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Scaleup of Mild Gasification to a Process Development Unit

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OBJECTIVES

The overall objective of this project is to develop the IGT Mild-Gasification (MILDGAS) process for near-term commercialization. The specific objectives of the program are to:

- design, construct, and operate a 24-tons/day adiabatic process development unit (PDU) to obtain process performance data suitable for further design scaleup
- obtain large batches of coal-derived co-products for industrial evaluation
- prepare a detailed design of a demonstration unit
- develop technical and economic plans for commercialization of the MILDGAS process.

BACKGROUND INFORMATION

The MILDGAS process is a continuous coal carbonization process operating in a closed system for producing liquid and solid (char) co-products at mild operating conditions up to 50 psig and 1300°F. It is capable of processing all eastern caking and western noncaking coals. It is flexible to offer options in the product slate by varying the process conditions and by blending different feed coals. The liquids can be processed as feedstocks for chemicals (e.g., BTX, phenol, cresols, xyenols, naphthalene, and indene), pitch for use as a binder for electrodes in the aluminum industry, and fuels. Depending on the feed coal characteristics and the operating conditions, the char can be used as an improved, reduced sulfur and moisture, and higher heating value fuel for power generation, or briquetted to make form

coke for steel-making blast furnaces or for foundry cupola operations. The briquetting process offers options for blending various chars and other additives (like alloying agents) to tailor the properties of the form coke. In addition, the binder for the form coke may be either pitch or a caking coal. Using coal as the binder for the form coke has the advantage of saving the pitch as a valuable co-product. The mild gasification and briquetting processes are done entirely within closed vessels which offer significant advantages over conventional coking practices for control of fugitive emissions.

Mild gasification, an alternative approach to coal conversion, may be an affordable route to increase coal utilization in the present economic climate. Because of its mild operating conditions and process simplicity, the technology could be commercialized within the next 5 to 10 years. Systems analysis studies by Bajura and Ghate¹, and Klara and Hand², as well as current DOE-sponsored projects, all support this prospect.

The MILDGAS reactor can process both caking and non-caking coals. The process splits the coal feed into coarse (+60 mesh) and fine (-60 mesh) fractions. The coarse fraction of the feed coal is fed into the fluidized bed section of the reactor. The fluidization gas distributor is designed to promote rapid mixing and dispersal of the incoming coal throughout the bed of char. The fines fraction of the coal feed is fed to the entrained section, with short residence times sufficient for conversion of the smaller particles. Under a previous program funded by DOE, IGT built and operated a 100-lb/h coal capacity isothermal process research unit (PRU) for the design of the MILDGAS reactor. The program was conducted with the participation of an industrial advisory committee consisting of steel, coal, and tar-processing companies. In the course of PRU testing and data analysis, it was determined that the relationship of coal feed rate to the mass inventory of char in the fluidized bed

is critical in controlling agglomeration with caking coals.

Two Illinois No. 6 coals, a West Virginia coal, and a subbituminous coal from Rochelle, Wyoming were tested in the PRU. The results of the successful PRU tests, conducted at temperatures in the range of 1035° to 1390°F, with Illinois No. 6 and West Virginia coals show yields of 54% to 76% char, 13% to 28% oils/tars, and 7% to 19% fuel gas.

The fate of sulfur is, of course, of paramount importance in assessing process performance and selecting co-product upgrading methods for the targeted markets. Analysis of the fate of sulfur with increasing temperature shows that the sulfur content in the char and gases decreases while the sulfur content of the oils/tars remains the same or increases slightly. In general, for the Illinois coal, the char retained 36% to 54% of the sulfur in the feed coal, the oils and tars contained 9% to 15% of the total sulfur, and the product gas accounted for 31% to 52% of the total sulfur.

The criteria for co-product selection were identified from a market survey³ and the advice of the advisory committee. Since char is the major co-product, estimated to comprise about 54% to 76% of the total co-product stream, its beneficial utilization is critical to commercialization of MILDGAS technology.

Many of the existing coking plants in the United States are reaching the limits of useful age. The supplies of metallurgical coke and high-quality coal tar pitch have been declining in the U.S. because of the closing of coke oven operations due to aging and environmental concerns. A portion of the demand for these products is being met by imports. Therefore, environmentally acceptable methods to produce supplementary supplies of coke and tar are urgently needed. The form coke production from the MILDGAS char is continuous with superior

environmental control that is difficult and costly to achieve in present coke oven batteries.

Char upgrading studies completed during the PRU program show that the MILDGAS char can be used to produce a form coke with sufficient strength and an acceptable reactivity for metallurgical use. The form coke co-product from the MILDGAS char has two sub-markets. The larger of the two markets for form coke is in blast furnaces for the production of iron. A smaller market for foundry coke is in cupolas for re-melting and alloying iron for steel castings.

