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Author(s)	Chiba, Tadatoshi; Yamagishi, Tohru; Moritomi, Hiroshi et al.
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Preferential Separation of Low Ash-Content Coal Particles by CO_2 Absorption and Liquid Fluidization

Tadatoshi CHIBA, Tohru YAMAGISHI, Hiroshi MORITOMI
Yuzo SANADA, Tadao JIMBA*, Masatoshi NAKAI* Tomoyasu ESAKI*
and Yukuo KATAYAMA**

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Abstract

Preliminary experiments were carried out in an effort to evaluate the effect of CO_2 absorption on the separation process of coal particles with different ash contents. A mixture of coal particles and water was exposed to CO_2 gas under pressures up to 2.0 MPa and then released to atmospheric pressure. This resulted in the formation of froths of CO_2 gas on the surface of each coal particle. The froth formation depended on the ash content of individual particles and some of low ash-content particles floated onto the water surface. However, since the amount of the particles recovered from the water surface remained only a few weight percents, liquid fluidization was applied to classify the CO_2 treated particles, the apparent density distribution of which was broadened as a result of the selective froth formation. Weight fractions of the coal particles and ash elutriated from the bed at increasing liquid velocities were measured for several kinds of coals having different size ranges and ash contents. An improved separation by the CO_2 treatment was obtained for some coals.

1. Introduction

Ash content of coal particles must be reduced efficiently prior to the slurry transportation and conversion processes. In recent years much effort has been made to improve existing classification methods such as washing, dense medium cyclone separation, froth flotation, oil agglomeration, selective flocculation and high-intensity magnetic separation. In froth flotation, air pressurization and use of surface-active agent have been applied in such a way as to generate increarirgly tiny foams of air in water and to pick up lower ash-content particles preferentially. In such cases the air foams are usually still much larger than coal particles and only a very limited increase in the separation efficiency can be expected.

Thus, this paper describes a new method[1] to improve the efficiency. The method utilizes two facts i) that carbon dioxide is absorbed preferentially onto the carbonaceous portion of coal[2],[3] and ii) that the expansion of a liquid fluidized bed depends on the size and

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Metals Research Institute
*Office of Business Planning and Development, Kubota Ltd., Osaka, Japan
**Research and Development Division, Institute of Applied Energy, Tokyo, Japan

the apparent density of the bed materials.[4),5)] When a mixture of coal particles that have a different ash content is pressurized by CO_2 gas in water, more absorption occurs on the particles with a lower ash content (see Figure 1). Hence, subsequent depressurization to atmospheric pressure results in an increasing froth formation on the outer surface or within the pores of the individual particles with a lower ash content. If the apparent density of a particle is lowered to less than the density of water, the particle floats on the water surface.

The depressurization may also result in a rise of a swarm of very tiny bubbles in water which are generated by CO_2 dissolved in water. These bubbles may entrain some particles by coalescing with froths on the surface of particle and may enhance the flotation to some extent without adding any surface-active agent.

This short paper describes the results from preliminary experiments on a batch flotation by the CO_2 pressurization and depressurization and on a semi-batch classification in a liquid fluidized bed of the CO_2 treated coal particles.

2. Experimental

All experiments were conducted at room temperature. A 50cm^3 stainless steel micro-autoclave was used for batch CO_2 treatment. A mixture of 3 g coal particles and about 20cm^3 water was loaded in the autoclave. Before each run, air in the autoclave was purged by an atmospheric CO_2 gas stream. The autoclave was then pressurized up to a programmed pressure by CO_2 gas from a cylinder. N_2 gas was also used instead of CO_2 for comparison. After the pressure was kept steady for a given time, the autoclave was depressurized to atmospheric pressure and the particles floating near the top surface of water were recovered by suction without disturbing the layer of particles settled at the bottom. A 50cm^3 glass autoclave was also used to visualyze flotation after the depressurization. Size distributions of untreated and CO_2 treated particles were inspected by microscope photographing and no essential difference was found between them in the present pressure range.

The residual mixture was subjected to subsequent classification in a liquid fluidized bed.

