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Beneficiation of coal–heavy oil coprocessing residue by oil-phase agglomeration*

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Surface characterization techniques (scanning electron microscopy and X-ray photoelectron spectroscopy) were used to examine toluene-insoluble solids from coprocessing of coal and heavy oil in order to evaluate their physical separation potential. Washability studies, using float–sink tests, were also carried out to provide an empirical estimate of the ash separation attainable. On the basis of the results, agglomeration tests were performed in an attempt to optimize the beneficiation of the organic matter in the residue pitch. Ash rejection in these tests ranged from 30 to 40%. Analysis of the ash from the agglomerated and reject fractions suggested that most of the iron was retained in the agglomerates.

(Keywords: coal–oil coprocessing; residues; oil-phase agglomeration)

The liquid-phase agglomeration technique developed at the National Research Council of Canada can play a major role in the beneficiation of finely divided carbonaceous solids[1–4]. The principle of selective liquid-phase agglomeration is the preferential wetting of a specific solid component in liquid suspension, by a second, immiscible liquid. The amount of second, or bridging liquid, controls the formation and growth of agglomerates formed from the selected component. Surface tension forces in the capillaries between the agglomerated particles bind them together. This technique has the advantage of rejecting non-wetted solid impurities while maintaining high recovery of combustible carbonaceous material.

The work described here concerns the beneficiation of the solid residue from a coal–heavy oil coprocessor. In the coprocessing reactor an iron sulfate catalyst precursor is eventually converted to pyrite or pyrrhotite under severe hydrogenation conditions. Iron sulfides cannot be readily separated from a hydrophobic matrix, owing to their hydrophobic character[5]. This fact may be utilized advantageously in de-ashing of vacuum pitch, where it is beneficial to leave the iron compounds in the cleaned oil, thereby reducing catalyst makeup requirements. Surface chemical characteristics play an important role in the selective separation of one component from a complex mixture such as coprocessing residue[3]. In this work, the surface characteristics of toluene-extracted solids from coprocessing residue were studied, using SEM and XPS, to evaluate the potential for their separation by means of oil phase agglomeration.

Ash dissemination, size and degree of liberation are also primary determinants for separation. The degree to which a carbonaceous material can be beneficiated by agglomeration is limited by the extent to which the ash can be liberated from the material[5]. Washability characteristics derived from float–sink analysis and specific gravity fractionation[6] usually provide a good measure of the degree of dissemination of the ash associated with coal. In this investigation, float–sink tests on the toluene-extracted solids from coprocessing residue provided an empirical measure of ash separation attainable. The selective de-ashing of pitch residue from a coprocessing reactor was attempted by selectively agglomerating the hydrocarbon-rich fraction, using a suitable oil as the bridging liquid. A number of conditioning treatments to improve ash removal were also examined.

EXPERIMENTAL

Materials

The vacuum residue was obtained from a CANMET bench-scale coal–oil coprocessor. The composition of this test sample is reported in *Table 1*. Pitch samples (100 g) were dispersed in distilled water (500 ml) and ground using 2 kg of 6 mm zirconia balls in a 25 cm porcelain ball mill. The average particle size of the ground material was $9.8 \pm 1.4\ \mu m$ as measured with a Malvern particle sizer.

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Surface analysis

Scanning electron micrographs were recorded using samples coated with a thin carbon layer to impart conductivity. X-ray photoelectron spectroscopy (XPS) was performed with monochromatic Al Kα radiation and dry samples pressed into indium foil. Survey spectra were collected using a pass energy of 188 eV, and high-resolution spectra were recorded with a 22 eV pass energy. An electron flood gun was used to neutralize the charge during the experiment. Bonding energies were referenced to the carbon–carbon bond, which was assigned a bonding energy of 284.6 eV. Atomic compositions were estimated using standard programs provided with the instrument. During analysis, the pressure inside the instrument was always $< 5 \times 10^{-9}$ torr (0.67×10^{-6} Pa).

Float–sink tests

Triplicate samples of ground coprocessing residue were extracted with toluene in a Soxhlet apparatus. Each sample was wrapped in three layers of Whatman No. 2 filter paper to ensure that no solids were lost into the toluene. The toluene extract was diluted and centrifuged to ensure that no solids were lost during extraction. Three separate extractions with fresh solvent were made on each sample to ensure removal of all soluble organic material.

