Solid/solid separation by selective agglomeration with agglomerant recovery by thermal desorption

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Abstract

In this study, selective agglomeration is used as a simple physical technique to reduce the ash content of coal tailings, in order to reduce environmental pollution and to increase the value of these materials for combustion and energy recovery. A pilot plant based upon this separation method was constructed, permitting the de-ashing of the tailings to a high level and the recycling of the agglomerant used for these operations by thermal desorption from the agglomerates. The effects of many operating variables have been examined for two different tailings and the efficiency of the process for cleaning tailings and agglomerant recovery has been demonstrated. Moreover, a tailings de-ashing efficiency of 90% was achieved for this process, and a recovery and re-cycling efficiency for the agglomerant of 82% was also realised. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Tailings de-ashing; Selective agglomeration; Thermal desorption; Recycling

1. Introduction

In the past, large reserves of coal resulted in much wasteful mining and preparation practices. Traditionally, coarse-coal cleaning has been rated as an effective, economical method for the reduction of the ash content of raw coal. This method was performed almost exclusively by gravity concentration methods. The efficiency of the process was low. As a consequence, fine coal particles were frequently discarded along with the refuse, which was stored in basins, sometimes for >20 years. This product contained a high ash level (25–50%), but also ≈50–75% combustible matter.

In this study, selective agglomeration was used as a simple solid/solid separation method to reduce ash in coal tailings, in order to reduce environmental pollution and to increase the value of these materials as an energy source through the combustion of the carbonaceous matter.

The treatment of tailings in aqueous suspension consists of separating the carbonaceous fraction from the ash-forming mineral matter. The separation process, based on selective agglomeration, involves the principle of preferential wetting of hydrophobic carbonaceous particles by agglomerants (hydrophobic). This selectivity allows the coal (carbonaceous matter) to be wetted by the agglomerant and the impurities (minerals, trace metals), which are typically hydrophilic, to remain in aqueous suspension. In the presence of an adequate amount of agglomerant and sufficient mechanical agitation, the agglomerant-coated coal particles collide with each other and form agglomerates of a sufficient size to permit separation by screening with a sieve.

Agglomerate growth depends on many process variables and affects the de-ashing and recovery efficiency of coal as well as its moisture content. The following phenomena may affect the agglomeration process [1]: the nature and concentration of the agglomerant, the nature of the coal, the coal particle size, the concentration of coal/water suspension, the agitation duration and intensity, the pH and temperature.

The originality of this study essentially concerns the recovery and recycling of the agglomerant by thermal desorption from the agglomerates and resultant dry carbonaceous matter.

Many studies have already been performed that apply this method to coal [2–8], although the majority of these have been limited to laboratory level, without recovery and recycling of the agglomerant. In this study, a pilot plant was constructed, which allowed the de-ashing of tailings to a high level, and recycling of the solvent (volatile) that was used as the agglomerant. The different factors which influence the process have been examined and the optimal conditions for

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Table 1
Identification and analyses of feed tailings

Type of sailing	Ash content (%)	Density	Elementary analysis				
			C	H	N	S	O
SL 57	42.2	1.70	48.3	2.94	1.05	0.74	4.77
SL 60	33.4	1.61	58.8	3.5	1.24	0.65	2.41

	Major elements in ash (wt.% of tailings)										
	Na	Mg	Ca	K	Mn	Fe	Al	Si	P	Ti	S
SL 57	0.27	1.19	2.13	3.10	0.05	4.40	12.4	26.0	0.10	0.65	0.96
SL 60	0.34	1.17	1.47	3.40	0.04	3.50	13.6	30	0.09	0.66	0.63

the process have been found. The efficiency of the process has also been demonstrated.

2. Materials and methods

The tailings used for this study came from Simbourg in Belgium. Their composition is illustrated in Table 1. The mean diameter of tailing particles was 52 μm, with a maximum size of 500 μm. The ash content of the tailings was 42% for SL 57 and 33% for SL 60.

The ash content in the tailings and agglomerates was determined according to the ASTM D3174 method [9]. Carbonaceous matter recovery was also determined.

A pilot plant (see Fig. 1) was constructed in order to de-ash the tailings using the technique of selective oil agglomeration and recovery of oil by desorption from agglomerates in order to recycle the oil used for the agglomeration process.

