unit.

Features of the Continuous Flow Reactor Scheme. The significant features of this novel reactor scheme for the coal liquefaction process are:

§ Use of dispersed catalysts in the coal-solvent slurry.
§ Temperature-programmed liquefaction (TPL) in a multistage (2 stages) reactor scheme.
§ Option of adding water to the coal-catalyst-solvent slurry to the 1st stage reactor.
§ Incorporation of an interstage separator between the two stages of the reactor.
§ A feed gas compression system.
§ Continuous sampling and G.C. analysis of gas and liquid from the slurry lines at several vantage points to periodically monitor the progress of the reaction.

Details of the Multistage Continuous Flow Reactor System for Coal Liquefaction Process.

* Slurry Feed Mix Tank - Feed slurry to the reactors consisting of coal, dispersed catalyst, and solvent (or petroleum resids) will be thoroughly mixed in this 3 gallon feed mix tank equipped with an agitator, to maintain this slurry in suspension. The temperature and the pressure of this vessel will be maintained at around 80-100 °C and at 50-80 psi., respectively.

* Slurry Pump - The slurry from the feed mix tank will be fed to the reactors through a preheater by the slurry pump. This pump will be capable of handling high concentration of solids loading in slurry (35 wt %) and pressures as high as 4000 psi.

Prior to entering the preheater, the slurry will be contacted by pure feed gas (H_2 or syngas - $H_2 + CO$). CO will be used in an attempt to improve the economics by incorporating the water-gas shift step. Although it is necessary to recycle gas in an industrial-scale process, we have eliminated all recycle gas streams and the compressors in these lines in our design of the laboratory-scale reactor system, due to budget and space limitations.

* Feed Gas Compression System - The maximum pressure of the feed gas from the manufacturer's gas cylinders is 2500 psi. Since we will require a steady flow of feed gas at pressures around 2500 psi., it was necessary to design a feed gas compression system. The feed gas (H_2 or syngas - $H_2 + CO$) will flow from the gas cylinders into a pneumatic compressor, where it is compressed to the desired high pressure and stored in the compressed gas storage tanks. The pneumatic compressor will be capable of compressing the inlet feed gas to pressures as high as 5000 psi, depending on the inlet pressure and the desired exit flow rate to the compressed gas storage tanks.

The outlet lines from the compressed gas storage tanks are provided with a vent line, to depressurize the system at the completion of the experimental runs. The outlet from these storage tanks serves as the feed inlet to the reactor system. The feed gas inlet to the slurry line before the preheater, is equipped with a forward pressure regulator and a mass flow controller.

* Preheater - The gas and slurry that enters the 300 mL agitated autoclave vessel that serves as a preheater, will be maintained at a temperature of 200-300 °C. The preheated gas and slurry then flows into the 1st stage reactor.

* 1st Stage Coal Liquefaction Reactor - The 1st stage reactor is a 1-liter vessel equipped with a dispersimax turbine type impeller, that will be maintained at a temperature of 350-400 °C. The gas and slurry entering from the preheater, will undergo primary reactions here.

Provision is made for continuous injection of water from the water feed reservoir (3 gallon), into the 1st stage reactor, with the aid of a water pump. This water reacts with the gas and slurry to form primary products which includes the product vapors and the coal liquids which remains in the slurry. The unreacted gas, product vapors, and slurry (along with the product liquids) then flows into the interstage separator. A sampling loop for G.C. analysis of liquid is provided.

* Interstage Separator - This interstage separator is a 1-liter vessel equipped with a turbine type impeller, whose main function is to ensure the complete removal of water remaining in the system. The product vapors exiting this separator flows through a condenser to remove any condensables remaining in the system like H_2O, H_2S, and NH_3, that are collected in the 300 mL liquid collection bomb. The noncondensables (H_2, C_1-C_4) initially flows through a back pressure controller and a flow meter, and then vented to atmosphere. Provision is made for analysis of gas and liquid from this separator.