Float–sink tests on the extracted solids were carried out using Certigrav liquids having specific gravities of 1.3, 1.4, 1.5, 1.6, 1.8, 2.0 and 2.2. Each separated fraction was analysed for ash using a Leco proximate analyser. Size analyses were conducted on the extracted residue and on the specific gravity fractions using a Malvern particle sizer.

Agglomeration

A slurry containing 10–20 g of ground pitch was first conditioned with an appropriate reagent by agitation in a blender at 250 rev s^{-1} for 1 min; thereafter the agitation rate was reduced to 150 rev s^{-1}. Bridging liquid was then added dropwise, with mixing, until discrete agglomerates formed. At this stage the blending rate was increased to 200 rev s^{-1} for 2–3 min to facilitate ash liberation. The pitch agglomerates were then separated from the suspending aqueous phase by screening on a 149 μm sieve. The product was washed several times with distilled water, dried at 100°C and then ashed to determine the degree of beneficiation. The aqueous phase (< 149 μm fraction) was returned to the blender and the residual carbon was floated using a frother. The froth was carefully removed, dried and designated as secondary product. The remaining aqueous phase was transferred to a preweighed glass beaker and the contents were dried in an oven at 110 ± 10°C to constant weight. The difference between the final and initial weights of the beaker was taken as the amount of aqueous-phase ash.

Table 1 Composition (wt%) of CANMET coprocessing residue

C	77.5
H	6.2
N	1.2
S	4.4
Al	4.3[a]
Fe	20.5[a]
Si	3.0[a]
Toluene-extracted solids (TES)	29.7 ± 1.3
Ash at 600°C (feed)	11.8
Ash at 600°C (TES)	37.3
Average particle size of ash (μm)	10

[a] Ash basis

RESULTS AND DISCUSSION

Washability studies using float–sink tests are routinely used to determine the relative ease of separation of coal from refuse[6]. These studies are usually helpful in determining the efficiency of a separation process or what might be done to improve its performance. The authors' experience with coal beneficiation using oil-phase agglomeration is also consistent with the usefulness of these tests in determining the suitability of this technology for cleaning different types of coals.

Float–sink tests

The float–sink process can be subdivided into two processes involving ash liberation and coal cleaning[6]. In the liberation process, the bonds between coal and impurities are fractured. In the separation process, the resulting liberated particles are sorted using a dense medium. One fraction is a float product rich in coal; the other is a high-ash reject. These tests provide information on the theoretically attainable separation. However, in actual coal cleaning processes the separation may be incomplete; some sink material always reports to the clean coal product and some float material to the refuse[7].

Table 2 summarizes the float–sink analysis data for the toluene-extracted solids from coprocessing residue. No material floated at < 1.4 specific gravity. As the specific gravity was increased from 1.5 to 2.2, successive samples of float material, with increasing ash contents, were removed from the starting material. The quality of these separated fractions was very different from that of

Table 2 Float–sink data (wt%) for toluene-extracted solids from coprocessing residue

Specific gravity (1)	Individual fractions			Cumulative floats			Cumulative sinks			Ordinate
	Yield (2)	Ash (3)	C (4)	Yield (5)	Ash (6)	C (7)	Yield (8)	Ash (9)	C (10)	Z (11)
1.4 × 1.5 float	12.3	3.1	19.1	12.3	3.1	19.1	100	37.3	100	6.2
1.5 × 1.6	1.5	10.1	2.1	13.8	3.9	21.2	87.7	42.1	80.9	13.1
1.6 × 1.8	8.3	19.0	10.7	22.1	9.5	31.9	86.2	42.7	78.8	18.0
1.8 × 2.0	17.8	26.0	21.0	39.9	16.9	52.9	77.9	45.2	68.1	31.0
2.0 × 2.2	25.7	39.4	24.8	65.6	25.7	77.7	60.1	50.8	47.1	52.8
Sink 2.2	34.4	59.4	22.3	100	37.3	100	34.4	59.4	22.3	82.8

Table 3 Comparison of float–sink data for toluene-extracted solids (TES) from coprocessing residue and unreacted feed coal