The pilot plant can treat ≈10 kg/h of tailings. The process can be divided into five phases.

2.1. Tailings/water suspension preparation

In this phase, the solid is mixed with water and agitated in order to wet the particles and to homogenise the suspension. The agitation is performed using a high-speed double-turbine agitator and was driven by compressed air, in a tank with a 50-l capacity.

2.2. Selective agglomeration

The agglomeration is carried out in a 250-cm^3 agglomeration vessel (see Fig. 2), in which the tailings–water suspension and the agglomerant are continuously introduced, using two volumetric pumps. The flow rates of both the suspension and agglomerant are measured using an electromagnetic
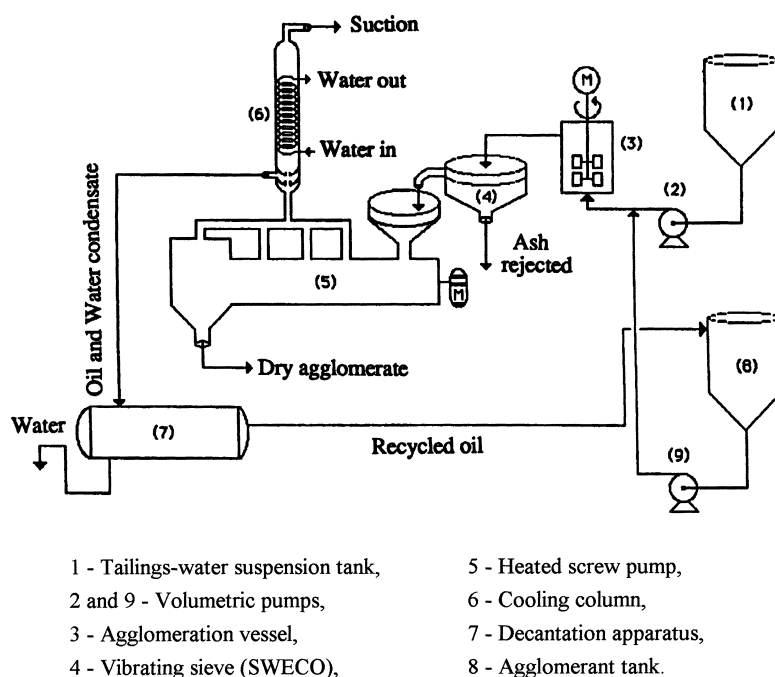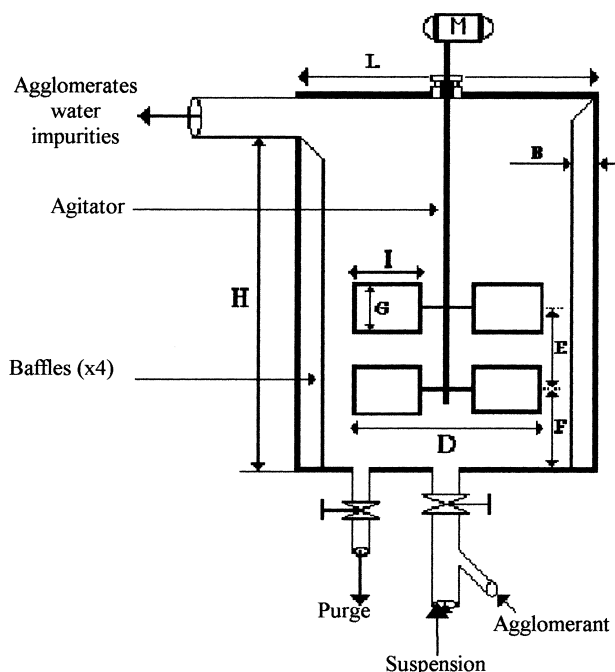

1 - Tailings-water suspension tank,
2 and 9 - Volumetric pumps,
3 - Agglomeration vessel,
4 - Vibrating sieve (SWECO),
5 - Heated screw pump,
6 - Cooling column,
7 - Decantation apparatus,
8 - Agglomerant tank.

Fig. 1. Process flow plan of continuous pilot plant.