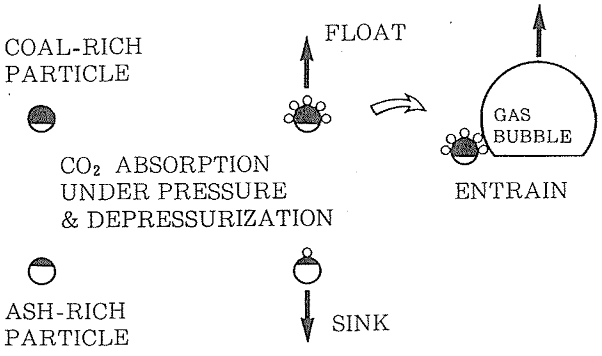


Fig. 1 Conceptual mechanism of selective separation of lower ash-content coal particles.

A schematic diagram of the apparatus is shown in Figure 2. A 20mm i.d. and 500mm high PMMA-resin column with a porous-sintered brass-beads distributor was used for the fluidized bed. Water was fed as the fluidizing liquid to the column through one of the calibrated rotameters and was allowed to overflow at a height of 450mm from the distributor. The liquid velocity was increased step-wise at intervals for complete elutriation of the particles at each velocity. Collection of the elutriated particles was conducted successfully by using a 400 mesh stainless-steel gauze. All collected samples were dried overnight and weighed. Their ash contents were determined on the basis of JIS (M8812−1963) analysis.

Four different coals were so far used. Their properties and the result of ultimate analysis are listed in Table 1. The ash content for each coal depended more or less on its size so that the mean content was determined after each run for all particles recovered. The mean values thus obtained for each size range still varied from run to run. Therefore, the minimum and maximum values of the means values for each size range are shown in the table.

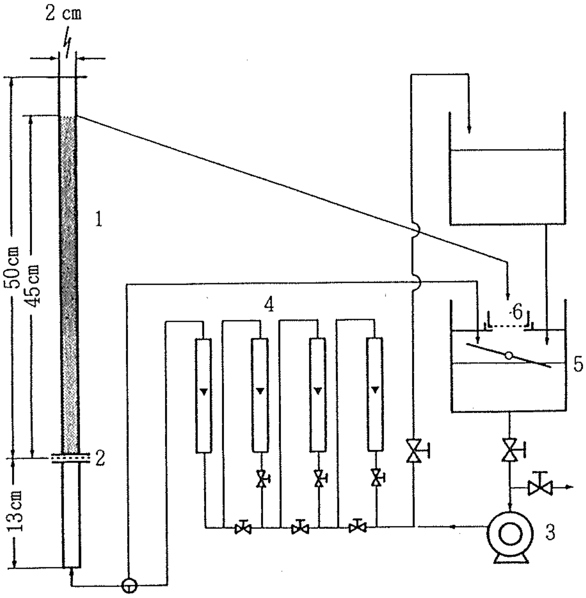


Fig. 2 Schematic diagram of liquid fluidized bed apparatus; 1, fluidized bed column; 2, porous-sintered brass-beads distributor; 3, water circulation pump; 4, rotameters; 5, water feed tank; 6, particle collector.

Table 1 Properties of coal particles so far used

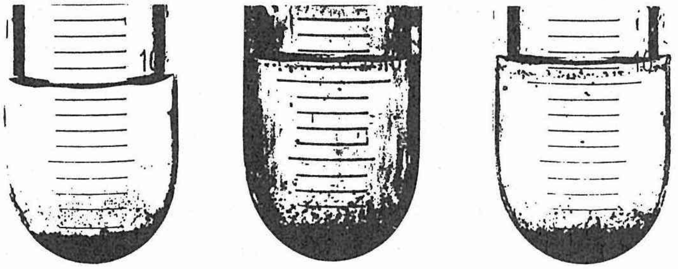
Coal	Particle Size Range [mm]	Ultimate Analysis [d. a. f. wt%]					Mean Ash Content [wt%]
		C	H	N	O	S	
A274	0.250−0.297	82.5	5.9	2.2	8.6	0.8	2.0− 2.3
A149	−0.149						4.1− 4.6
M250	−0.250	81.9	5.6	1.3	10.0	1.2	2.5− 3.1
T399	0.297−0.500	84.7	6.5	1.1	6.5	1.2	12.1−12.4
T112	0.074−0.149						13.2−15.3
W200	0.149−0.250	80.6	5.1	1.9	11.7	0.7	3.3− 3.6
W250	−0.250						7.6− 8.0
W149	−0.149						8.6− 9.7