Specific gravity	Yield (wt%)		Ash (wt%)		Carbon (wt%)		Carbon:ash ratio	
	TES	Feed coal	TES	Feed coal	TES	Teed coal	TES	Feed coal
1.4 × 1.5 float	12.3	50	3.1	12.0	30.6	53.4	9.9	4.5
1.5 × 1.6	1.5	40.3	10.1	18.9	27.6	39.6	2.7	2.1
1.6 × 1.8	8.3	4.6	19.0	23.7	25.4	4.3	1.3	0.18
1.8 × 2.0	17.8	3.1	26.0	44.0	23.2	2.1	0.9	0.05
2.0 × 2.2	25.7	0.5	39.4	64.2	19.0	0.2	0.48	0.003
Sink 2.2	34.4	1.5	59.4	80.4	7.4	0.4	0.21	0.005

Table 4 Representative properties of coal lithotypes[a]

Lithotype	General appearance	Specific gravity	Ash (wt%)
Vitrain	Uniform shiny black	1.3	0.5–1 (mainly plant ash)
Clarain	Laminated: composed of shiny & dull bands	1.3	0.5–2.0
Durain	Dull, nonreflecting, poorly laminated	1.25–1.45	1-5 (much extraneous ash)
Fusain	Charcoal-like fragments	Soft 1.35–1.45, hard 1.6	5–10

[a] Ref. 9

typical coprocessing feed coals. In all cases, the ash content of the material separated at any specific gravity was much less than that of typical unreacted coal separated at the same specific gravity.

The data for toluene-extracted solids (TES) from coprocessing residue and unreacted feed coal are compared in *Table 3*. Float fractions separated from TES at specific gravity 1.4–1.6 appear to be unreacted coal. A comparison of the ash content of these fractions with that of coal lithotypes[8], *Table 4*, suggests that these fractions are unreacted durain and fusain from the feed coal. The higher carbon content of the remaining fractions from TES, compared with the corresponding fractions from feed coal, suggests that during treatment in the coprocessor the ash liberated is coated with layers of carbonaceous material. This is consistent with the results of SEM surface analysis, which clearly demonstrate that the solid particles of the residue are more or less coated with carbonaceous material.

Experience with oil agglomeration has shown that particles of specific gravity >2 are too hydrophilic to be oil-wetted and collected as agglomerates. If it is assumed that the particles in the coprocessing residue behave similarly, then it should be possible to predict the amount of coprocessing residue solids that can be selectively separated from the oil phase. The data from the float–sink tests are plotted in *Figures 1* to *3* as washability curves. *Figure 1*, cumulative weight per cent floats against specific gravity fraction (*Table 2*: column 5 vs. 1), shows the theoretical yield of washed product from the toluene-extracted solids for any specific gravity of separation. It is evident from this curve that a maximum of 60 wt% of the solids has a specific gravity >2 and therefore may be rejected to the aqueous phase during agglomeration.

Figure 2 shows two curves of cumulative ash in the float product (*Table 2*: columns 5 vs. 6) and the sink

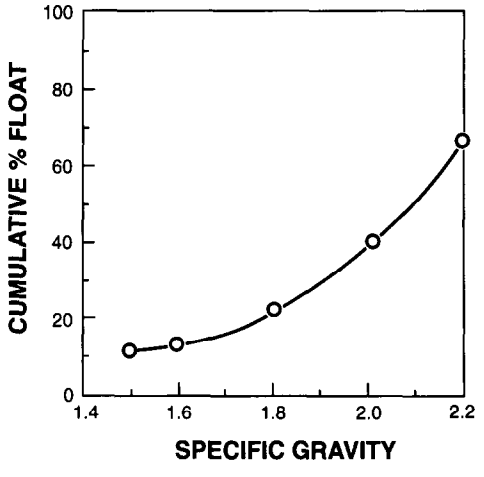

Figure 1 Washability curve, showing yield of float product separable at different specific gravities

