Dry Superconducting Magnetic Cleaning of Pulverized Coal

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There are wet and dry methods of cleaning pulverized coal for thermal power stations. However, it may be desirable to use a dry process because dewatering finely pulverized coal is difficult and expensive, and burning wet coal reduces the thermal efficiency of the combustion process. The largest advantage is that dry cleaning methods require the lowest initial capital investment and have the lowest maintenance costs of all currently used methods of upgrading fine coal. Also, influencing the choice of dry cleaning beneficiation is the lack of sufficient water in arid regions, localities of high altitude or cold climates. It has been shown that high gradient magnetic filters can be constructed which will extract micron-sized paramagnetic particles from a pulverized coal-air stream passing through the filter. Conventional magnetic separators are generally restricted to separating strongly magnetic materials, such as iron and magnetite. High gradient magnetic filters are capable of separating weakly paramagnetic particles, the magnetic force being proportional to the product of field strength and gradient. Superconducting magnets can produce extremely intense and uniform magnetic fields, of up to 20 Tesla or more, with gradients of 70 Tesla/m or more. A high gradient magnetic cleaning device consists of a matrix made of fine ferromagnetic fibers located in a high magnetic field. The fine wire of the matrix provides high magnetic gradient sites for the capture of paramagnetic particles which therefore can be separated from an incoming coal and impurities mixture. The presence of wire of high magnetic susceptibility in air of low susceptibility results in a highly nonuniform magnetic field with large magnetic gradients. The finer the wire, the smaller the distance over which the field changes and thus the higher the gradient. Only superconducting magnets are currently economic to generate sufficiently large background magnetic fields of, say, more than 2 Tesla, over the volumes required for the desulfurization of coal.

Deterioration in the quality of coals supplied to the power industry (particularly as regards calorific value and mineral element content) is causing serious problems in terms of meeting the increasing environmental demands being made on power stations. This situation means that ways must be found of improving and expanding the industrial cycle on coal-fired thermal power stations. The sulfur content of coals ranges from 0.2 to 10 wt. %, but most coals have less than 5%. Pulverized coal for U.K. power stations has an average value of 1.6%. The typical proportion of inorganic sulfur to organic sulfur is approximately 2:1. The inorganic sulfur occurs mainly as two forms of FeS_2, namely pyrite and marcasite, which mostly exist as small particles widely imbedded in the coal substance. The organic sulfur is chemically bound to the hydrocarbon matrix of the coal. Beside sulfides and sulfates, other minerals occurring in coal are classified into clays, carbonates, oxides, chlorides and trace elements. During coal combustion in power stations, these minerals are converted into ash which leads to a number of boiler operating problems (slagging, fouling, and corrosion), and at the same time emit gaseous pollutants harmful to the surrounding environment. These mineral impurities are amenable to high gradient magnetic cleaning processes, if they are well liberated from the coal particles. The effectiveness of the process lies in the fact that coal is diamagnetic whereas most of minerals, including pyritic sulfur are paramagnetic. In principle, therefore, the magnetic impurities can be removed from the nonmagnetic coal. In practice, this means that the impurities must be liberated from the coal by grinding, as is normal in the combustion of pulverized coal. In some early studies (Trindade and Kolm, 1973; Burnley and Fells, 1985), the degree of grinding was probably inadequate to liberate the minerals so that much coal was lost with the magnetic mineral matter. Furthermore, even with fine grinding (to normal pulverized fuel specifications) care must be taken in laboratory studies to redisperse the particulates as a monodisperse aerosol, avoiding agglomerations of coal and liberated mineral matter. Thus led to disappointing results in these early studies.

The geological processes which formed coal also concentrated trace elements in coal. The average concentrations of arsenic, antimony, cadmium, mercury, and selenium are approximately ten times the average concentration found in all the other rocks that make up the earth's crust. Many trace elements in coal are associated with particular mineral matter species. Arsenic and mercury are commonly associated with pyrite, cadmium with sphalerite, selenium with lead selenide, pyrite and other sulfides (Finkelman, 1980). There are also some cases in which some of these elements are organically bound. The dry, magnetic desulfurization technique is

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effective in removing mineral matter from coal and can potentially clean at least some of the trace elements associated with specific minerals, thereby reducing the release of these elements into the atmosphere.