3. Results and Discussion

3. 1 Batch Flotation

Batch flotation experiments were carried out in a range of gas atmosphere, pressure and holding time under pressure. Photographs in Figure 3 were taken for A274 coal particles after holding them in water for 60 min under 0.50 MPa-CO_2 gas atmosphere and reducing the pressure to normal atmosphere. Immediately after the depressurization no particle movement was observed and all particles were found to stay as a layer in the bottom part of the autoclave. Then, froth formation on the surface of each particle and foam generation in the water initiate simultaneously within one minute. Some particles leave from the top surface of the particle layer and start to rise through the water. When the particles reach the top surface of water, some of them remain there while others descend by leaving the froth on the water surface. The latter repeats the froth formation during descent through water or settling on the surface of the bottom layer, resulting in circulation of them in the water. The particles remaining on the top surface of water gradually increase with time but no significant difference in their amount can be seen after 10 min, as shown in the photographs (2) and (3).

In Figure 4 the weight fraction of particles recovered near the top surface of the water is shown for A274 coal as a function of the holding time under different gas atmospheres and pressures. The ash contents of these "floating" particles were less than 1 wt%. For CO_2 gas treatments the weight fraction seems to increase up to a few weight percents with the holding time within 20 min and to decrease with the pressure from 0.5 to 2.0 MPa. However, the time and pressure effects are rather ambiguous since these data were obtained with a poor reproducibility; for example, the observed weight fractions for 30 min treatments by 0.5 MPa CO_2 gas fluctuated between 0.018 and 0.043, as shown in the figure. Even with the scattered data, it can be seen that CO_2 is more effective than N_2 for the flotation. This result might reflect the difference of the flotation mechanism between the gases. The



(1) 0 min (2) 10 min (3) 30 min

TIME AFTER DEPRESSURZATION

Fig. 3 Batch flotation of A274 particles after CO_2 treatment at 0.5 MPa for 60 min.

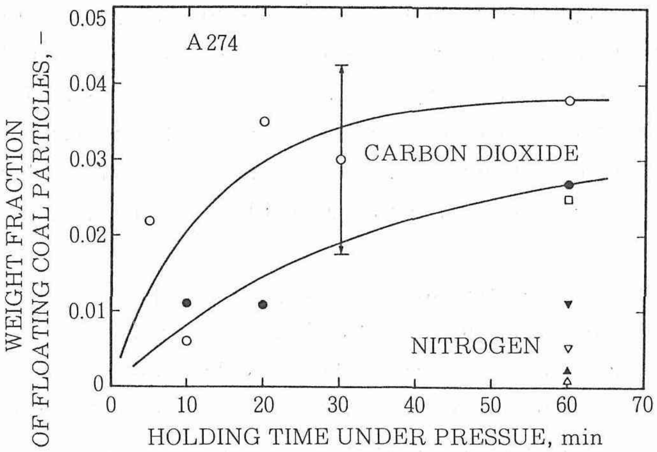


Fig. 4 Change of weight fraction of floating coal particles with time under pressures.

solubility of CO_2 gas into water is much greater than that of N_2 (at 293 K and atmospheric pressure, the solubilities are 0.88 and 0.016 [cm^3-gas at 273 K and 0.1 MPa]/[cm^3-water at 293 K], respectively for CO_2 and N_2) and the Henry constant for CO_2 at 293 K is less than one-fiftieth of that for N_2. This means that the amount of CO_2 dissolved in water is much higher than N_2, which in turn causes incteasing generation of CO_2 foams after the depressurization. In addition, CO_2 is absorbed by coal much more easily than N_2. This gives rise to increased froth formation by CO_2 as compared to N_2. Thus, in the case of N_2, only the entrainment of particles by the rising foam in water contributes to the flotation which increases slightly with pressure whereas in the case of CO_2 the flotation is promoted not only by the entrainment but also by the froth formation.

3. 2 Elutriation from Liquid Fluidized Bed

Since the amount of low ash-content particles recovered by the flotation was much less than expected, a further classification of the CO_2 treated particles was tested by applying liquid fluidization on the basis of a simulation which is briefly described below.

Elutriation of particles from a liquid fluidized bed occurs when the liquid velocity exceeds their terminal velocity. When a bed consisting of size- and/or density-different multicomponent particles is fluidized at a velocity, the bed stratifies into layers forming a layer of particles with the smallest terminal velocity among those of the whole constituents on the top[4),5)] and the particles elutriated are those having terminal velocities lower than the fluidizing liquid velocity. Hence, a successive small increase of the velocity makes it possible to separate the particles based on their terminal velocity differences. On the other hand, the froth formation by absorbed gas on coal particles brings about a change in the distribution of the apparent particle densities. If CO_2 absorbs preferentially onto the carbonaceous portion of a coal particle, the lower ash-content particles can be expected to be concentrated through a greater decrease in the terminal velocity. This expectation was first examined by a simple model simulation with the following assumptions:

(1) The size distribution of coal particles is log-normal as expressed by Eq.(1) in Table 2.