	L	H	B	F	D	E	G	I
Standard	/	/	L/10	L/7	L/2,3	L/3,25	L/7	L/7
Agglomeration vessel (mm)	100	35	10	14,3	43,5	30	14,3	14,3

Fig. 2. Vessel of continuous agglomeration process.

flow meter and a rotameter, respectively. The agglomeration period and agglomerant/tailings mass ratio are imposed by the flow regulation. Two high-shear impeller mixers are used for an efficient agglomerant dispersion.

2.3. Separation of agglomerates from mineral parts by screening

The formed agglomerates and free minerals in the water are conducted from the bottom of the agglomeration vessel to a vibrating sieve (SWECO). The mineral particles pass through the screen, while the agglomerates are collected by the screen and are conducted to a heated screw pump. At this point in time, the agglomerates contain both water and the agglomerating agent.

2.4. Agglomerant recovery by desorption

The screw pump is heated using electrical resistance (see Fig. 3). During the residence time in the pump, the agglomerant and water evaporate, forming an azeotropic mixture. A cooling tower allows the recovery of the water/agglomerant vapour mixture and facilitates its condensation. The condensate is conducted to the decantation apparatus for separation,

and the dry agglomerates are recovered in the output of the heated screw pump.

2.5. Agglomerant–water mixture separation

The agglomerant–water mixture is recovered in the decantation apparatus, which permits the separation of the two liquid phases on account of their difference in density. The solvent's density is less than that of water, hence, it is recovered from the top of the decantation apparatus, whereas the water is recovered from the bottom. The recovered water is retained for preparation of the tailings suspension, while the solvent is sent to the agglomerant storage tank for recycling.

3. Results and discussion

Different agglomerants have been tested (heptane, hexane, pentane, heptanol, hexanol) and their performances evaluated. Other factors, such as residence time in the agglomeration vessel, concentration of tailings in the water suspension, agglomerant concentration, and other factors affecting the recovery of the agglomerant from the agglomerates have also been studied.
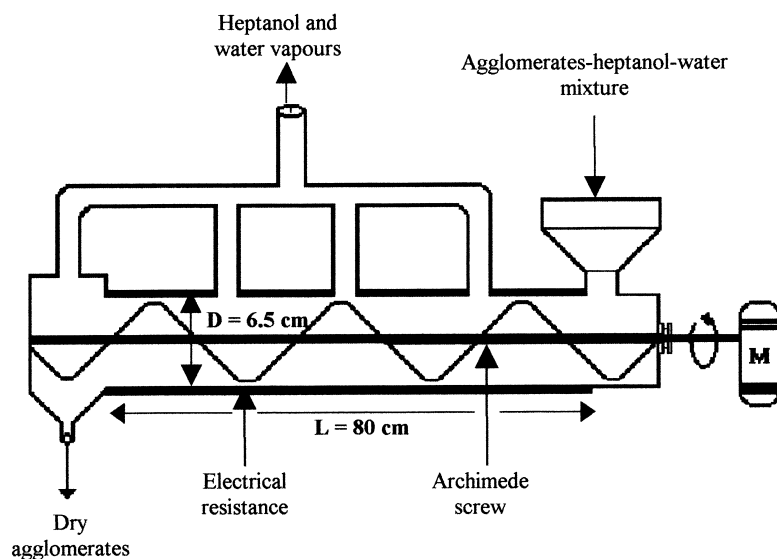
Fig. 3. Heated screw pump.

Each of the operating conditions has been examined in relation to the major objectives of coal preparation, namely: (a) reduction of the ash content in tailings, (b) recovery of the purified coal while rejecting impurities, (c) recovery and recycling of the oil used for the agglomeration.

3.1. Effect of agglomerant nature

The viscosity and density of the agglomerant have an important influence on the agglomeration quality and efficiency of de-ashing. The influence of several agglomerants upon ash reduction and carbonaceous matter recovery has been studied for SL 57 tailings, with an initial ash content of 42% (w/w). The experimental conditions of the agglomeration process are depicted in Table 2. The results obtained are stated in Table 3.

Tailings cleaning increases when there is an increase in the density and viscosity of the agglomerant (see Table 3). Heptanol and hexanol produced the best results for tailings purification, with about 11.3 and 12.8% residual ash contents, respectively. It should also be noted that an increase

in the carbon content of the aliphatic hydrocarbons reduces the de-ashing of tailings.

