Beneficiation of Ultrafine and Micronized Coals through Oil Agglomeration

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Introduction

When coal is comminuted, the resulting particles can range from coarse lumps of several centimeters to micron-sized respirable dust. The amount of coal in any one size-class depends on several factors including the coal’s geologic rank, presence of cleft or fractures, petrographic composition, and mining, handling, and processing methods employed. It is not unusual for coal in the cleaning plant to contain 5% by weight, particles less than 1 mm in size. Collectively referred to as ‘coal fines,’ this amount includes the ‘fine’ coal fraction (from 1 mm to 75 microns), the ‘ultraclean’ coal fraction (from 75 microns to 10 microns), and the ‘micronized’ coal fraction (particles less than 10 microns). Conventional froth flotation technology can process these sizes, but it is often sparsely practiced in the industry because of problems with coal and efficiency. More often, this coal is simply ‘passed through’ the cleaning plant and disposed of in a pond or landfill.

The Center for Applied Energy Research (CAER) is actively researching and developing novel processes to effectively handle, clean, and use coal fines. This study focuses on a process known as oil agglomeration which can produce dramatic changes in a coal’s maceral and mineral content as well as improve its handling characteristics. The formation of coal agglomerates is initiated when slurried particles become preferentially wetted by an immiscible bridging liquid such as fuel oil or pentane. Coal macerals see, to various degrees, oleophilic (oil attracting) and tend to form large, agglomerated masses while oleophobic (oil repelling) coal minerals tend to remain suspended in the slurry. Thus, the agglomerated clean coal possesses lower ash and sulfur values than the parent coal. Significant partitioning of the coal’s maceral or microlithotype content may also occur.

Five commercially cleaned coals from Eastern Kentucky were chosen for laboratory agglomeration studies. The samples were selected to provide an adequate range of composition (vitamorph ranging from 60% to 85%) and rank (Renus ranging from 0.78 to 1.09). Each was prepared in a rod mill at a pulp density of 35% W/W until 80% was less than 45 microns (725 mesh). slurries were then prepared at 8% W/W and subjected to standard laboratory blender experiments using pentane as the bridging liquid. Experiments were performed under various conditions of slurry pH and pentane concentration. Nine runs were made for each sample and each run generated a clean coal and tailings fraction for a total of 18 samples per coal.

Samples in the data base were characterized for ash, sulfur, maceral, and microlithotype content (Kuehn, 1992). CLEAN COAL RECOVERY After blending, agglomerated clean coal floats as a layer upon the denser mineral–rich suspension. This mixture was carefully poured onto a 100 mesh (150 micron) sieve thereby defining the yield of clean coal as all the >100 mesh material and the tailings as all <100 mesh material. In these runs, under varying conditions, the average clean coal recovery was 87% ± 4.5%

Figure 1 illustrates a typical clean coal re-

Estimation of Physical Properties of Hydrocarbons in a Coal-Derived Liquid by a Group Contribution Method

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INTRODUCTION

A simple method of calculating the physical properties (boiling point, density and refractive index) of hydrocarbons in a coal derived liquid was developed using the group contribution method. The component groups adopted in this study are aromatic rings, naphthenic rings, aromatic compound carbons, and aromatic center carbons. The contributions of component groups to physical properties were calculated by summing the calculated contributions weighted by the mole fractions of the component groups in the liquid. These values are applicable in predicting liquid hydrocarbon mixtures’ physical properties such as coal derived liquids, as well as pure hydrocarbons. The correlation of the bond energies and their physical and thermal properties was facilitated by least-squares fitting of the experimental data.
Beneficiation, continued

covvy curve for the pentane experiments. The graph shows that initially, small amounts of pentane yield small percentages of clean coal. At 2.5ml per test, the yield is less than 5%, but quickly reaches 80% at 10ml pentane. At this point the curve flattens and very little additional clean coal is recovered even upon doubling or tripling the amount of pentane. Presumably, a critical amount of pentane is needed to sufficiently wet the particles and once wetting is achieved, agglomeration proceeds and is not aborted by further pentane additions.