(2) Particles of the same size consist of two components (Eq.(2) in Table 2), one having the lower ash content whereas the other the higher, and the difference between the ash contents is wider for finer particles (Figure 6(c)). As an example, the ash distribution is assumed to be described by Eqs. (3) and (4) for the lower and higher ash-content components, respectively.

(3) CO_2 is absorbed only onto the carbonaceous portion of particles and all absorbed CO_2 contributes to the froth formation after the depressurization.

(4) The fluidizing water flow is uniform in the bed and is laminar around a particle so that the terminal velocity is given from the Stokes law as expressed by Eq.(5) in the table.

Figure 5 shows distributions of the particle size and ash content calculated when values for the properties relevant to the above correlations are assumed as shown in Table 3. In this case the ash content of the lower ash-content component was assumed to increase in proportion to the particle size from the minimum of 0.005 to the average of 0.025 while that of the higher ash-content component to decrease exponentially from the maximum of 1.0 to the average. The calculated cumulative weight fractions of elutriated particles and their ash contents of this coal vary with the superficial liquid velocity as shown in Figure 6 for both untreated and treated particles. It is seen that the cumulative weight fraction of treated particles is always greater than that of the untreated, revealing an earlier elutriation of the former due to a decrease in apparent particle density by CO_2 froth formation. Since the decrease is greater for the lower ash-content particles, the terminal velocities of some lower ash-content coarser particles are smaller than those of the higher ash-content finer ones which were smaller than the formers before the treatment. Accordingly, with a certain amount of particles elutriated the cumulative ash weight fraction for treated particles becomes in general small in comparison with that for the untreated. For the example in the figure where the mean ash content is 10.5 wt%, a rapid increase in the ash reduction starts

Table 2 Correlations used for simulation

Size distribution of coal particles : f(d)

$$f(d)=\frac{1}{\sqrt{2\pi}\sigma d}exp\ [-\frac{\{ln(d/d_{av})\}^2}{2\sigma^2}] \tag{1}$$

Ash content of a particle : a(d)

$$a(d)=m_1a_1(d)+m_2a_2(d) \tag{2}$$

where

$$m_1+m_2=1$$

$$a_1(d)=a_{av}-(a_{av}-a_{min})\frac{d_{max}-d}{d_{max}-d_{min}} \tag{3}$$

$$a_2(d)=a_{av}\ exp\ [ln(a_{max}/a_{av})\frac{d_{max}-d}{d_{max}-d_{min}}] \tag{4}$$

Terminal velocity of a particle : u_{ti}

$$u_{ti}=\frac{g}{18\mu}\{\rho_i(V_i/V)-\rho\}(6V/\pi)^{2/3} \qquad i=1,\ 2 \tag{5}$$

where

$$\rho_i=1/\{a_i/\rho_a+(1-a_i)/\rho_c\}$$

$$V=V_i\{1+V_G(1-a_i)\rho_i\}$$

Symbols:

$f(d)$; probability density function, σ ; standard deviation, d ; particle diameter, d_{av}, d_{min} and d_{max} ; mean, minimum, and maximum particle diameter, respectively, $a(d)$; weight fraction of ash in particles with diameter d, $a_1(d)$ *and* $a_2(d)$; lower and higher weight fractions of ash in particles with diameter d, respectively, m_1 and m_2 ; weight fractions of lower and higher ash content particles, respectively, g ; gravitational acceleration, μ ; fluid viscosity, ρ, ρ_a, ρ_c, and ρ_i ; densities of fluid, ash, d.a.f. coal, and particle, respectively, V_i ; particle volume, V_G ; volume of absorbed gas per weight of d.a.f. coal

when about 70 wt% of treated particles are elutriated and at 80 wt% elutriation it reaches as high as about 5.8 wt%. Then, it decreases gradually towards complete elutriation. Figure 7 illustrates typical experimental results of elutriation curves when the fluidized bed classification is applied to A274 coal particles untreated and treated by 0.5 MPa-CO_2 gas for 60 min. The shift of elutriation curve of the treated to the left clearly demonstrates a practical reduction of the terminal velocities caused by the CO_2 treatment. Furthermore, as expected from the above simulation, the ash content of the treated particles is lower than of the untreated in a range of particle recovery of more than 60 wt%.