Carbonaceous matter recovery also depends on the nature of the agglomerant, as can be observed in Table 3. A very low agglomerant viscosity decreases the coal recovery, due to a lower agglomerant adhesion or dispersion, respectively, during agitation. There is an optimum range for the density [10], which allows a better adhesion of particles with agglomerant and a higher degree of agglomeration. Mazzone et al. [10] illustrated the fact that, under certain conditions, a continuous deformation of the liquid bridges occurs. In this case, the strength of the bridges (attraction force due to the presence of the liquid) and consequent agglomerate growth depend not only on the shape of the interface, but also on the fluid viscosity.

Aliphatic alcohols exhibit a higher carbonaceous matter recovery and a better tailings purification than aliphatic hydrocarbons [14]. It seems that alcohols permit the collection of particles that are less hydrophobic as in the case of weathered or oxidised coal, which was the case for the tailings used for this study.

Heptanol was used for the next set of experimental trials, especially on account of its good efficiency for tailings

Table 2
Experimental conditions for studying SL 57 tailing

Type of tailings	Tailing SL 57
Tailings size	$0 \times 500 \,\mu m$
Mean size	$52 \,\mu m$
Temperature	$20°C$
Suspension density	0.30 tailings/water (w/w)
Time of tailings wetting	90 s
Ratio of oil/tailings (w/w)	0.15
Agitation period	60 s
Agitation speed	2000 rpm
Sieve size	$90 \,\mu m$

Table 3
Effect of oil nature on ash content and coal recovery for SL 57 tailings[a]

Ser. No.	Type of oil	Density (20°C)	Viscosity (C_p) 20°C	Coal recovery (%)	Ash content (%)
1	Pentane	0.63	0.240	53.00	10.75
2	Hexane	0.65	0.326	74.35	12.45
3	Heptane	0.66	0.409	79.92	13.21
4	Heptanol	0.82	5.81	89.56	11.31
5	Hexanol	0.85	4.58	85.55	12.83

[a] Tailings suspension concentration=30%; agglomerant/tailings ratio= 0.15; and agglomeration time=60 s.
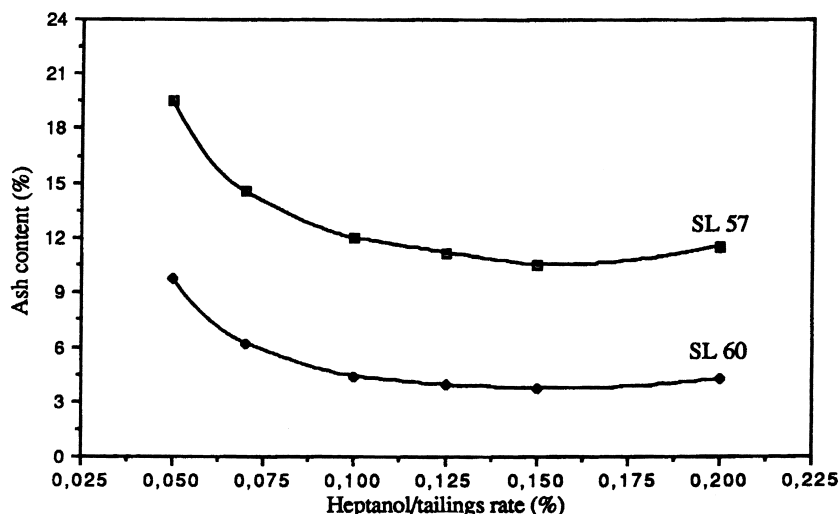
Fig. 4. Ash content as a function of the agglomerant/tailings ratio (tailings suspension concentration=30%, agglomeration time=60 s).

purification. It also displayed the best results for carbonaceous matter recovery during agglomeration and had a low temperature of desorption from agglomerates that form an azeotropic mixture with water.

3.2. Effect of heptanol concentration

Agglomerant concentration is a critical factor for agglomerate growth. Both ash reduction and coal recovery are functions of an optimum oil concentration, which gives significant differences between agglomerate and mineral matter sizes.

