



Energy, Mines and  
Resources Canada

Énergie, Mines et  
Ressources Canada

## CANMET

Canada Centre  
for Mineral  
and Energy  
Technology

Centre canadien  
de la technologie  
des minéraux  
et de l'énergie

### BENEFICIATION OF LIGNITE BY OIL AGGLOMERATION AS AN INTEGRAL PART OF COPROCESSING

MICHIO IKURA, JAMES F. KELLY AND C. EDWARD CAPES

JANUARY 1988

For presentation at the Symposium of Coprocessing organized by the ACS Division of Fuel Chemistry at the Third Chemical Congress of North America, June 5-11, 1988, Toronto, Ontario and for publication in the ACS preprints.

CANMET LIBRARY



3 2329 00056602 1

ENERGY RESEARCH LABORATORIES  
DIVISION REPORT ERL 88-04(OPJ)

ERL 88-04 (OPJ) L.2

## BENEFICIATION OF LIGNITE BY OIL AGGLOMERATION AS AN INTEGRAL PART OF COPROCESSING

Michio Ikura<sup>1)</sup> James F. Kelly<sup>1)</sup> and C. Edward Capes<sup>2)</sup>

1) Energy Research Laboratories, CANMET, Energy, Mines and Resources Canada. 555 Booth St., Ottawa, Ontario K1A 0G1

2) Chemical Engineering Section, Division of Chemistry, National Research Council, Montreal Rd., Ottawa, Ontario K1A 0R9

### Abstract

The low cost of Canadian low-rank coals makes them prime feedstocks for coprocessing with bitumens and heavy oils. Depending on the coal concentration in the feed slurry, the high ash level in some of these coals can cause operational problems as well as reduce the overall process efficiency and the net liquid product yield. To reduce the ash content in the feed slurry, oil agglomeration can be used for coal beneficiation (deashing) as an integral part of coprocessing technology. However, an optimum integrated approach requires the use of undiluted bitumen or heavy oil feedstocks as a bridging liquid. Also, low-rank coals as well as oxidized higher rank coals often possess a relatively hydrophilic surface. This makes the application of oil agglomeration extremely difficult.

A novel method of beneficiating low-rank coals for coprocessing by oil agglomeration using a part of the feed bitumen for coprocessing has been conceived and developed. This method overcomes the hydrophilic surface problem of low-rank coals and allows the use of undiluted coprocessing feed oils as viscous as vacuum bottoms.

Experimental results on the beneficiation of a Canadian lignite with vacuum bottoms are presented. The efficiency of the process was evaluated in terms of combustibles recovery and ash rejection. The agglomeration of Willowbunch lignite (ash content from 20 to 22 wt % on a mf basis) with Interprovincial Pipe Line (IPPL conventional crude) vacuum bottoms resulted in 51 to 97 wt % combustibles recovery and a corresponding 77 to 42 wt % ash rejection. The effects of vacuum bottoms concentration, operating temperature, and processing time on the beneficiation performance are discussed.

### Introduction

Low-rank coals that are often recovered by surface mining are inexpensive and good feedstocks for coal-oil coprocessing. Unfortunately, Canadian low-rank coals are often characterized by their high ash content. For instance, the ash content of Canadian lignites is between 15 and 30%. Most of the low-rank coal deposits in the USA, Australia and other countries also have high ash contents. When such a coal is subjected to coprocessing with heavy oil or bitumen, the high ash content can reduce the net liquid product yields and the overall efficiency of the plant. Also, the handling of process residues with high solids contents may cause operational problems.

An effective route to overcome the high ash problem in the feed coal is coal beneficiation, i.e., deashing of the coal to a certain degree prior to coprocessing. The choice of an appropriate beneficiation process depends on coal rank, composition, surface properties, dissemination of mineral matter, and the end use of the cleaned coal.

Coal beneficiation based on gravity separation of carbonaceous and mineral particles is the most widely used commercial method. However, the efficiency of this technique depends on coal washability, i.e., percentage of mineral matter which can be separated by gravity from various size fractions of coal. For low-rank coals in which mineral particles are often found in a very fine state, beneficiation based on gravity separation may be ineffective.

Selective oil agglomeration, alone or in combination with other methods, may be appropriate for the beneficiation of low rank coals. This technique involves the addition of oil to an aqueous suspension of coal and ash particles. The oil preferentially wets the carbonaceous constituents and causes adhesion of the coal particles by capillary interfacial forces (1). The carbonaceous constituents are agglomerated by oil and recovered from the suspension whereas the ash remains in suspension and is rejected. The oil agglomeration method is based on differences in the surface properties between organic and inorganic matter, the first usually being hydrophobic and the latter hydrophilic. For successful oil agglomeration of coal, the carbonaceous constituents must be less hydrophilic than the ash. As coals decrease in rank from bituminous to subbituminous and to lignite their surface properties become more hydrophilic. This is the major reason that oil agglomeration becomes more difficult for low rank coals (1-3).