In Figure 8 the effect of the treatment on the degree of de-ashing is evaluated for different kinds of coals in terms of the weight fraction of ash separated, which is defined by

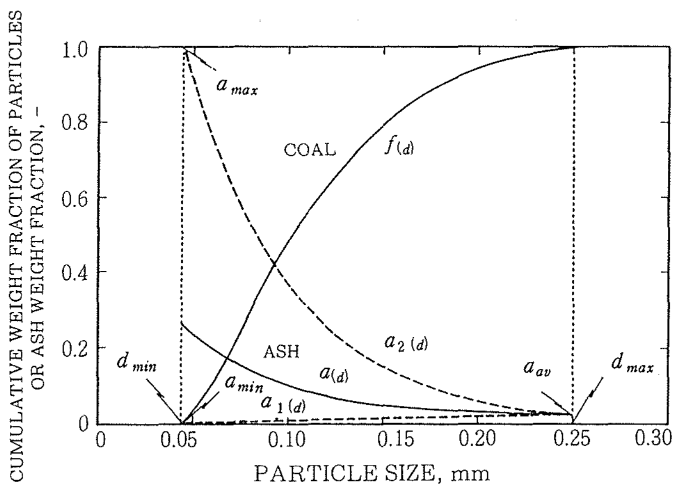


Fig. 5 Assumed distributions of coal particle size and of ash contents (See Tables 2 and 3).

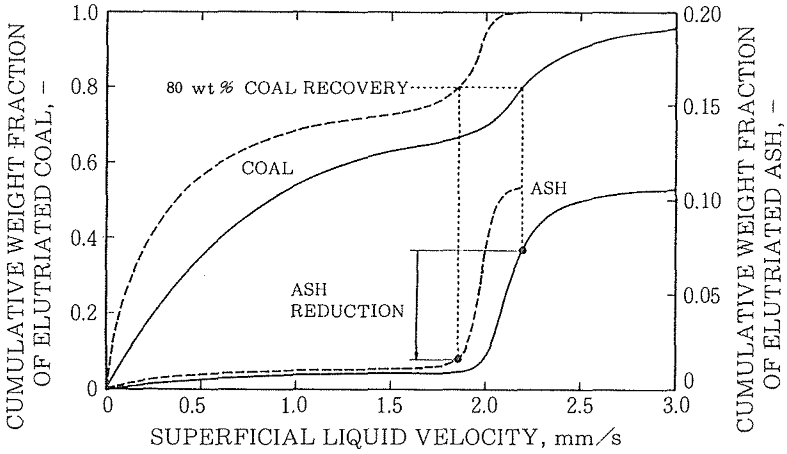


Fig. 6 Calculated result for elutriation of coal particles and ash from fluidized bed.

$$1-\frac{[\text{cumulative weight fraction of elutriated ash}]}{[\text{mean ash content}]}$$

against the cumulative weight fraction of the elutriated coal particles. As can be seen, for all cases the fluidization itself has an effect on de-ashing. Therefore, the net de-ashing efficiency of the CO_2 treatment is evaluated here as the difference between the weight fractions of separated ash of the treated and untreated coals at the same fractions of the coal particles elutriated. Among the coals employed, the treatment is most effective for A274 coal (Figure 8(a)), giving about 10 wt% increase in the fraction of separated ash at fractions of elutriated particles between 0.7 and 0.9. The increase seems to be almost independent of

Table 3 An example of assumed particle and water properties for simulation (Simbols are shown in Table 2)

Coal particles ;	ρ_c	$=1.1$	$\mathrm{g\ cm^{-3}}$
	d_{av}	$=0.194$	mm
	d_{min}	$=0.044$	mm
	d_{max}	$=0.254$	mm
		$=0.50$	
Ash ;	ρ_a	$=3.0$	$\mathrm{g\ cm^{-3}}$
	a_{av}	$=0.025$	
	a_{min}	$=0.005$	
	a_{max}	$=1.000$	
	m_t	$=0.75$	
water ;	ρ	$=1.0$	$\mathrm{g\ cm^{-3}}$
	μ	$=0.010$	$\mathrm{g\ cm^{-1s-1}}$
CO_2 absorption ;	v_c	$=0.05$	$\mathrm{cm^3-CO_2/g-d.a.f.\ coal}$

CO_2 pressures from 0.5 to 2.0 MPa. Such a de-ashing effect is also seen for W200 and W250 coals treated by CO_2 at 0.5 MPa (Figure 8(b)) although the improvement is rather smaller, particularly very slight for the latter. On the other hand, for A149 and M250 coals, the effect can not be detected (Figure 8(c)). Though the results are not shown here, the effects for T399 and T112 coals are similar to those for W200 and A149, respectively.