The ash content of agglomerates decreases with an increase of oil concentration up to an optimum value (see Fig. 4). Above the optimum value, the agglomerate sizes

become very large and may trap some mineral matter. The optimum value of de-ashing for the studied coals was obtained between 0.125 and 0.175 of the heptanol/tailing ratio for SL 57 and SL 60 tailings.

For a good recovery of carbonaceous matter, a minimum amount of heptanol was also necessary for a good coal recovery, as is indicated in Fig. 5. For the two tailings tested, a minimum heptanol/tailings ratio of 0.15 is necessary, in order to obtain a 90% recovery of carbonaceous matter.

3.3. Effect of agitation intensity and agitation time

Agitation for agglomeration is performed with a high-speed agitator equipped with a high-shear impeller. It
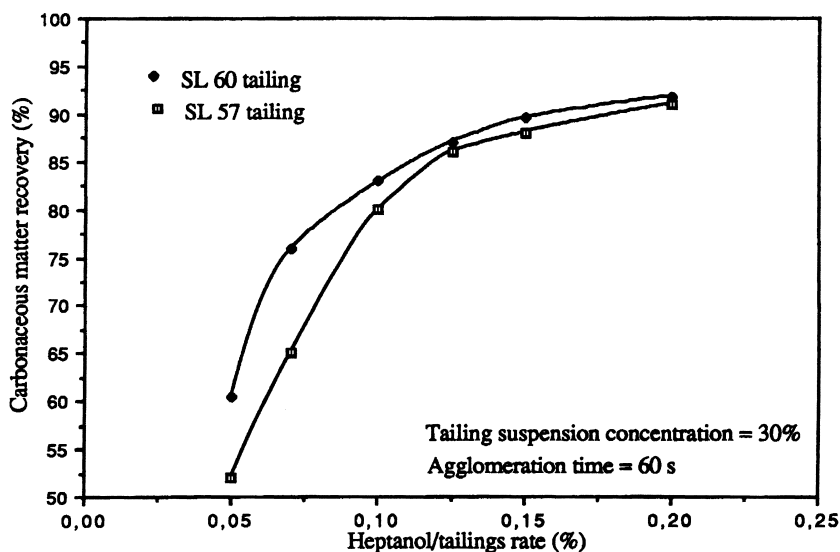

Fig. 5. Carbonaceous matter recovery as a function of the agglomerant/tailings ratio (tailings suspension concentration=30%, agglomeration time=60 s).
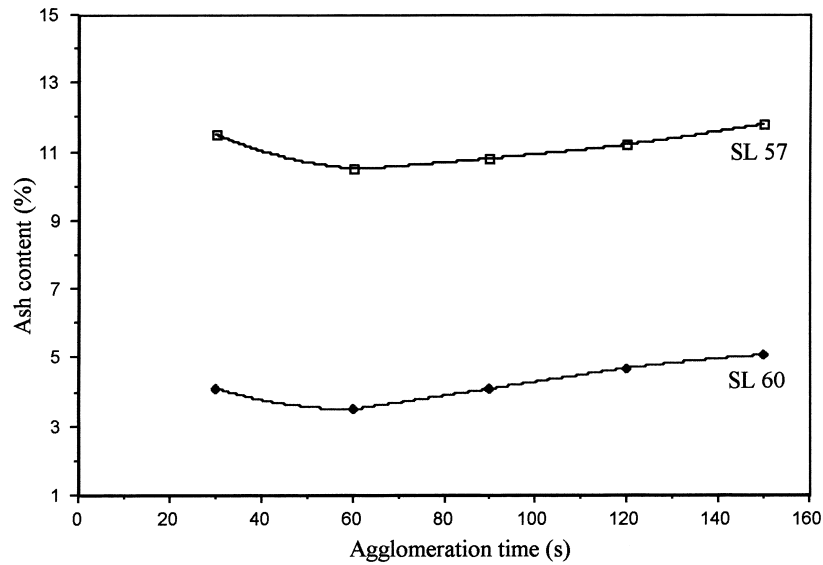
Fig. 6. Ash content as a function of the agglomeration time for SL 57 and SL 60 tailings (tailings suspension concentration=30%, heptanol/tailings ratio=0.15).

causes the agglomerant to disperse effectively into the tailings/water suspension, and it facilitates the contact of agglomerant with coal particles.