The application of selective oil agglomeration to fine coal beneficiation may be especially advantageous if it can be used as an integral part of coprocessing. In coprocessing, coal is ground and mixed with heavy oil to prepare the feed slurry. Normally the feed slurry contains oil and coal at approximately a 2 to 1 ratio. Thus a portion of the heavy oil or bitumen for coprocessing could be utilized for the selective oil agglomeration beneficiation of the feed coal prior to the preparation of the feed slurry (Figure 1). Clean coal particles in the form of agglomerates would then be sent to coal-oil slurry preparation. This integrated process configuration does not result in any additional cost for coal grinding and bridging oil. Also, the overall efficiency and economics of coprocessing will improve by reducing the need for dilution of high ash content residues to permit easier downstream handling.

This paper presents experimental results on a method for lignite beneficiation using vacuum bottoms only as an agglomerating oil. It was shown that at elevated temperatures the vacuum bottoms performed well as a bridging liquid and a hard-to-agglomerate lignite was successfully agglomerated and beneficiated.

## EXPERIMENTAL

All beneficiation experiments were carried out using Willowbunch lignite ground to less than 208  $\mu\text{m}$  (less than 65 Tyler mesh size). A nominal 500 mL of an aqueous solution containing coal surface conditioner and surfactant was prepared to make approximately a 10% coal suspension. The coal suspension was heated in a closed high pressure mixer to a predetermined temperature and a measured quantity of melted vacuum bottoms was injected into the aqueous solution. High shear mixing (4800-5000 rpm) for 5-20 min was used to create microagglomerates followed by low shear mixing (2000-2300 rpm) for 4-10 min to allow for agglomerates growth. Upon completion of an experiment, the vessel contents were discharged into a vessel filled with water at atmospheric pressure. The agglomerates were then separated over a 100 mesh sieve while the tailings were collected in a dish under the sieve. Residues in the vessel were collected by rinsing with water, then Varsol.

Moisture and ash analyses of lignite samples were carried out prior to oil agglomeration tests in accordance with ASTM procedures. Moisture, ash and oil analyses of agglomerates and tailings were performed at the completion of each run. For the determination of the moisture content of agglomerates, a 2-3 g sample of air-dried agglomerates was weighed, then dried in an oven at  $107 \pm 3$   $^{\circ}\text{C}$  to a constant weight. For the determination of the ash content of agglomerates, a 2-3 g sample of air-dried agglomerates was weighed, then heated in a closed crucible in a furnace to  $750^{\circ}\text{C}$  and kept at this temperature until no further weight change was recorded.

To determine the ash content of agglomerated lignite on a moisture-oil-free basis, the oil had to be removed from the agglomerates prior to ashing. Since bitumens used for agglomeration of the lignite have high boiling temperatures, they were removed by solvent extraction using toluene. A 3-4 g sample of dried agglomerates was placed in a predried Soxhlet extractor thimble. Upon completion of the extraction, the thimble was dried in a vacuum oven at  $90^{\circ}\text{C}$  to remove any toluene residues. The moisture-oil free lignite sample was then weighed and ashed.

Ash rejection and combustibles recovery were calculated as follows:

$$\text{Ash rejection (wt \%)} = B/(A+B+C) \times 100 \quad 1)$$

where A, B, and C denote ash in agglomerates, in tailings, and in residues in the agglomeration vessel respectively (all on a moisture-oil-free basis).

$$\text{Combustibles recovery (wt \%)} = (E+F)/(D+E+F) \times 100 \quad 2)$$

where D, E, and F denote combustibles in tailings, in agglomerates, and in residues in the agglomeration vessel respectively (all on a moisture-ash-free basis).

Tables 1 and 2 give typical ultimate and proximate analyses of Willowbunch (Saskatchewan) lignite and the properties of IPPL vacuum bottoms respectively.

## RESULTS AND DISCUSSION

Figure 2 summarizes the effect of vacuum bottoms concentration on the ash rejection and combustibles recovery. The vacuum bottoms concentration was varied from 20 to 64% based on moisture free lignite. When small amounts of vacuum bottoms are used, little agglomeration occurs and the largest part of the ash remains in the tailings. The results in Figure 2 were obtained at 150°C in high and low shear mixing stages. The duration of the high shear mixing was kept at 20 min and that for the low shear mixing was varied between 4 and 10 min.