SULFUR AND ASH

It is significant that the coals used were already cleaned in a commercial process. Thus, any further cleaning through comminution and agglomeration represents an actual improvement to existing product quality. The five samples initially ranged from 3.47% to 9.40% in ash content (dry basis) and from 0.53% to 2.45% in total sulfur content (dry basis). These figures are very acceptable for most purposes. The experimental data, however, show further improvements to quality by reducing ash by an average of 27.8% to 4.1% and sulfur by an average of 7.0% to 1.7%.

MACERALS AND MICROLITHOTYPES

Figure 2 illustrates two photomicrographs the typical appearance of the clean coal and talc fraction derived from the same agglomeration run. The clean coal fraction, which is quite low in mineral matter, also has a simpler maceral composition which tends to be enriched in vitrinites. The associated talc fraction tends to show a concomitant increase in inertinites and lampinites as well as extremely high, high and medium contents. Overall, 50% of the 45 samples analyzed microscopically showed a significant amount of this maceral pairings between clean coal and talc. Results obtained for one coal is given in Table 1.

Characterization of the coal's microlithotype composition shows how macerals are associated and how individual particles reacted in the agglomeration experiments. Three of the five coals showed significant partitioning of microlithotypes as well. Clean-coal fractions tended to be enriched in bright microlithotypes while the associated talc fractions showed increase in dull types such as inertite and durvitrinite.

DISCUSSION

The high percentages of recovered clean coal viewed with their lowered ash and sulfur contents suggest a potential benefit from upscaling the agglomeration process. In fact, agglomeration using fuel oil as the bridging liquid has been demonstrated both on the pilot scale and in a commercial process. Fuel oil, however, has the disadvantage of being non-recyclable because it adheres tightly to coal particles. It also emits a strong odor and is corrosive to the conveyors and belts in a cleaning plant. Interest thus has turned to pentane whose high vapor pressure under ambient conditions may permit partial recovery and recycling of the liquid on a large scale. The ability to process fine coals commercially and add them to the product stream would mean additional revenues for the industry and less discarded coal.

The coal fines already discarded in ponds represent a significant potential resource if they could be efficiently recovered, cleaned, and used. The recent study (Kushn and Beene, 1990), it was determined that one year of one Western Powder coal pond contains approximately 7 million tons of discarded fines. The oil agglomeration process can handle the handling problems by creating a larger effective particle size more amenable to on-site recovery operations. Our results concerning the partitioning of macerals and microlithotypes indicate that for some purposes, it may be worthwhile to intensively sort the entire coal feed to ultralime and minorized sizes. As coal is comminuted, breakage occurs along preexisting fractures in the organic matrix, and it is well known that deeper cleaning is possible as particle size decreases owing to mineral and maceral liberation. Pyrite, for example, the predominant form of sulfur in most U.S. coals, commonly occurs in euhedral, blebs, or framboids less than 25 micrometers. Clay minerals, the main contributor to coal ash, crystallize at 4 microns or less. Certain coal macerals, such as the inertinites, which are friable, tend to concentrate in the coarsest range while sporeinite tends to be concentrated in coarser sizes ranges. Thus, crushing coal can have profound effects on the organic matrix, inorganic composition of smaller sizes ranging from medium to fine coal. Ultimately, it may be possible to tightly control the quality characteristics of ultrimers and minoritized coals in a commercial process. The prospect of processing reliable clean coal feedstocks for specialized uses as well as liquidation or alternative diesel fuels is intriguing.

In the past, chemical processes were hampered by extremely slow diffusion rates into fine grained coal of only a few microns per hour. Thus, any significant reduction in particle size should be advantageous. For example, solvent penetration to a depth of 2 microns into a spherical particle whose diameter is 75 microns affects only 15% of the particle volume. The same penetration depth into a spherical particle of 5 micrometers diameter would affect 99% of its volume. This advantage could help advance the development of practical chemical beneficiation and liquefaction processes. These possibilities have been pointed to in a detailed study on the chemistry and reactivity of minoritized coals conducted by the Western Kentucky University Center for Coal Science. This research has also established that commercially available attritor mills can produce large quantities of submicron, micronized coal in a cost effective manner (Lloyd and others, 1988).

