Development of a Continuous Mild Gasification Process for the Production of Co-Products

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Introduction

The primary objective of the Mild Gasification program at the University of North Dakota Energy and Environmental Research Center (EERC) has been to demonstrate a process that will produce several value-added products from a high-sulfur, midwestern bituminous or a low-sulfur, western subbituminous coal. Indiana No. 3 and Wyodak coals were used for the majority of testing. The products of the process are a low Btu gas, a hydrocarbon condensate and a low-velocity char.

An extensive survey was conducted to determine possible markets for the products. The surveys indicated that the best slate of products would consist of a metallicurgical coke substitute from the solids, a feedstock for specialty chemicals from the liquids and a fuel for the utility heat plant or a small electric cogenerating unit from the gas. The program was then tailored in an attempt to optimize the raw products to meet the industry standards for the desired end-products. In the area of form coke, we investigated organic (form coke) and inorganic binders (Peller Technology Corp) in both pelletizing and briquetting schemes. The heavy pitch fraction of the liquid products can be utilized as an organic binder in the form cok process. The lighter liquids have been analyzed to determine their potential as a source of creosote acid, as well as for use as a liquid motor fuel.

Coal is the largest indigenous energy resource in the United States. With the increasing consumption of petroleum products and electricity in the country, it is becoming increasingly important to develop processes that will delay the waste of fuel, including use as a substitute for petroleum in energy and chemical markets. One approach is to develop processes that would be analogous to coal in petrochemical refinery in which several products are produced that meet the needs of different end-users. Thus, within the process boundaries, a high-value product could be maximized at the expense of lower-value products. Under the mild gasification concept, for which research is sponsored by the Morgantown Energy Technology Center (METC) of the Department of Energy (DOE), a rapid devolatilization of coal under mild conditions of temperature and pressure would produce three products: a reactive char, a valuable hydrocarbon condensate, and a low-Btu gas. The process under development at the EERC produces metallurgical coke substitutes, diesel fuel additives, and chemical feedstocks. The coals selected for study in this project are high-sulfur, eastern bituminous (Indiana No. 3) and low-sulfur, western subbituminous (Wyodak).

Work at the EERC in the area of mild coal gasification was initiated on a bench-top scale.

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Scanning Tunnelling Microscopy (STM) as a means of Examining Fischer-Tropsch Process

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The invention of the scanning tunneling microscope (STM) revolutionized high resolution surface imaging in the last decade. According to the physical properties of the materials' surface being measured, different sensing mechanisms have been developed in the last few years. The two most well developed techniques are scanning tunneling microscopy (STM) and atomic force microscopy (AFM). STM has real space atomic scale resolution in both lateral and vertical directions, and has been used to study numerous surface problems, from silicon surface reconstruction to biological species and vertex imaging of superconductors.

The capability of STM is a driving force made of paramagnetic materials which can be expanded or contracted by an electric field. With applied voltage on the specimen device, the voltage of the probe can be precisely controlled with ingenuity resolution. The tunneling current between the probe (actually a metallic tip) and the sample is measured by the STM. This current depends on the distance between the tip and the sample, hence surface topography can be imaged (Figure 1): Atomic resolution for several materials can be easily achieved by STM in ultra high vacuum (UHV), etc., or even liquid. In order for tunneling to occur, both the sample and tip must be conducting. A major disadvantage of the STM is that it is an insulating layer (such as oxide layer) on the sample surface's top can prevent steady tunneling. This severely limits its use in an imaging tool.