Distribution of Sulfur Content

The impurities occurring in coal may be classified broadly into those that form ash and those that contribute to sulfur emissions. From the standpoint of coal cleaning, both the ash-forming and the sulfur-containing impurities may be subdivided into two classes—impurities that are structurally a part of coal and hence inseparably combined with it, and segregated and/or disseminated impurities that can be eliminated to a greater or lesser extent by the dry magnetic cleaning method. Iron pyrites are present in all coal, occurring either diffused through the mass, in visible laminae, or in layers and nodules, sometimes of considerable size. It may exist in the state of sulfuric acid in combination with a base, in combination with iron as iron pyrites or bisulfide of iron, and, probably, also in combination with the organic elements of coal as it exists in albumen. The mode of distribution of minerals in coals significantly effects the cleaning efficiency of the magnetic benefication of coal. The problem of reducing the sulfur content of coal becomes one of reducing the pyritic sulfur content, and the degree to which it can be accomplished will be influenced strongly by the mode of distribution of the pyritic sulfur in the coal such as in concentration and in particle shape and size.

In high gradient magnetic cleaning, it is the degree of fineness of size and shape of the impurities that is of importance. Comprehensive photomicrographs have been produced with reflected light, and the photomicrographs illustrate the distribution of minerals in a British coal of Spencroft seam, open cast site, Stoke-on-Trent. The photographs displayed in Figures 1, 2 and 3 have been selected essentially to show the possible ways in which pyrites, marcasite, and ashes are distributed through the coal, and particularly the small sizes in which it is disseminated.

Figure 1a, for example, shows isolated marcasite crystalline layer less than 4 mm wide and more than 20 mm long which could be easily cleaned by froth flotation and magnetic separation. In Figure 1b the distribution can be seen of discrete pyrite grains in durain. These grains are all about 30 microns in diameter. This pyrite cannot easily be cleaned by other physical means, but seems to be easily removed by dry high gradient magnetic separation. It will be noted that some grains segregate both in number and in size. Figure 2a shows a fool's gold corn roughly 5 mm in size. This discrete pyrite corn is surrounded by ash cavity fillings and isolated ash layers. These minerals can be withdrawn by froth flotation, but not so effectively as by dry magnetic separation. Figure 2b shows primary discrete pyrite grain layer in fusain, some isolated pyrite grains and segregated groups, with pyrite grains again filling the fractures. Figure 3a shows a fusain layer in which the cavities and fractures are filled with ash and increase both in number and size as they coalesce into bands along the bedding plane. The full liberation of these ashes prior to the dry magnetic separation depends on the size which it is ground and the pulverizing method. The photomicrographs mentioned above were taken with fusain and clarain. Figure 3b displays a photomicrograph of a vitrain coal in Spencroft seam, open cast site, Stoke-on-Trent, for reference to them. It is very difficult to find pyrite and other impurities, although some less fine pyrite and marcasite concretions found in vitrain has been reported (Gray et al., 1963).

A photomicrographical investigation was undertaken to obtain data on the occurrence of sulfur and ash forms in various size and gravity fractions. One objective of the work was to determine how effective the dry magnetic separation will be for this special coal. An additional objective of the investigation was to determine ways to bring about improvements of matrix loadability and reductions of coal entrapment in the equipment. It did indicate that adequate liberation of coal feed particles should be the key to the problem. Therefore, a dry fluidized-bed aerosol generator was employed to monodisperse pulverized coal particles in air streams prior to the magnetic separation, and a vertically upward air-particle flow separator will be of some significance to the problem since the settling velocities of pyrites and marcasite particles in air are much greater than coal powder for the same size. They will thus travel upward at a lower velocity than the airstream and coal, which thus improves their probability of capture.

Theoretical

The fundamental unit in the dry magnetic cleaning process is a cylindrical ferromagnetic wire, located transversely into a strong applied magnetic field. The cleaning process consists of numerous extraction elements of the interaction between the ferromagnetic wire and small paramagnetic particle in a uniform background field, which includes the effects of the magnetic tractive force on the particle, air viscous drag force and buoyant force, and particle inertia and gravity.

Consider a ferromagnetic wire or radius a and saturation magnetization M_s placed transversely to an uniform magnetic field $\boldsymbol{H}$, large enough to saturate the wire. Spherical paramagnetic mineral particles of susceptibility χ, radius b, and ρ_p flow past the wire with the pulverized coal. The vertically upward airstream has a viscosity μ, a permeability μ_0 and density ρ_f, and flows with an uniform superficial velocity $\boldsymbol{U}_0$. The magnetic force $\boldsymbol{F}_m$ on the particle can be calculated by using

$$\boldsymbol{F}_m = \frac{2}{3}\pi\mu_0\chi b^3\nabla(\boldsymbol{H}\cdot\boldsymbol{H})$$