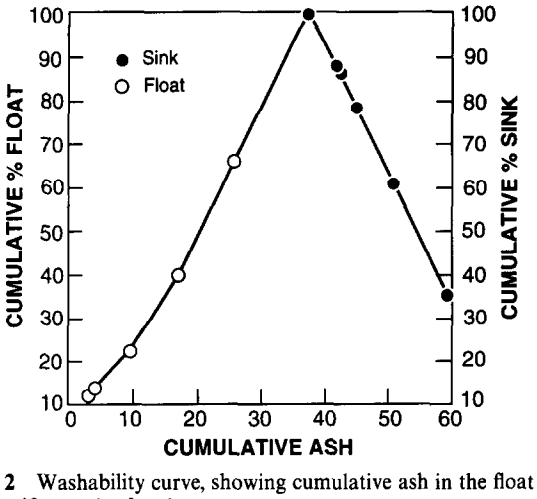

Figure 2 Washability curve, showing cumulative ash in the float and sink specific gravity fractions

fraction (*Table 2*: column 8 vs. 9). These curves give the theoretical percentage ash of the washed product and sink fractions at any given yield of washed product. These plots suggest that ~50 wt% of the total ash is contained in the 60 wt% float product separated at specific gravity >2. These results also suggest that theoretically it might be possible to reduce the ash content of the pitch by 50%, provided that the sample is ground to the ash liberation size.

The plot shown in *Figure 3*, known as the elementary ash curve[6], is a derivative of the cumulative percentage

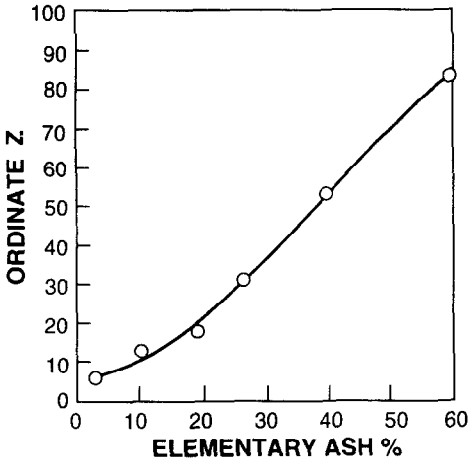

Figure 3 Elementary ash curve, showing rate of change of ash content at different specific gravities

ash in the float material and is intended to show the rate of change of ash content at different specific gravities or yields. The curve is designed to show the highest ash content of any individual particle found in the float product at any specific gravity. It is constructed by finding a new ordinate Z and plotting its value against the actual ash content of each specific gravity fraction as abscissa. Z is given by the equation

$$Z = X + Y/2 \quad (1)$$

where X is the cumulative weight per cent floated for all materials of lower specific gravity and Y is the weight per cent of material at the given specific gravity directly above X. The values of Z are shown in *Table 2*, column 11, and are plotted against the data in column 3.

The slope of the elementary ash curve indicates the relative ease of separation of the carbonaceous fraction from the refuse[6]. Steep slopes represent relatively small ash changes for large differences in yield, whereas gentle slopes indicate easy separation. *Figure 3* suggests that the float product separated at a specific gravity <2 will contain ash that is closely associated with the carbonaceous material and thus will be difficult to separate. The ash from the float product obtained at a specific gravity >2 will be relatively easy to separate. This is consistent with previous unpublished data.

The particle size distribution of the solids separated at various specific gravities was also determined. In general, the high-ash fractions, separated at specific gravity $\geqslant 1.8$, had a smaller mean particle size (10 ± 1 μm) than the low-ash fractions obtained at lower specific gravities (25 ± 5 μm). This observation suggests that the sample must be ground to an ash liberation size of <10 μm to achieve separation.

Characterization of float–sink fractions

The lightest and heaviest fractions (1.5 floats and 2.2 sinks respectively) were analysed by SEM and XPS. *Figure 4* shows the SEM micrographs. Both samples are composed of aggregates of swollen particles. However, differences in the surface texture of the two samples are clearly seen. The 1.5 float aggregates contain primary particles of diameter 0.1–3 μm. The particles are round and their surfaces are smooth; the 2.2 sinks have much rougher surfaces. The aggregates in the 2.2 sinks seem to be platy, clay-like particles. XPS spectra of the two samples are shown in *Figure 5*. Both spectra show similar features. Peaks for S, C, N and O are observed. The atomic concentration of each element is listed in *Table 5*.