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scale system using thermogravimetric analysis (TGA). This work was used to determine the basic product yields and the temperatures at which devolatilization occurred. Using data from the early TGA work, a continuous 4 lb/hr fluid-bed reactor (CFBR) system was designed. The relatively high consumption rate of the CFBR and the use of an indirectly cooled quench train aided in obtaining accurate material balances. Some processing difficulties were encountered in using the Indiana No. 3 coal. This is an agglomerating, high-sulfur, eastern bituminous coal. When the bed temperature exceeded the temperature at which the coal swells, the bed would begin to agglomerate. The relatively gentle action of the bubbling bed did little to break up these agglomerates, and, shortly, the entire bed would become a single sticky particle plugging the reactor and ending the experiment. It was possible to operate using dwelling costs only if a staged approach was taken. The coal and subsequent char were run through the reactor a number of times, with a slight increase in temperature at each step. By this method, the process could be completed and material balances were obtained by full summation of the products of each step. No operational problems were encountered using the Wyodak sub-bituminous.

The yield and operability data produced by the CFBR test program were used to design a 100 lb/hr process development unit (PDU). The PDU is a refractory-lined vessel with an 8 inch ID bed section expanding to a 16 inch freeboard section. Process heat is provided by a natural gas-fired burner. The PDU was designed for improved operability using the swelling eastern coals. The PDU is operated as a spouting bed. The 1/4 x 0 inch coal is pneumatically fed into the gasifier by a high-velocity gas jet axially located in the distributor plate. The jet rapidly carries the fresh coal into the reactor and through the bed. A fountain-like cascade is set up as the gases lose velocity in the freeboard and the particles drop back into the bed. This results in a very turbulent bed that breaks up any agglomerates as they are formed. The gas exits the reactor overhead. Any fines that may be entrained in the gas are removed by two high-efficiency cyclones. The gas quench loop on the PDU is composed of two parallel trains of direct cooling devices. The first train consists of two venturi scrubbers and a sieve tower. The second venturi uses hot liquid tar as a heat exchange fluid. Lighter fractions of oil are collected in the sieve tower. The second venturi condenses water vapor from the gas stream. The other gas quench train consists of two water-quenched venturi scrubbers. Following the quench trains, the gases pass through a pressure letdown system and are sampled and disposed of in the system.

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Gasification Process, (continued)

In a commercial mild gasification plant, the gas produced would be burned in either an electrical cogeneration unit or in a boiler to raise utility steam. The liquids have been tested for use as motor fuels, feedstocks for the specialty chemical industry and binders for a briquetted char product. Figure 1 lists data for selected liquid samples.

Char is removed from the system via a gravity bed drain. Figure 2 lists data for the chars produced during the project. The Wyodak char was the only char that met metallurgical coke specifications. The volatile content was higher than typical coke; however, this is a general specification that is tied to the briquette strength only. Typically, the higher the volatile content results are, the lower the briquette strength. For the Wyodak char, the volatile content was satisfactory. The Cannellon char data is reported without physical cleanliness. If the relative reduction of ash in the Indiana No. 3 char can be used as a comparison, the Cannellon char could be reduced to 12%-13% ash, which would be slightly higher than the specifications. The volatile content is quite low, which should produce a high-strength coke substitute. The Indiana

<table>
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<tr>
<th>Coal Specifications</th>
<th>Wyodak</th>
<th>Indiana</th>
<th>Indiana</th>
<th>Cannellon</th>
</tr>
</thead>
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<tr>
<td>90°F</td>
<td>60.0</td>
<td>74.0</td>
<td>81.7</td>
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</tr>
<tr>
<td>111°F</td>
<td>49.0</td>
<td>68.0</td>
<td>54.4</td>
<td>73.1</td>
</tr>
<tr>
<td>129°F</td>
<td>35.1</td>
<td>30.1</td>
<td>74.3</td>
<td></td>
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</tbody>
</table>

Proximate Analysis

<table>
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<th>Temperature, °F</th>
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<th>1290</th>
<th>1290</th>
<th>1290</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volatiles, %</td>
<td>&lt;10</td>
<td>15.0</td>
<td>10.3</td>
<td>10.3</td>
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<tr>
<td>Moisture, %</td>
<td>1.5</td>
<td>2.0</td>
<td>0.6</td>
<td>0.3</td>
</tr>
<tr>
<td>Ash, %</td>
<td>&lt;10</td>
<td>8.9</td>
<td>22.5</td>
<td>16.1</td>
</tr>
<tr>
<td>Sulfur, %</td>
<td>0.5</td>
<td>3.5</td>
<td>2.9</td>
<td>0.8</td>
</tr>
</tbody>
</table>

*Unchanged.