The growth of agglomerates increases with agglomeration time [6]. Ash reduction increases with agitation time until an optimum value is reached (see Fig. 6). A prolonged agglomeration time substantially increases the agglomerate size and greatly increases the risk of trapping impurities within the agglomerates. The optimum agglomeration time obtained for the two tested tailings (SL 57 and SL 60) is ≈60 s.

3.4. Effect of tailings/water suspension concentration

The concentration of coal in water is generally not a critical factor in selective agglomeration, according to Swanson et al. [11]. However, it can affect the quality of coal cleaning, according to our results. A high concentration of coal does not give the necessary free space for the mineral matter to be separated from the coal particles. Some impurities are then imprisoned among the coal particles during agglomeration and there is an increase in the ash content of the coal (see Fig. 7).
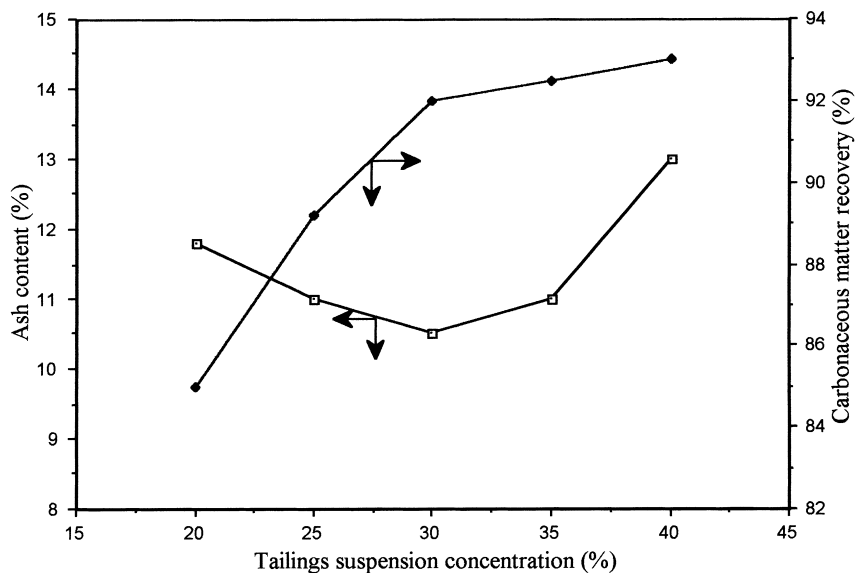

Fig. 7. Ash content and carbonaceous matter recovery as a function of the SL 57 tailing suspension concentration (heptanol/tailings ratio=0.15, agglomeration time=60 s).

A large dilution would require prolonged mixing to ensure that contacts between particles occur. Very low concentrations of coal are undesirable, due to the large volume of water which is required. A mass concentration of ≈30% of tailings in water has produced favourable results and has, therefore, been adopted for this stage of the experiments. This concentration is considerably greater than that used in the flotation process for coal cleaning [3,12,13].

3.5. Heptanol–water mixture recovery

The last phase of the process is the agglomerant (heptanol) recovery by desorption of the azeotropic mixture (heptanol/water) included in agglomerates. In order to optimise the vaporisation operation, the process was studied using a fixed optimal residence time of 4 min for the solids in the screw pump. Air was drawn through the heated screw pump and condenser by applying suction at the condenser outlet. The air flow rate was measured using a rotameter and the suction pressure was measured by the use of a water gauge manometer.

The results obtained are presented in Table 4 and illustrate the effect of the suction pressure and air flow on the desorption efficiency. It is possible to observe that residual moisture decreases with an increase in suction, because of the improvement in mass transfer.

There was, however, a decrease of condensation in the cooling column when the suction exceeded 1.5 mm of water gauge (Table 4). When less suction was applied, less heptanol and water were vaporised.

The results of desorption from the agglomerates made from SL 60 tailings are shown in Table 5. The residual

water and heptanol were very low at 4.34% (3.45% heptanol and 0.9% water) when an optimum suction (1.5 mm water gauge) and the temperature within the heated pump (solid temperature) of 105°C were applied.