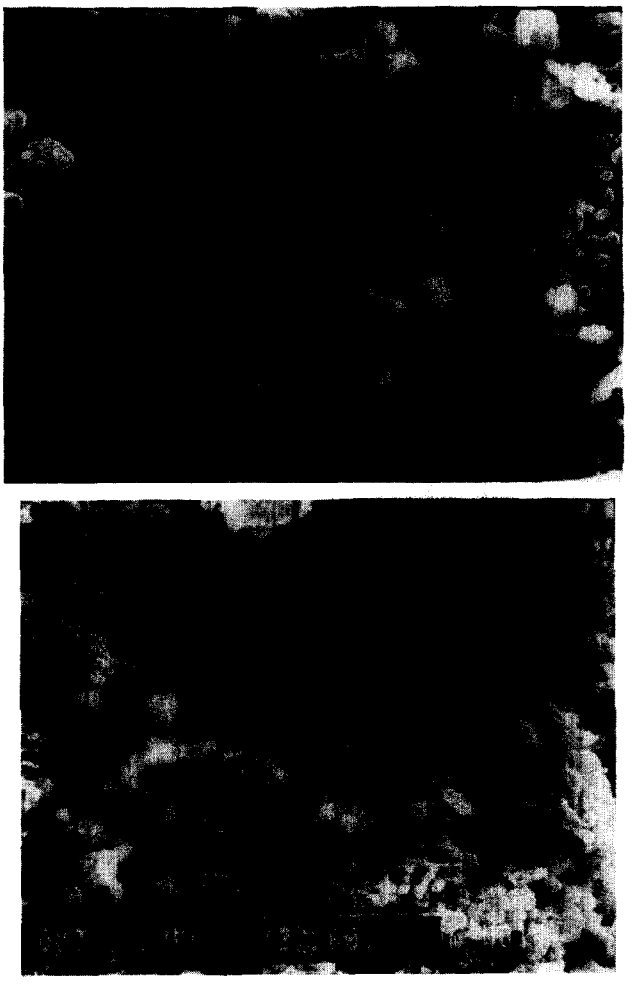

Figure 4 SEM micrographs: a, 1.5 floats; b, 2.2 sinks

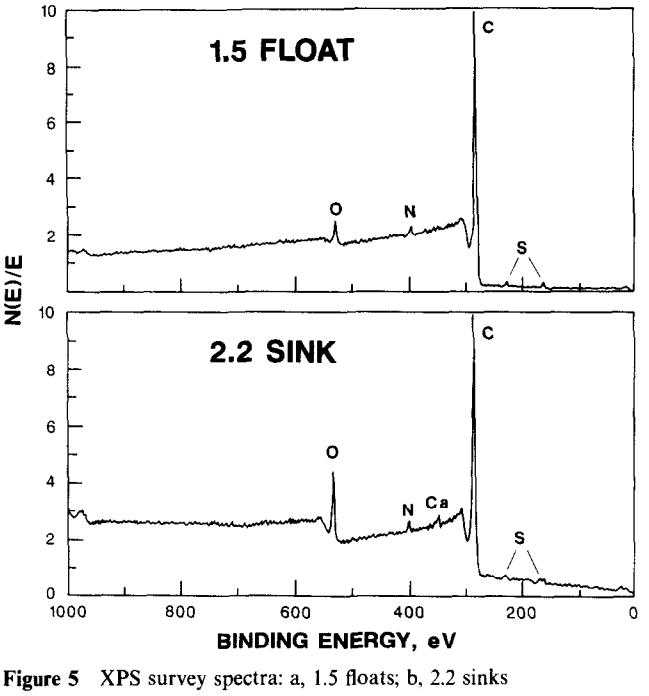

Figure 5 XPS survey spectra: a, 1.5 floats; b, 2.2 sinks

Table 5 Concentrations (at.%) of elements by XPS

	1.5 Floats	2.2 Sinks
C	91.5±0.5	85.2±0.5
O	3.9±0.2	8.7±0.4
N	3.6±0.3	3.9±0.3
S	1.0±0.1	1.7±0.2
Ca	<0.05	0.5±0.1

The combination of SEM and XPS data provides information that allows speculation on the structure of the samples. XPS provides information on the topmost 10 nm layer of the particle. The fact that no Fe, Cu, Al or Si is seen in the XPS spectra for either sample suggests that the particles are covered with a layer of organic material. This organic layer consists mostly of carbon atoms with small amounts of O, N and S, suggesting that functionalities such as C–OH, C=O, HO–C=O, C–NH_2 and C–SH could be present. The high-resolution spectra of the carbon peak also suggest the presence of some functionalities. In the 2.2 sinks some calcium was detected in association with a higher oxygen concentration, suggesting the presence of $CaCO_3$ or oxides of calcium. The fact that Ca is observed at all suggests that the organic layer covering these particles is thinner, or less uniform (patchy), in the 2.2 sinks. These results indicate that the sink fraction is more hydrophilic than the float material, making a separation based on surface selectivity possible.