* No internal oxidation.

* Cleaned (low yield due to coal reject in cleaning stage).

A wide range of binders was tested in the char briquetting program, including the heavy pitch fraction of the liquids recovered by the quench train. The form coke produced with mild gasification char and coal-derived pitch binders demonstrated excellent properties. The briquettes had very high reactivity and great physical durability demonstrated by low solids handling tests.

Brian Range has been a researcher at the University of North Dakota’s Energy & Environmental Research Center since 1988.

CAER Educational Partnerships

The CAER held its first summer internship program for high school students June 7-16. The participants were: first row, left to right: Mi Lin, Virginia Golden; second row: Sarah Wagner, Casey Crouch.

Interns performed extensive laboratory work at the CAER.

The Center recently hosted 80 fifth graders from the Scott County Northern Elementary School. The students and their science teachers participated in hands-on demonstrations in the areas of combustion, coal cleaning, oil shale sintering and petrology.
Workshops on Adsorbent Carbons and Fullerenes

I want to take the opportunity in these columns, to mention two workshops that the CAER recently organized, and were inspired by an earlier meeting. In 1991, we first held a workshop on Adsorbent Carbons. The meeting was scheduled to attract researchers who were travelling to the biennial U.S. Carbon Conference. It was considered to be very successful, and a collection of several of the presented papers were subsequently published in the journal Carbon.

This year, from (to) 11th June at the Hyatt Regency Hotel in Lexington, we repeated the exercise, with the same timing (just before the Carbon Conference in Buffalo, NY) and with the addition of a second parallel workshop on Superconductivity in Fullerenes. In most other respects the format was the same: short, focused meetings, and limited numbers of participants to encourage discussion. The two workshops had similar audiences of around 60 each, collectively representing several countries: Canada, England, France, Italy, Japan, The Netherlands, Spain, USA, and Venezuela.

The workshop on Adsorbent Carbons was once again strongly supported by industrial companies which provided financial assistance and were well represented. Thanks are due to Air Products & Chemicals Inc; Amoco Corporation; Ashland Oil; Atlanta Gas Light; Calgon Corporation; Norit NV; and Westvaco Corporation.

The workshop on Fullerenes had a somewhat different sponsorship which included generous support from the University of Kentucky. Special thanks are extended to UK administrators Dr. Lee Magid, Vice President for Research and Graduate Studies; Dr. David Watt, Vice Chancellor for Research and Graduate Studies; and Dr. Richard Edwards, Dean, Arts and Sciences.

Once again we seem to have found a formula that works. The feedback has been very positive, and follow-up meetings are planned for 1995. While there are no proceedings of either meeting - most people already have more than sufficient writing duties - we will be pleased to supply a copy of either program and/or abstracts upon request.

Last, it is my pleasant duty to give public thanks to the speakers and chairs of the two workshops: the workshop organizers - Peter Eklund and Kambie Subhanwary of the UK Department of Physics and Astronomy for the Fullene workshop; my co-organizer Marit Jagiery of the CAER for the Adsorbent Carbon workshop; and the supporting team of Teresa Epperson, Marybeth McInturff, and Kathy Sauer of the CAER, now veterans of many such campaigns.

Figure 1. Scanning tunneling microscope showing piezoelectric tube and computer generated surface image.

There are two methods to solve this problem. One is to measure the probes deflection as it scans along the surface by an optical method instead of measuring the tunneling current. This is essentially the working principle of an AFM. As a result, even insulating surfaces can be imaged. However, since tunneling current is not measured, information on the electronic structure cannot be obtained. The second method is to install the STM in an UHV chamber and perform the experiment in-situ to prevent formation of the oxide layer. This is our approach in catalytic thin film studies.