As the vacuum bottoms concentration is increased from 20 to 40%, combustibles recovery increases from 50 to 93%. However, further increase in vacuum bottoms concentration shows no significant enhancement in combustibles recovery. Ash rejection decreases monotonously with increasing vacuum bottoms concentration.

These results can be explained as follows: When the amount of vacuum bottoms is insufficient for the effective agglomeration of lignite particles in the suspension, the agglomeration is simply incomplete and a large fraction of the lignite particles passes through the 100 mesh sieve with the tailings suspension. As the vacuum bottoms concentration is raised, more vacuum bottoms droplets become available for collision and eventual agglomeration of lignite particles; thus the combustibles recovery further increases. However, at higher combustibles recovery, ash rejection is reduced since there is ash associated with incremental lignite recovery. In addition, an excessive amount of vacuum bottoms in the aqueous suspension results in the formation of emulsion-like agglomerates which contained most of the lignite particles bridged together, as well as a substantial amount of entrapped ash particles. As a result, combustibles recovery is marginally higher but there is an attendant decline in ash rejection.

Figure 3 shows the effect of operating temperature at 40% vacuum bottoms concentration. At 130°C and 140°C, the combustibles recovery was as high as 97.6 to 98.1% at an ash rejection of between 52.5 and 53.1%. The tailings and washings had a clear appearance with very light colored ash settling at the bottom of the tailings collection pan. When the temperature was increased to between 150-160°C, the combustibles recovery slightly decreased to between 94.0 and 95.7% whereas the ash rejection remained essentially constant. This may be explained by the increased viscosity of vacuum bottoms droplets in the suspension. The vacuum bottoms are more viscous at the lower temperature, and when the vacuum bottoms droplets collide, they tend to aggregate resulting in higher combustibles recovery. Because the concentration of suspension is in the order of 10 to 15%, the overall viscosity of the suspension will not change appreciably by the increased vacuum bottoms viscosity. Therefore, it is more advantageous to operate with a vessel temperature as low as practical to promote high combustibles recovery without sacrificing ash rejection. It should be noted, however, that when the temperature of the vacuum bottoms is too low, it becomes impossible to create the fine oil droplets that are needed for oil agglomeration.

Figure 4 shows the effect of processing time used for the high shear mixing. This series of runs was conducted at 150°C and 40% vacuum bottoms concentration. It is seen that high shear mixing for 5-7 min achieved high combustibles recovery and sufficient ash rejection. A further increase of high shear mixing time resulted in a slight decrease of combustibles recovery. This may reflect the fact that some of the microagglomerates were destroyed during prolonged high shear mixing. It was observed that after 3 to 5 min of high shear mixing, the vessel contents had gradually changed from black to gray and then to white. After 7 to 9 min of high shear mixing, a darkening of the vessel contents was observed. Also the appearance of the tailings obtained after 7 min of high shear mixing was clear with mostly white ash settled to the bottom of the tailings collection pan whereas a darkening of the settled particles was observed for the prolonged runs at 9 to 20 min. At 300 min of high shear mixing the combustibles recovery was down to 49.4% and ash rejection increased to 66% indicating unsatisfactory agglomeration. It can therefore be concluded that the present process requires only a very short processing time, and an excessively long mixing time has detrimental effects on the overall agglomeration performance.

#### CONCLUSIONS

An oil agglomeration technique using vacuum bottoms only as a bridging liquid was developed for low-rank coal beneficiation. It was shown that at elevated temperatures the vacuum bottoms performed well as a bridging liquid and a hard-to-agglomerate lignite was successfully agglomerated and beneficiated. In this work a processing time of 5 to 10 min was sufficient to achieve greater than 90% combustibles recovery and more than 55% ash rejection. The present process is ideally suited for ash reduction in coprocessing feed because it can use a part of the coprocessing feed oil for agglomeration before preparation of the slurry feed to the reactor.

#### REFERENCES

- 1) Capes, C.E. and Germain, R.J.; "Selective Oil Agglomeration in Fine Coal Beneficiation", in the "Physical Cleaning of Coal", Editor Liu, Y.A., Marcel Dekker Inc., pp 293-351, 1982.
- 2) Pawlak, W., Turak, A., Janiak, J., Ignasiak, B., Szymocha, K., Ignasiak, T. and Rodkiewicz, C.; "Agglomeration of Low Rank Alberta Coals with Bridging Liquids Derived from Indigenous Bitumen and Heavy Oil Deposits", Proceedings of the International Conference on Coal Science, pp 525-528, Sydney, Australia, Oct.28-30, 1985.
- 3) Capes, C.E., Coleman, R.D. and Thayer, W.L.; "Oil Agglomeration for Fine Coal Processing", Proceedings of 4th International Symposium on Agglomeration, Iron and Steel Society, pp 857-866, Toronto, Canada, June 2-5, 1985.