Beneficiation by oil-phase agglomeration

The results from the washability studies, as well as the surface analysis data, suggest that components in the pitch ash are finely disseminated. Spherical agglomeration techniques are well suited to handle solids in such a finely divided state[5,9]. Several tests were carried out to agglomerate the carbon from a wet, ground pitch sample (average particle size 9.8 ± 1.4 μm) slurried in water, using Stoddard solvent, no. 4 fuel oil or dodecane as bridging liquids. Conditioning agents added to render the surface of the ash particles more hydrophilic included tannic acid, sodium silicate, sodium hydroxide, sodium oxalate, hydrogen peroxide, copper nitrate, iron sulfate and aluminium nitrate. Other variables investigated included slurry pH and the amount and type of collector oil.

Effect of pH. One objective was to explore the possibility of selective separation of the carbon and iron compounds from the siliceous solids. This would allow a reduction in overall catalyst use if the pitch was recycled. Previous work[1] demonstrated that pH has a significant effect on the selective agglomeration of iron in the presence of siliceous matter, provided that the siliceous matter is liberated. Several tests were performed to investigate the effect of pH on the beneficiation of coprocessing residue by oil-phase agglomeration. The pH of the slurry was adjusted by HCl or NH_4OH. The results are summarized in *Figure 6*, which shows weight per cent ash rejection as a function of the pH of the slurry. A dramatic effect of pH was observed. Best ash rejection was achieved at pH 4–5. This improvement in ash rejection may be related to the point of zero charge[10], at which there is in theory no ionized charge at the surface of the particles and therefore a maximum carbon hydrophobicity should appear[11]. This should lead to higher carbon recovery by oil-phase agglomeration. The point of zero charge for coals varies between pH 2 and pH 7 depending on rank, degree of oxidation etc.[12]. The clay minerals illite, montmorillonite and kaolinite, which are usually the major ash constituents of coals, are expected to be negatively charged in this pH range[13], and therefore should remain dispersed in the aqueous phase; this would account for the steep rise in ash rejection seen in *Figure 6*.

Effect of conditioning agents. The effects of various conditioning agents on ash separation are demonstrated in *Table 6*; the results are compared with blanks at the same pH. None of the additives gave results better than that achieved at pH 4–5.

Effect of oil characteristics. The type of oil used as the bridging agent is as important as its concentration in the agglomeration of hydrophobic materials[5]. Lighter, more refined oils, having a high alkane content, are usually more efficient for selective agglomeration, especially when the rejection of siliceous material is an important consideration. In addition to their more

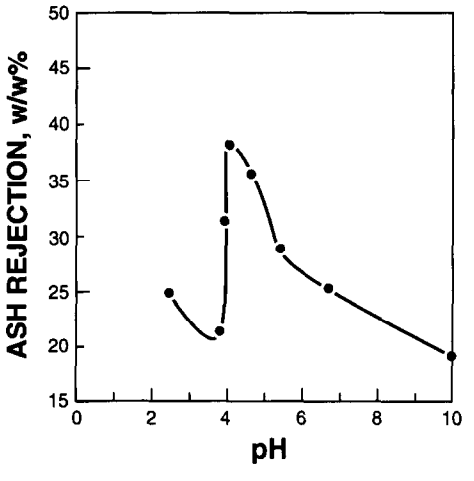

Figure 6 Effect of pH on beneficiation

Table 6 Effect of conditioning agents on the beneficiation of pitch

Test no.	Conditioning agent[a]	pH	Ash rejection[b] (wt%)
1a	Blank	6.7	25.4
1b	Blank	4.2	39.0
2	Tannic acid (16)	6.7	21.2 (25.4)
3	Tannic acid (30)	6.7	35.3 (25.4)
4	Tannic acid (20)	4.0	33.9 (22.5)
5	Tannic acid (30)	8.0	36.2 (23.0)
6	Sodium silicate	7.0	34.7 (25.0)
7	Triethylamine	7.0	27.1 (25.0)
8	Sodium oxalate (4)	–	22.9
9	Hydrogen peroxide	–	30.5
10	Copper nitrate (3.8)	–	23.7
11	Ferric sulfate (5.4)	–	23.7
12	Aluminium sulfate (4.3)	–	28.0