In the present experiments no further attempt was so far made to have more improved results. Future work is therefore needed in particular on coal comminution to produce the size and ash-content distributions of a coal suitable to the CO_2 treatment.

4. Conclusions

A simple method to classify coal particles depending on their ash contents was described, which utilized the characteristics of coal to absorb a fair amount of CO_2.

1. When a mixture of coal particles and water was exposed to CO_2 gas under pressure and then released to atmospheric pressure, froth formation was observed on the surface of the individual particles having the lower ash contents. Together with foam formation in water, this resulted in flotation of the low ash-content particles onto the top surface of water. However, the amount of the floating coal particles was less than a few percent for all coals presently employed.
2. Liquid fluidization was applied to further classification of the CO_2 treated particles, the apparent density distribution of which was broadened by the selective froth formation. For all coals, the treated particles were elutriated at lower liquid velocities than the untreated

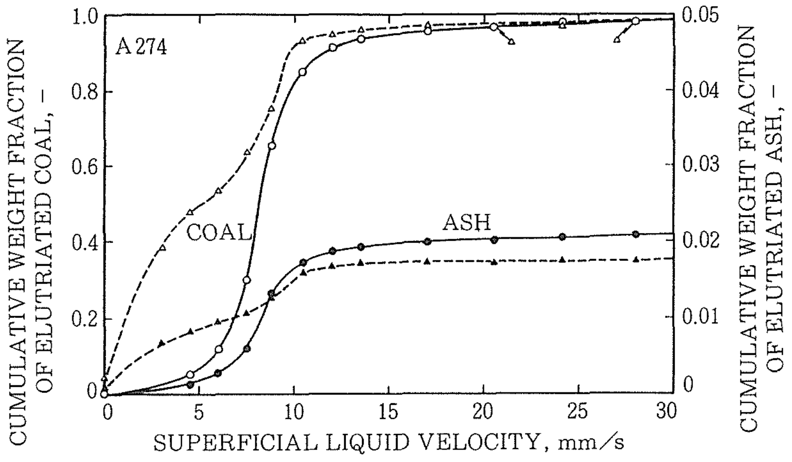


Fig. 7 A typical change of cumulative weight fractions of elutriated A274 coal particles and their ash contents with liquid velocity.

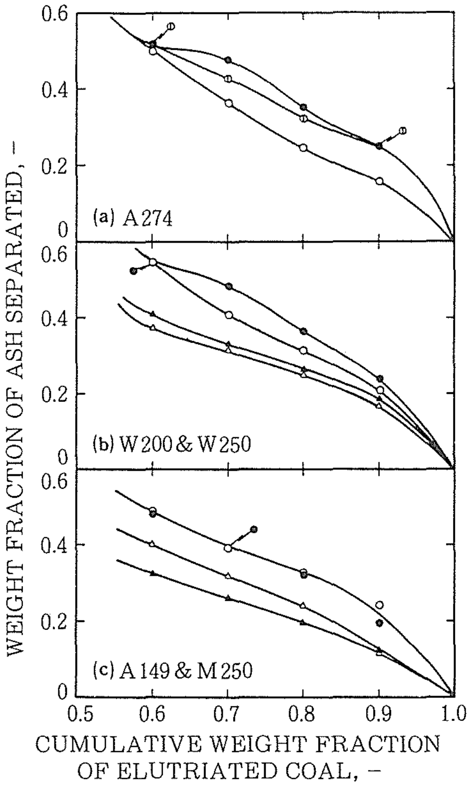


Fig. 8 Degree of deashing as a function of cumulative weight fraction of elutriated coal paticles.

particles due to a decrease in the terminal velocities. Based on the net efficiency of the treatment evaluated from difference between elutriated ash weight fractions, it was shown that the treatment is effective for some coals.

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