<table>
<thead>
<tr>
<th>TABLE 1. Examples of Maceral Partitioning</th>
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<tbody>
<tr>
<td>Coal KCER-91846 (25m) (pontane) Vitrinates Inertites Liptinites</td>
<td>%</td>
</tr>
<tr>
<td>pH1 clean</td>
<td>70.3</td>
</tr>
<tr>
<td>talcites</td>
<td>60.0</td>
</tr>
<tr>
<td>pH1 clean</td>
<td>70.9</td>
</tr>
<tr>
<td>talcites</td>
<td>59.3</td>
</tr>
<tr>
<td>pH1 clean</td>
<td>72.8</td>
</tr>
<tr>
<td>talcites</td>
<td>66.0</td>
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Physical Properties (continued)

Extensive knowledge about coal derived liquids' physical properties is necessary for the optimum design and operation of coal liquefaction and upgrading processes. Significant amounts of aromatic hydrocarbons are contained in coal derived liquids. The data on physical properties are generally available for low molecular weight hydrocarbons, but to date, data on heavier compounds is still not easily available data. However, a correlation is needed to estimate the values of physical properties of the limited available data.

Physical properties of heavy hydrocarbon molecules are closely related to their chemical structures. A molecule, it is possible to assign partial values for the properties to each component group in the molecule on the premise that as long as the additive rules are followed for the property the additive property is the sum of the group contributions. This is useful in estimating the physical properties of coal-derived liquids. Generally, prediction methods involving group contribution methods are obtained by regression analysis, and the greater accuracy, the more the number of atomic component groups increase. In practice, these methods might be available for the physical property estimation, however, it is difficult to find an academic interest in them. For example, we cannot quickly answer a plain question like how a certain physical property changes by increasing aromatic rings or naphthenic rings. In this paper, estimation methods, which could answer the above mentioned question of the most fundamental properties, were proposed using a group contribution method.

In applying a group contribution method to a coal derived liquid, information on its chemical structure is necessary. Because of its complexity it is impossible to identify single components of the coal derived liquid. The authors have proposed a new analytical method to reveal the chemical structure in the liquid.

By our methods, aromatic hydrocarbon structure can be expected with the number of aromatic rings, naphthenic rings and carbons of alkyl side chains. Furthermore, two additional component groups are proposed to represent the condensed polynuclear aromatics. For instance, both pyrene and chrysene have four aromatic rings. These compounds are distinguished by using aromatic conjunction carbons and aromatic inner carbons. For example, the number of those component groups in trimethylphenylidanaophenopyrene, K_{37}H_{46} are defined as shown in figure 1.

The authors' first question was how the physical properties change with the total carbon number on the homologue of polynuclear aromatics with no naphthenic ring such as a series of alkylbenzenes, alkyl-napthenoalkanes, and alkylnaphthalenes. The changes of boiling point, molar volume and M/I (molar index) for the various components and alkyl side chain are shown in figures 2, 3, and 4, respectively.

Two conclusions may be drawn from these figures. One is that for the properties of the homologue with the increase of total carbon number is similar to that of normal paraffins, the boiling points group roughly about 30 K per aromatic ring, whereas the molar volume and values are not dramatically arrived at 40 molar and 10 per aromatic ring, respectively.

(continues on page 4)
Naphthenic rings are deserve serious consideration. Changes in properties with the total carbon number are also similar to that of normal paraffins. The total carbon number for the same, the boiling points rise about 10 K per naphthenic ring, where as the smaller volumes and values of M/n fall about 2 ml/mol and 5 per naphthenic ring, respectively.

In the boiling point prediction, contributions of aromatic condensation carbons and aromatic inner carbons are considered. The contribution of aromatic condensation carbons appears to be negligible, where the boiling point declines about 5 K per aromatic inner carbon, if the total carbon number is equal.

The variation with total carbon number in physical properties of homologues of non-paraffin compounds is close to that of paraffin homologues. The group contributions to a property of a given hydrocarbon are almost constant, regardless of total carbon number changes. Consequently, the boiling points of hydrocarbon...
ONE-STOP SHOPPING
By Jack Siegel

Last year a delegation from Thailand stepped off an airplane at Dallas International Airport outside of Washington, D.C., to begin a shopping tour of the United States. But this was no ordinary shopping trip.