The viscous drag force $\boldsymbol{F}_d$ acting on a moving particle of velocity $\boldsymbol{U}_p$ is given by

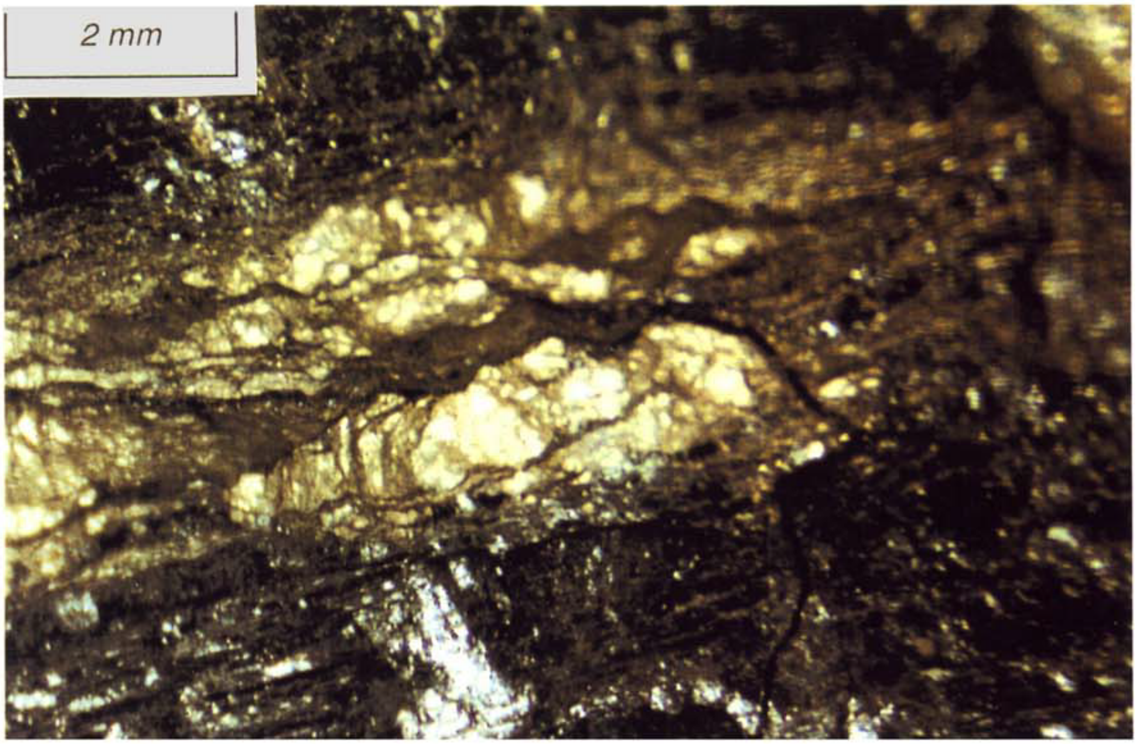
$$\boldsymbol{F}_d = \frac{1}{2}\pi C b^2\rho_f(\boldsymbol{U}_0 - \boldsymbol{U}_p)^2$$

Where C is air viscous drag coefficient. The sum of an air buoyant force and a gravitational force acting on the particle is