In our STM, the piezoelectric tube holding the tip is mounted on an optical translator. The tip will approach the sample as the translator is driven by a

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Environmental accountability - impact assessment for coal

by Tim Jones, IEA Coal Research

Environmental matters continue to be an important part of the political agenda. Last year world leaders left the United Nations Conference on Environment and Development to sign a view that it was the beginning of a new process of cooperation on environmental concerns. As a result, environmental accountability is an issue to be faced by every enterprise throughout the world.

Environmental accountability may be brought about by an approach involving these distinctive phases. The environmental impact assessment (EIA) which is carried out during a project's planning stage. The second phase, the environmental management system, consists of environmental performance measures during a project's operation. Finally there is restoration and rehabilitation at the end of a project's life.

Formal procedures for EIA are relatively new to most countries. The greatest experience has been gained in the USA since the implementation of the National Environmental Policy Act (NEPA) in 1970. Requirements for EIA vary from country to country, and even within countries at each level of government. The process may be completed in a few months or may take several years where extensive data collection is deemed necessary.

Legislation overlap causes complications and for coal industries, consultation at all government levels is necessary to clarify requirements and procedures. Where an attempt has been made to introduce a uniform set of rules at the international level, as in the European Community, some nations appear to act faster than others. The EIA procedure can be improved where there is an external independent body to advise on requirements and procedures. The Commission for EIA in the Netherlands performs such a role, and in the United Kingdom, the Institute of Environmental Assessment was set up to carry out a similar function.

Most coal-related projects have a potentially significant impact on the environment, because of their general scale and nature. However, mitigating measures to reduce potential impacts to acceptable levels are possible. This applies throughout the coal chain, from extraction through transport, to utilization. Such measures may include site layout, transport corridors or containment methods. Visual impacts may be reduced by suitable screening, or color schemes. The problems of atmospheric emission may be overcome by using technologies like flue gas desulphurization (FGD) and low-NOx burners.

Considering technologically feasible alternatives during the formal EIA would be prudent. It increases credibility by demonstrating a more complete assessment, and delays in permit decision making may also be reduced. EIA often yields a better project which is not necessarily less efficient or more costly. More cost data is needed to obtain an adequate assessment of various procedures. The cost of EIA for coal-related projects is likely to be at least $300,000, and can run to millions of dollars.

EIA experience improves efficiency of subsequent similar assessments. Hence, a database to promote information exchange on coal-related projects would benefit coal producers and users, and improve cost-effectiveness.

These are the findings of a recent IEA Coal Research report, Environmental Impact Assessment for Coal. The study presents an international perspective by examining the situation in selected coal producer and/or user countries. The report summarises environmental legislation in those countries, the requirements for EIA, the procedures involved, and the potential effects of EIA. Five case studies (continued, page 6)

Scanning Tunnelling, (continued)
Commentary, continued

of recent coal-related projects serve to evaluate the experience gained by IFA for coal.

Tim Jones is a senior environmental policy maker at IEA Coal Research in London, England. His interests include environmental impact assessment and environmental auditing.

Scanning Tunnelling, continued

provide groundwork for catalytic studies in coal FT process conditions.

The Center of Applied Energy Research (CAER) initiated this research by providing the necessary equipment (such as the UHV chamber) and the support of a graduate student to begin the experiments. During this time, we synthesized and carefully characterized the catalytic Fe-Mn thin film of different compositions, and also constructed an FTIR that is especially designed for the experiments.

Kwok-Yat Ng, graduated from the University of Hong Kong with a BSc in Physics. He came to the U.S. in 1981 to attend graduate school in Texas State University, and graduated in 1986 with a PhD degree majoring in Solid State Physics. He joined the Dept. of Physics and Astronomy at the UK as an assistant professor in 1988.

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