It is to be noted that desorption and recovery efficiency were calculated as follows:

$$\text{Desorption efficiency} = \frac{\text{MC2}}{\text{MC1}} \times 100 \text{ and}$$

$$\text{Recovery efficiency} = \frac{\text{MR}}{\text{MC1}} \times 100$$

where MC1 represents the heptanol and/or water mixture content in the agglomerates before desorption, MC2 the heptanol and/or water mixture content in the agglomerates after desorption and MR the heptanol and/or water mixture recovered after evaporation and condensation.

4. Conclusions

Our study shows that selective agglomeration can be used as an effective physical method to reduce the ash content of tailings to a lower level and to recover the agglomerant by desorption from the agglomerates for recycling.

Improvements in ash reduction and carbonaceous matter recovery efficiencies were achieved by optimising the parameters studied. A mass concentration of ≈30% tailings in water and 60 s agitation time, combined with an agitator speed of 2000 rpm, produced favourable results. Heptanol was utilised in view of its high efficiency for tailings purification and carbonaceous matter recovery and its ease of desorption from the agglomerates. The optimal heptanol/tailings ratio for the tailings studied, in order to obtain a minimal ash content and a maximum combustible matter recovery is between 0.13 and 0.17.

The used agglomerant was recovered by desorption from the agglomerates as an azeotropic mixture (heptanol–water), then separated by decantation. Altogether, 82% of the heptanol used was recovered and recycled.

Table 4
Desorption and recovery efficiency as a function of the suction pressure (tailing used SL 60; temperature of the solid within the screw pump 105°C)

Suction pressure (mm of water)	Air flow rate (m³/h)	Residual moisture (%)	Desorption efficiency (%)	Recovery efficiency (%)
0	0	9.6	68	67
0.1	18.3	8.1	73.5	73
0.5	27.5	4.86	83.8	83.4
1.5	35.6	4.35	89.4	87.8
2.6	42.7	3.74	87.5	78.6
4.3	50.9	3.41	88.6	75.4
8.2	69.2	3.25	89.2	70.2

Table 5
Results of the heptanol and water desorption and recovery from SL 60 tailings agglomerates

	Residual water and heptanol in the dry agglomerates (%)	Evaporation efficiency (%)	Recovery efficiency (%)
Global	4.35	89.41	87.78
Heptanol	3.45	83.62	82.09
Water	0.9	95.49	93.76

References

[1] G.A. Robbins, R.A. Winschel, C.L. Amos, F.P. Burke, Fuel 71 (1992) 1039–1046.

[2] A. N'kpomin, A. Boni, G. Antonini, O. François, Chem. Eng. J. 60 (1995) 49–54.

[3] C.E. Capes, R.J. Germain, in: Y.A. Liu (Ed.), Physical Cleaning of Coal, Marcel Dekker, New York, 1982, pp. 293–351.

[4] C. Skarvelakis, M. Hazi, G. Antonini, 4ème Congrès Français de Génie des Procédés, Grenoble, France, 7, 29, 21–23 September 1993, pp. 239–244.

[5] .D. V Keller Jr., W. M Burry, Coal Prep. 8 (1990) 1–17.

[6] C. Skarvelakis, Ph.D. thesis, Compiegne University of Technology, France, 1992.

[7] P. Hopital, Ph.D. thesis, Compiegne University of Technology, France, 1987.

[8] .C.E. Capes, K. Darcovich, Powder Techn. 40 (1984) 43–52.

[9] ASTM D3174, Annual Book of ASTM Standards, Part 26, Philadelphia, PA, 1975.

[10] D.N. Mazzone, G.I. Tardos, P. Pfeffer, Powder Techn. (1987) 71–83.

[11] .A.R. Swanson, C.N. Bensley, S.K. Nicol, Agglomeration 77 (2) (1977) 939–951.

[12] R.P. Killmeyer, Coal Mining, Sep., 1985, pp. 45–49.

[13] R.E. Zimmerman, Froth flotation in coal preparation, in: J.W. Leonard, D.R. Mitchell (Eds.), 3rd Edition, AIME, New York, 1968, pp. 10–73.

[14] A. Bensakhria, DEA Report, Compiegne University of Technology, France, 1993.