* 2nd Stage Coal Liquefaction Reactor - The process stream consisting of gas and slurry from the exit of the interstage separator, and pure gas from the compressed gas storage tank, is the feed to the 2nd stage reactor, which is a 1-liter vessel equipped with a turbine type impeller, and maintained at 400-440 °C. The gas and slurry entering this reactor undergo secondary reactions to form more coal liquids. The reactor effluent flows into a vapor-liquid separator. A sampling line for G.C. analysis of the liquid reacting in the 2nd stage reactor will be installed.

* Vapor-Liquid Separator - The gas and slurry products exiting the 2nd stage coal liquefaction reactor flows into the vapor-liquid separator, which is a 1-liter vessel equipped with a turbine type impeller. The gas exiting this separator is first contacted with water from the reservoir and then cooled by a condenser. The condensables fill the liquid collection bomb, which is a 300 mL cylinder. Provision is made for G.C. analysis of the liquid collected in this bomb. The noncondensables initially flows through a back pressure controller and a flow meter, and then vented to atmosphere. Provision is made for G.C. analysis of this gas exiting the V/L separator.

The gas and slurry exiting the vapor-liquid separator initially flows through a condenser, and is then sent through a Bureau of Mines (BOM) valve which will be capable of handling this slurry, at high temperature and high pressure process conditions. The process stream (gas and slurry) then flows into an atmospheric flash tank.

* Atmospheric Flash Tank - The effluent from the V/L separator enters the atmospheric flash tank which is a 1-liter agitated vessel. The flow rate of the gas exiting this vessel is measured and then vented to atmosphere. Periodic G.C. analysis of this exiting gas will be performed. The slurry exiting this atmospheric flash tank flows into a coal liquids collector tank.

* Coal Liquids Collector Tank / Coal Liquids Upgrading Feed Tank - The slurry exiting the atmospheric flash tank, enters a 3 gallon coal liquids collector tank through a filter bag placed in the process line. This would aid in the separation of solids from the liquid in the slurry stream. The liquids are collected in this tank, which is also the feed tank for the upgrading section of the mini-pilot plant system. The solids containing insolubles, unreacted coal, resids, and catalysts are removed. The coal liquid is then pumped to the upgrading section of the mini-pilot plant.

Mini-Pilot Plant for Coal Liquids Upgrading Process

The schematic of the process flow diagram of the mini-pilot plant for upgrading of coal liquids is presented in Figure 2. The typical operating conditions of the reactor scheme for upgrading coal liquids is shown in Table 1.

Details of Continuous Flow System and Peripheral Units for Coal Liquids Upgrading Process.

* Coal Liquids Upgrading Feed Tank - The feed to the reactor consisting of coal liquids from the liquefaction section of the mini-pilot plant will be stored in this 3 gallon feed tank equipped with an agitator. The temperature and the pressure of this vessel will be maintained at around 80-100 °C and at 50-80 psi., respectively.

The coal liquids from this feed tank is pumped through a feed pump, into the Preheater. Please refer to the earlier section regarding details about the pump and the preheater. Prior to entering the preheater, the liquid feed will be contacted by pure feed gas (H_2) from the compressed gas cylinder. The gas and slurry from the preheater flows into the coal liquids upgrading reactor.

* Coal Liquids Upgrading Reactor - This reactor is a 1-liter autoclave vessel equipped with a Robinson-Mahoney Spinning Catalyst Basket, and will be maintained at a temperature of 375 - 440 °C. The catalysts in the spinning catalyst basket that will assist in the upgrading reactions, will include conventional supported catalysts, and catalysts prepared in the laboratory, such as mesoporous alumino-silicate molecular sieve based catalysts. Provision is made for periodic analysis of the liquid inside the reactor to monitor the progress of the reaction.

The effluent from this upgrading reactor consisting of unreacted gases, product gases, and upgraded coal liquid products, flows into the vapor-liquid separator. Please refer to the earlier section regarding the detailed functions of this separator.