Table 1 Ultimate and Proximate Analyses of Willowbunch lignite

<u>Ultimate analysis (wt %)</u>	
Carbon	52.1
Hydrogen	4.0
Sulphur	0.9
Nitrogen	0.5
Ash	21.5
Oxygen (by difference)	21.0
<u>Proximate analysis (wt % mf)</u>	
Volatile	39.6
Fixed carbon	38.5
Ash	21.0
Sulphur	0.9

Table 2 Properties of IPPL vacuum bottoms

<u>General</u>	
Minus 525°C fraction (wt %)	0
Plus 525°C fraction (wt %)	100
Aromaticity	30
Toluene insolubles (wt %)	0.92
Viscosity (cSt) at 100°C	906
	110
	130
184	
<u>Elemental analysis (wt %)</u>	
Carbon	86.4
Hydrogen	10.4
Nitrogen	0.43
Sulphur	1.71
Oxygen	0.56

Figure 1

Flowsheet: Oil Agglomeration Beneficiation Integrated with Coprocessing

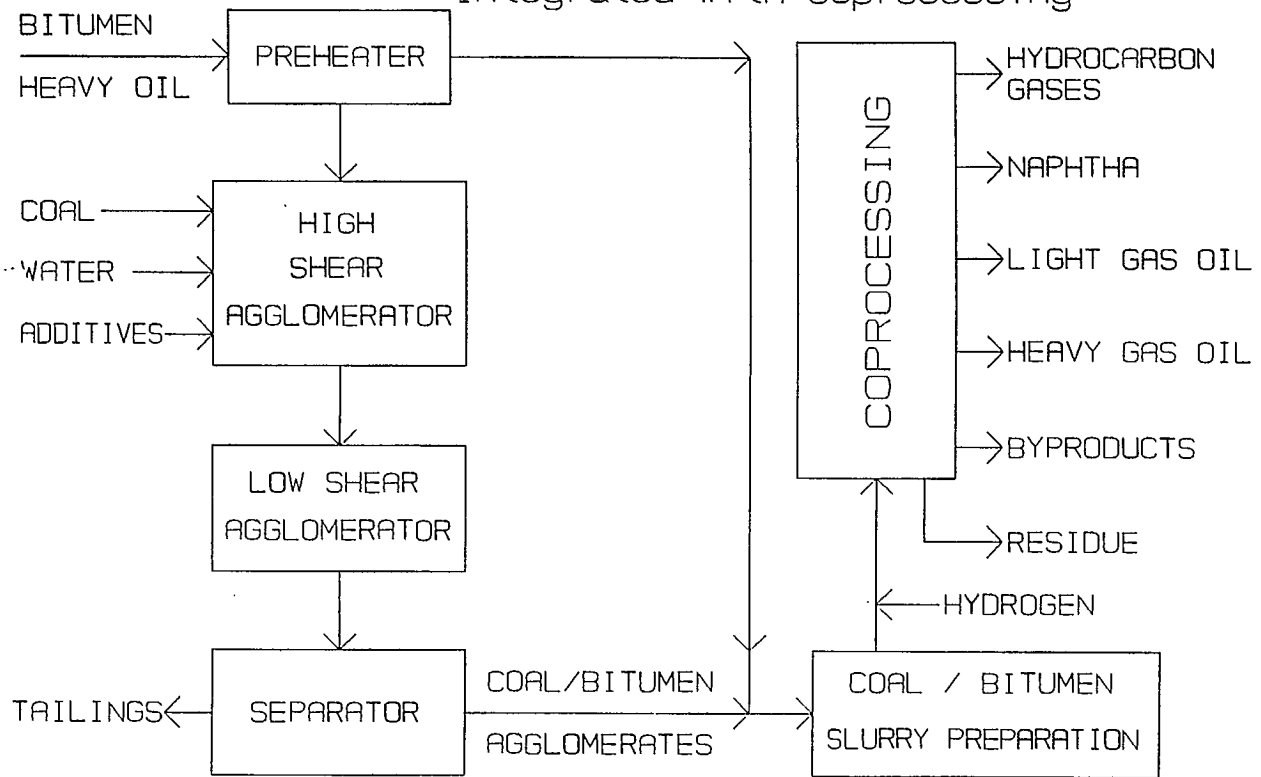


Figure 2  
Effect of Vacuum Bottoms Usage

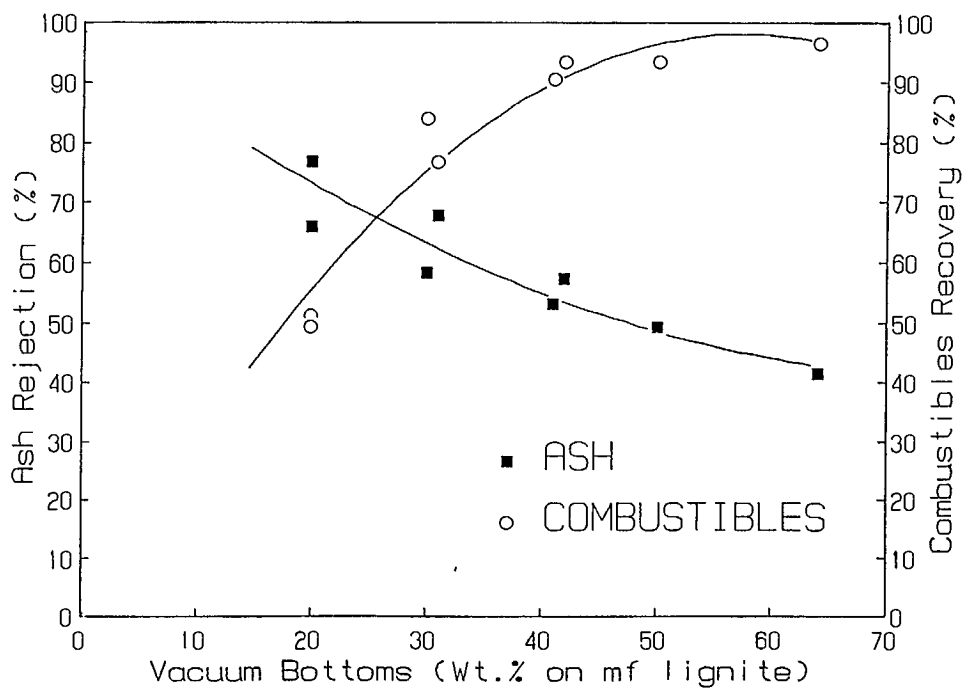




Figure 3  
Effect of Operating Temperature

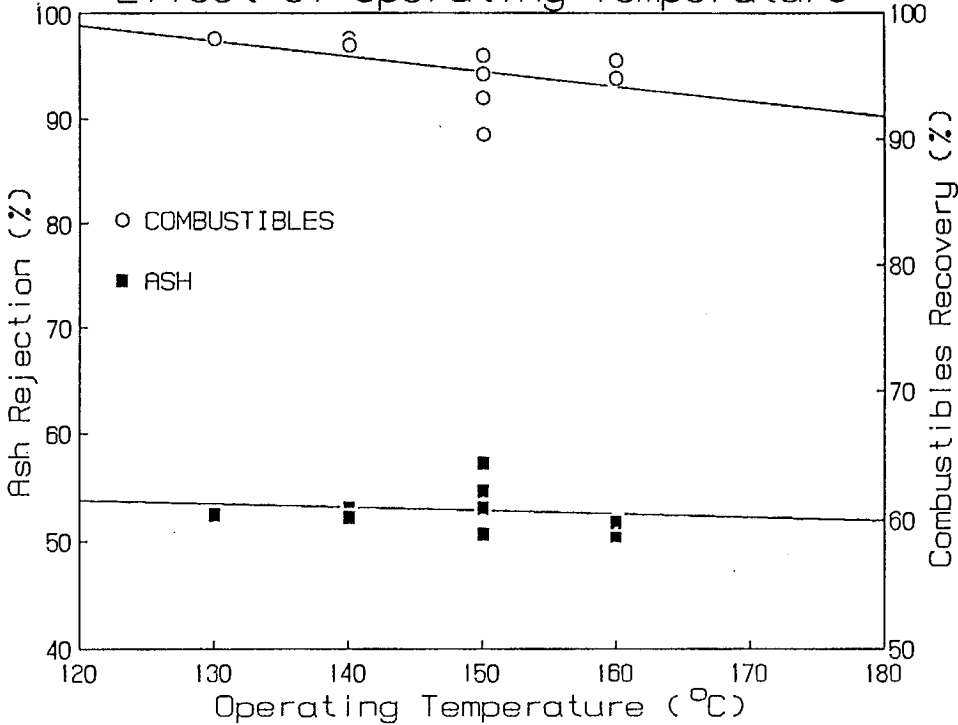


Figure 4  
Effect of Processing Time at High Shear Mixing