It is expected from the PRU data that the MILDGAS pitch would require post-treatment to increase aromaticity and remove heteroatoms in order to be acceptable for electrode binders. This may be accomplished either by an on-stream thermal upgrading step prior to tar condensation or by post-condensation heat-treating and catalytic dehydrogenation.

About 67% of the MILDGAS oils/tars are recoverable as soft pitch. For flat and low-slope roofing, coal-tar pitches are superior to asphalt in most characteristics, including water resistance and a "self-healing" property.⁴ A possibility also exists that soft pitch could compete with petroleum-based bitumen for road binders. Blends of coal- and petroleum-based binders have been shown to have improved adhesive properties, water resistance, and skid resistance of highway surfaces.

Yields of BTX and phenols from the MILDGAS PRU were greater than those from coke ovens. These chemical feedstocks are widely used as starting materials to make plastics, synthetic fibers, and building materials.

The MILDGAS liquids, or some fraction thereof, also have the potential for being converted into transportation fuels. The middle to heavy distillates can be used as a diesel fuel

blending stock. An alternate fuel application for some of the MILDGAS liquids with some upgrading could be in gas turbines.

PROJECT DESCRIPTION

The project team currently working on the present FDU development program are Kerr-McGee Coal Corporation (KMCC), the Institute of Gas Technology (IGT), Bechtel Corporation (Bechtel), and Southern Illinois University at Carbondale (SIUC).

The PDU to be constructed is comprised of a 2.5-ft ID adiabatic gasifier to process 24 tons per day of coal to produce char, coal liquids and gases. A block flow diagram of the 24-ton/day MILDGAS Process Development Unit is illustrated in Figure 1. The facility is planned to be comprised of two major sections: one devoted to the mild gasification of the primary coal feed to produce gases, coal liquids, and char; and the other to the processing of char to a briquetted metallurgical coke. It is also planned for the facility to incorporate equipment for incineration and scrubbing to environmentally dispose of all process-generated gases.

Coal liquids from the PDU will be evaluated as feedstock for high-value chemicals and fuels by Reilly Industries, Inc. Reilly will also conduct separate modification operations such as thermal treatment with or without a Lewis Acid Catalyst, fractional distillation, and hydrotreating to produce specification-grade products.

The major portion of the char produced will be used to produce the form coke for blast furnaces and cupolas. The form coke for blast furnaces will be evaluated by several steel companies, including LTV and Armco. The form coke for use in a foundry cupola will be evaluated by General Motors Research Laboratories. In addition, the form coke will also be tested for

foundry use at Pellet Technology Corporation's 60-inch cupola. The char from the PDU will also be evaluated at SIUC for relative reactivity and for suitability as a boiler fuel in a fluidized-bed combustor.

The product testing to be conducted in the proposed program will yield a realistic assessment of the quality and economic value of both the coal liquids and solids produced. This input is required to update the market potential of the co-products and determine the slate of products and the economics of the demonstration and commercial plants for the MILDGAS process.

Mild Gasification

The mild gasification system employs the IGT MILDGAS reactor concept specifically configured for processing both caking and noncaking coals. This configuration, illustrated in Figure 2, makes provision for separate feed of coal fines to the upper entrained-bed section. The coarser feed fraction enters the lower bubbling-bed section. The lower portion of the bubbling bed section has a turbulent mixing section to handle the caking coals.

The preheated coarse coal is introduced into this turbulent mixing zone, where it is rapidly mixed with devolatilized char and hot fluidizing gas. The rapid mixing and dispersion with the fluidized bed char in this zone prevents agglomeration of the incoming caking coal. The upper portion of the fluidized bed retains the char in dense-phase. The preheated coal fines are introduced into the bottom of the upper high-velocity entrained-flow zone. These fines require little time for heat penetration and pyrolyze rapidly in suspension flow. The preheated coal fines are heated to pyrolysis temperature by the hot gases leaving the fluidized bed.

Figure 1. BLOCK FLOW DIAGRAM

Figure 2. MILDGAS REACTOR SCHEMATIC

The heat required for raising the coarse coal feed from preheat to pyrolysis temperature is provided from two sources: recycle product gas from the liquids recovery section is boosted in temperature by partial combustion with preheated air; recycled fluidized bed char is heated by partial combustion of the char, and the heated char is separated from the gases and returned hot to the fluidized bed.