* Atmospheric Flash Tank - The gas and liquid flowing from the V/L separator, flows through a BOM valve and then enters the atmospheric flash tank which is a 1-liter agitated vessel. The flow rate of the gas exiting this vessel is measured and then vented to atmosphere. Periodic G.C. analysis of this product gas and the liquid collected in the vessel will be performed to monitor the progress of the reaction. The upgraded coal liquid stays in the vessel.

STATUS OF THE MINI-PILOT PLANT REACTOR SYSTEM AND FUTURE PLANS

According to the process configuration in the continuous-mode reactor scheme for coal liquefaction and upgrading of coal liquids, a cost estimation for the various individual components and accessories, that includes the reactors, slurry pump, water pump, etc., was performed. According to the cost analysis study and depending on budgetary constraints, the continuous-mode mini-pilot plant scheme will either have only manual controls, or have computer controls for selected equipments, or will be an entirely computerized system.

Several of the major process equipments (reactors, separators, flash tank, feed tanks, water reservoir, slurry pump, water pump, pneumatic compressor, compressed gas cylinders, liquid collection bombs, etc.) were ordered to be delivered from the manufacturers, and a few of them have been received. The other equipments and accessories will be ordered from the manufacturers soon. The assembly of this mini-pilot plant is expected to commence soon.

ACKNOWLEDGMENT

This project was supported by the U.S. Department of Energy, Pittsburgh Energy Technology Center (PETC), and the Air Force Wright Laboratory at the Wright Patterson Air Force Base (WPAFB), Dayton, Ohio. We wish to thank Mr. W.E. Harrison III of WPAFB and Dr. S. Rogers of PETC for their support. We also thank Dr. A. Cugini and Dr. D. Krastman of PETC for the helpful discussions on the liquefaction reactor system.

REFERENCES

1. Song, C., Parfitt, D. S., and Schobert, H. H.; *Energy & Fuels*, Vol. 8, No. 2, 313-319, 1994; Huang, L., Song, C., and Schobert, H.H.; *Am. Chem. Soc. Div. Fuel Chem. Preprints*, 37(2), 223-227, 1992.

2. Song, C. and Saini, A.K.; *Energy & Fuels*, Vol. 9, No. 1, 188-189, 1995; Song, C. and Saini, A. K.; *Am. Chem. Soc. Div. Fuel Chem. Preprints*, 39(4), 1103-1107, 1994; Song, C., Saini, A.K., and Schobert, H.H.; *Am. Chem. Soc. Div. Fuel Chem. Preprints*, 38(3), 1031-1038, 1993.

3. Provine, W. D., Porro, N. D., LaMarca, C., Klein, M. T., Foley, H. C., Bischoff, K. B., Scouten C. G., and Cronauer, D. C.; *Ind. Eng. Chem. Res.*, Vol. 31 (4), 1170-1176, 1992.

4. Lee, J. M., Vimalchand, P., Cantrell, C. E., and Davies, O. L.; *Proceedings of the Liquefaction Contractors' Review Conference*, September 22-24, 1992, Pittsburgh Energy Technology Center, U.S. Department of Energy, pp 35-96.

5. Lee, L. K., Comolli, A. G., Pradhan, V. R., and Stalzer, R. H.; *Proceedings of the Coal Liquefaction and Gas Conversion Contractors Review Conference*, September 27-29, 1993, Pittsburgh Energy Technology Center, U.S. Department of Energy, pp 547-559.

6. Pradhan, V. R., Comolli, A. G., Johanson, E. S., Lee, L. K., Stalzer, R. H., *Proceedings of the Coal Liquefaction and Gas Conversion Contractors Review Conference*, September 27-29, 1993, Pittsburgh Energy Technology Center, U.S. Department of Energy, pp 425-454.

7. Cugini, A. V., Krastman, D., Hickey, R. F., and Lett, R. G.; *Proceedings of the Liquefaction Contractors' Review Conference*, September 22-24, 1992, Pittsburgh Energy Technology Center, U.S. Department of Energy, pp 203-209.