Instead of browsing America’s stores for souvenirs, Thai officials were in the United States to shop for clean coal technologies—methods of advanced pollution control used in power generating systems.

The prospects for coal growth in Thailand are only a microcosm of the world as a whole, particularly the developing nations. It intends to more than triple its coal use—65 million tons by the year 2000—to keep up with the rising demand for power.

Coal consumption worldwide could expand by almost 1 billion tons by the year 2010 as other developing countries seek to modernize their industrial base. But, without the concurrent introduction of cleaner, more efficient coal-burning technologies, the inevitable growth in global coal consumption could spell trouble for the world’s environment.

In Thailand last year, sulfur dioxide emissions from the Mae Moh lignite power plant, which accounts for 27 percent of Thailand’s entire power generation, reached such health-threatening levels that the press called the incident a “mini Chernobyl.” So the Thai delegation’s “shopping” trip took on an added degree of urgency.

The United States is positioning itself as the ideal place for international buyers to come and “kick the tires” of environmentally cleaner coal-burning technology. Largely because of our multi-billion dollar investment in the Clean Coal Technology Program (CCT), we now offer the most extensive array of new pollution control technologies of any country in the world.

The $5 billion-plus investment by government and industry in clean coal technology has provided this country with a tremendous marketing advantage—and the international market could be huge. We estimate that between now and 2010, advanced clean coal technology purchases worldwide could amount to about $24 billion per year. If the United States can capture only a quarter of this expanding market, as many as 80,000 jobs could be added to the nation’s work force.

Worldwide adoption of clean coal technologies can mean more to the earth’s environment than just dramatic reductions in smog and air pollution levels. Their deployment on a global scale also offers one of our best means for reducing the buildup of carbon dioxide (CO2). The reason is efficiency. Today, the worldwide average thermal efficiency of a coal-burning power plant is only about 25 percent, compared to the 33 to 35 percent efficiencies of power plants in the United States. For many developing nations, efficiencies will drop to 15 percent or less. A less efficient plant burns more coal to generate electricity and thus gives off more CO2.

We project that clean coal technologies with efficiencies of more than 40 percent penetrate just a quarter of the world’s coal use by 2025, global CO2 emissions from coal burning will actually begin to decline slightly from projected levels in 2000, despite growth in coal use. Perhaps more important, CO2 emissions would be more than 26 percent less than if no improvements to power plant technology are made. Center market penetration by the year 2030, coupled with even better technology could virtually level off CO2 emissions, even as worldwide coal use escalates sharply.

Trade missions to sell U.S.-demonstrated clean coal technologies, therefore, not only make good economic sense, but they clearly make good environmental sense. The reciprocal visits such as the trip by the Thai officials, allow prospective customers to talk with the engineers who design the clean coal systems, with the companies that construct them, and with the operators who have first-hand experience in using them.

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Commentary, (continued)

running, debugging and perfecting them. The “hands on” expertise of these people makes them the world’s best clean coal technology salespeople.

The U.S. government can help open the doors to America’s expanding clean coal showrooms. The Department of Energy is working closely with other government agencies to put U.S. sponsors of clean coal technologies together with potential buyers.

Working as a team, we have enormous opportunities to leverage our domestic investment in cleaner technology and a better environment. We have the opportunity to arrange many other such “shopping trips” for international buyers. I believe in terms of jobs, a better environment, and ultimately perhaps, an unattainable, prosperous world may be incalculable.

Jack Sigrf is the Department of Energy’s deputy assistant secretary for coal technology.

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1994 CAER Annual Advisory Board Meeting

Some of the CAER Advisory Board members at the recent annual meeting. From left to right: David Drake, East Kentucky Power; Frank Burke, CONSECO; Robert Alvington, Ashland Resources; and John Neuman, Ashland Carbon Fibers

Don Cromar, Ayco Research Center and Doug Horne, Atlanta Gas Light

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