Char Processing

The char from the gasifier will be used to produce briquettes that will be calcined to produce form coke. The char will also be blended with lime or limestone and briquetted to form a transportable smokeless solid fuel. The char may also be treated with steam to make activated carbon. The unprocessed char product may be tested in combustion systems to produce steam for power generation.

In the expected processing to produce a calcined form coke, dried and ground caking coal will be added to the hot char to act as a binder for the briquette. The char product from the mild gasification will be ground and blended with the coal. The blend will then be heated and masticated to form a matrix of the char in plasticized coal binder. The hot plastic mass, is then briquetted. The briquettes are next calcined to remove all volatiles and harden the binding matrix to yield form coke that meets the specification of coke for use in blast furnaces and foundry cupolas.

An alternative to the use of coal as a binder and the high temperature needed for blending the coal and char, the tar/pitch from the mild gasification may also be masticated with the ground char at lower temperatures. The green briquettes are then calcined at an elevated temperature to cure and harden the pitch/char matrix.

Liquids Upgrading

The recovered liquids will be separated during recovery into three fractions by successive reduction in condensation temperature. A provision is included to filter the high-temperature tar condensate for rejection of entrained char fines. It is planned to ship these liquid products offsite to coal tar processors for large-scale product evaluation testing. This evaluation will be supplemented by analytical characterization of representative liquid samples.

PDU Test Program

The objectives of this task are: to conduct mild gasification tests in the PDU with various types of coals; analyze the parametric test results; obtain large samples of co-products for upgrading and industrial testing and evaluation; and produce large quantities of form coke briquettes from MILDGAS char produced in the PDU.

When the PDU system is fully operational, tests will be conducted with three selected coals (a bituminous coal from Illinois, a bituminous coal from West Virginia, and a western subbituminous coal) at the ranges of operating conditions selected from the PRU tests. Specifically, the parameters to be studied in the Phase 1 testing are:

- Coal type (Illinois bituminous, West Virginia bituminous, subbituminous)
- Temperature (1000°, 1300°F)
- Recycle char:coal weight ratio (1:1, 0.75:1)
- Vapor-phase thermocracking of oils/tars

A series of parametric tests with the three coals are anticipated in Phase 1, including replicates to be selected after evaluation of initial test data. In Phase 2, sustained-operation tests

will be conducted with the three candidate coals, and at two temperatures to be selected based on the results of Phase 1 testing. The char:coal ratio and operation of the thermocracker will be based on the results of Phase 1 tests and co-product evaluation data. The tests in Phase 2 will produce larger quantities of co-products for industrial evaluation.

Co-Product Upgrading and Evaluation

One of the primary goals of this project is to investigate the necessary upgrading of the co-products to marketable forms and to evaluate the co-products in sufficiently large quantities so that the results can be reliably used for evaluating the potential economics of demonstration- or commercial-scale plants.

The char produced in the PDU will be upgraded by conversion to a calcined briquetted form coke, and to a non-calcined briquetted smokeless fuel. The form coke can be produced employing either coal or coal-derived pitch as a binder for the briquetting operation. Conversion of the char to activated carbon by steam activation will also be investigated.

Evaluation of upgraded char as form coke briquettes will be performed by Pellet Technology Corporation. PTC will evaluate the performance of the form coke in comparison to conventional foundry coke. General Motors Corporation will provide additional evaluation of the form coke for foundry use.

In addition to evaluation of form coke, mild gasification char will also be tested by SIUC for properties relating to its possible use as utility fuel, using a thermogravimetric analyzer (TGA) and a 4-inch-ID bench-scale circulating fluidized-bed.

On-site upgrading of MILDGAS liquids will consist of vapor-phase thermocracking. No other on-site upgrading of the liquids is anticipated. Rather, the segregated product fractions will be submitted to commercial tar processors for evaluation and upgrading through their customary processing operations. Selected samples of the separate or re-blended fractions will also be processed by distillation to produce gasoline and fuel oil fractions for testing in a gasoline-powered vehicle and for evaluation as a peaking gas turbine fuel.

The surplus gas produced in the mild gasification will be analyzed to verify its composition but will not be processed in any way for upgrading.

STATUS

During the first ten months of this project, the NEPA Application for construction and operation of the PDU facility at the SIUC site was written and submitted for approval. In addition, the process design for the PDU was completed, bid packages for the PDU modules were prepared and sent to a slate of prospective bidders, and bids were received from the participating bidders.

FUTURE WORK

After NEPA approval, the bids for the various sections of the PDU will be assessed and the cost for the fabrication of the PDU, site preparation, and construction of the remainder of the plant will be estimated. The total estimate for the construction of the PDU will be finalized. Once this plan is approved by DOE and the State of Illinois, contracts for the plant sections will be issued and construction started.

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