Table 1. Typical Operating Parameters of the Continuous Flow Reactor Scheme

	Coal Liquefaction Reactor	Coal Liquids Upgrading Reactor
Reactor Feed	Subbituminous Coal / Bituminous Coal	Coal Liquids Product from Liquefaction Reactor
Solvent	Wilsonville Middle Distillates	Wilsonville Middle Distillates
Catalyst	MoS_2 Catalyst from Soluble Precursors (Dispersed State)	Mesoporous Supported Catalysts
Reactor Temperature	1st Stage 350 - 400 °C 2nd Stage 400 - 440 °C	375 - 440 °C
Reactor Pressure	2500 psi of H_2	2500 psi of H_2

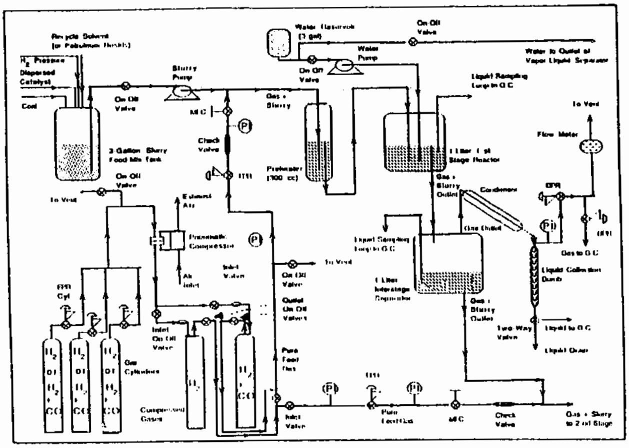

Figure 1. Process Flow Diagram of the Mini-Pilot Plant for Liquefaction of Coal

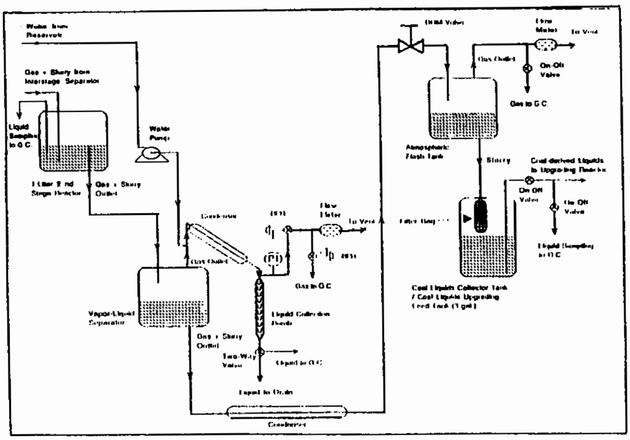

Figure 1 (contd.). Process Flow Diagram of the Mini-Pilot Plant for Liquefaction of Coal

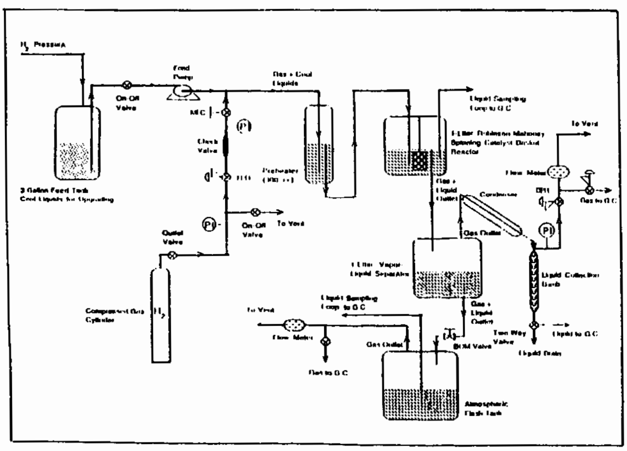

Figure 2. Process Flow Diagram of the Mini-Pilot Plant for Upgrading of Coal Liquids