[a] Values in parentheses represent amount of additive in milligrams per gram of pitch; all tests were carried out using Stoddard solvent as bridging liquid

[b] Values in parentheses are data from *Figure 6* at various pH values for tests without conditioning

Table 7 Ash and iron contents of various fractions of pitch under best separation conditions

Sample	Yield[a] (wt%)	Ash[b] (wt%)	Iron content of ash[c] (wt%)
Feed	100	11.8	24±1 (100)
Agglomerated product[d]	90	6.9 (57)	29 (77)
Secondary product	8	34.1 (25)	19 (23)
Aqueous phase ash from agglomeration	2	100 (18)	0 (0)

[a] Calculated on the basis of the total of the three products — agglomerates, secondary products and aqueous-phase ash — recovered
[b] Values in parentheses are percentages of total ash
[c] Values in parentheses are percentages of total iron
[d] > 149 μm product

desirable wetting properties, these lighter oils achieve efficient and economical coating of the organic particles during mixing. Denser, more viscous oils are generally less selective for the rejection of siliceous compounds. However, heavier oils, despite their poor selectivity, are usually a good compromise for the beneficiation of oxidized materials because of their ability to condition surfaces, rendering them hydrophobic. Most of the tests were done using Stoddard solvent, a reference oil normally used for comparison purposes[5]. However, tests were also made with dodecane and no. 4 fuel oil. The best results were obtained with no. 4 fuel oil, with which an ash rejection of 43 wt% was achieved. This compares with the 50% ash rejection predicted using float–sink tests. Ash rejection levels with Stoddard solvent and dodecane, under similar experimental conditions, were 30 and 36 wt% respectively.

Distribution of iron in reject and agglomerated fractions

The quantitative distributions of ash and iron in the feed and various fractions obtained from beneficiation by agglomeration are listed in *Table 7*. Agglomeration under optimum conditions resulted in an ash rejection of up to 43 wt%. ICP-AES analysis of the ash indicated that 77% of the total iron was retained in the agglomerates and 23% lost to the reject. These results suggest that two types of iron species are associated with the pitch. One, derived from iron sulfate catalyst added to the coprocessor and adsorbed at the coal surface, would be expected to remain associated with the coke on the surface of the particles. The other, originating from the mineral matter in the feed coal, would most probably be chemically bound with the inorganic solids. On treatment, the catalytic iron species are more likely to be collected with the agglomerates, whereas the species from the original coal feed would tend to remain with the ash-rich rejects. This is consistent with previous experience of the separation of pyritic sulfur from coal; such sulfur is difficult to separate from the hydrophobic coal matrix because it is itself hydrophobic[14,15]. Preferential collection of catalytic iron with the organic concentrate is beneficial because it could reduce catalyst makeup requirements during pitch recycling, as long as it is available at the particle surfaces.

CONCLUSIONS

The results of washability studies suggest that the ash particles associated with coprocessing residue pitch are finely disseminated and that the ash liberation size is < 10 μm. The data also predict a theoretical ash reduction of ~50 wt%, provided that the sample is ground to liberation size; grinding to finer sizes could produce better results. SEM and XPS results for the gravity-separated fractions suggest that the solids have an external coating consisting mostly of organic carbon. The lower-specific-gravity fractions have a thicker layer of carbon coating than do the heavier fractions. Liquid-phase agglomeration techniques applied to the coprocessing residue pitch for the selective separation of carbonaceous matter achieved >40 wt% ash rejection. Analysis of the ash from the agglomerated product and the reject material suggests that most of the iron from the added catalyst is retained in the agglomerates. Analysis of the ashed agglomerates showed an iron content of 29 wt%, representing 77 wt% of the total iron in the feed. This would be beneficial, as it could reduce catalyst makeup requirements if the pitch were recycled, provided that it occurs at the particle surfaces. The results also show that, compared with the feed material, the cleaned pitch is considerably poorer in siliceous matter.

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