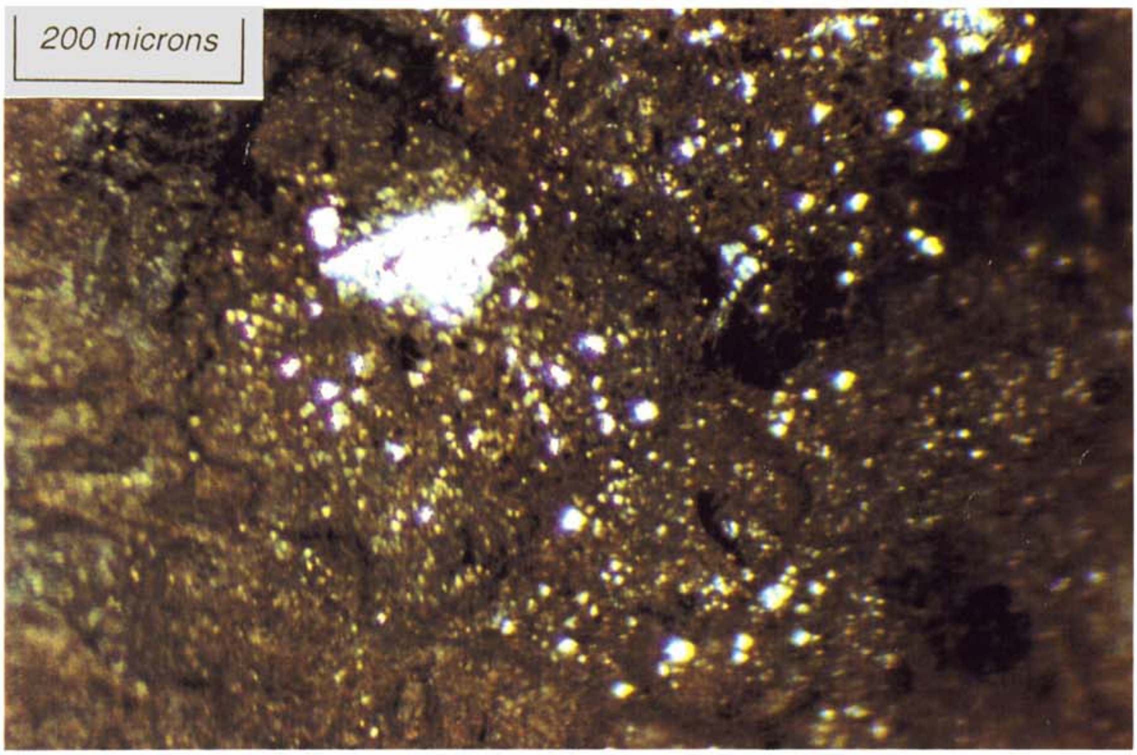
$$\boldsymbol{F}_g = \frac{4}{3}\pi b^3(\rho_p - \rho_f)\boldsymbol{g}$$

and the particle inertia can be expressed as

$$\boldsymbol{F}_i = \frac{4}{3}\pi b^3\rho_p\frac{d\boldsymbol{U}_p}{dt}$$

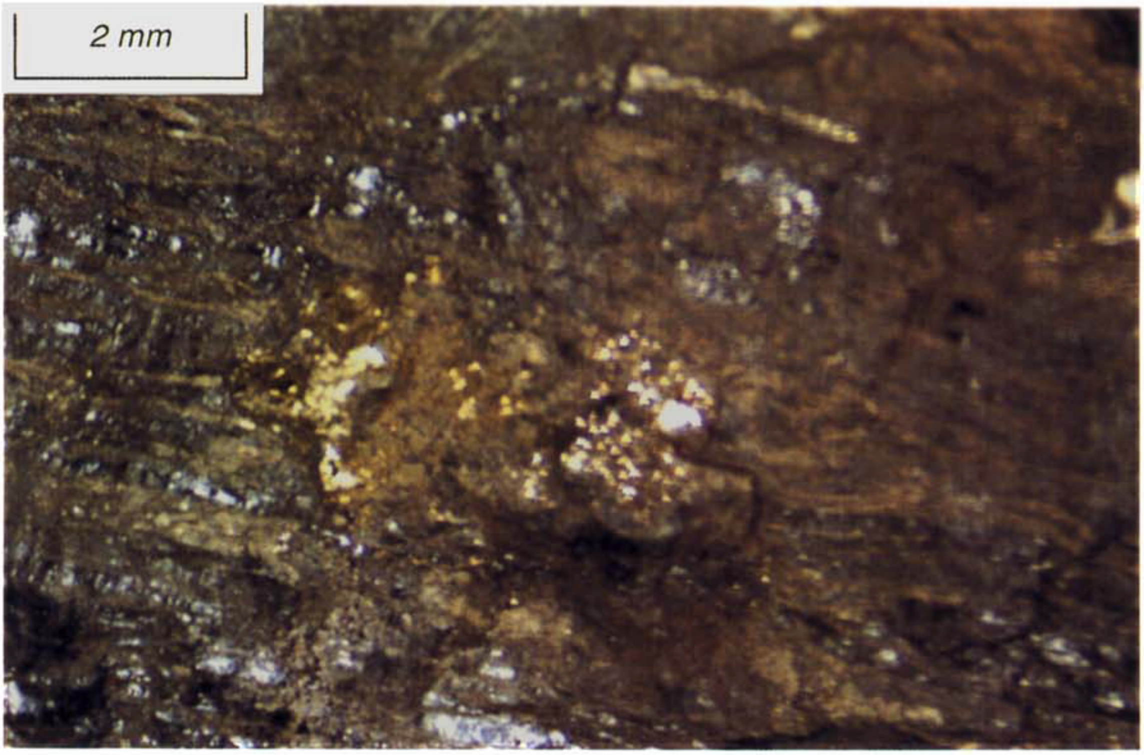


(a)

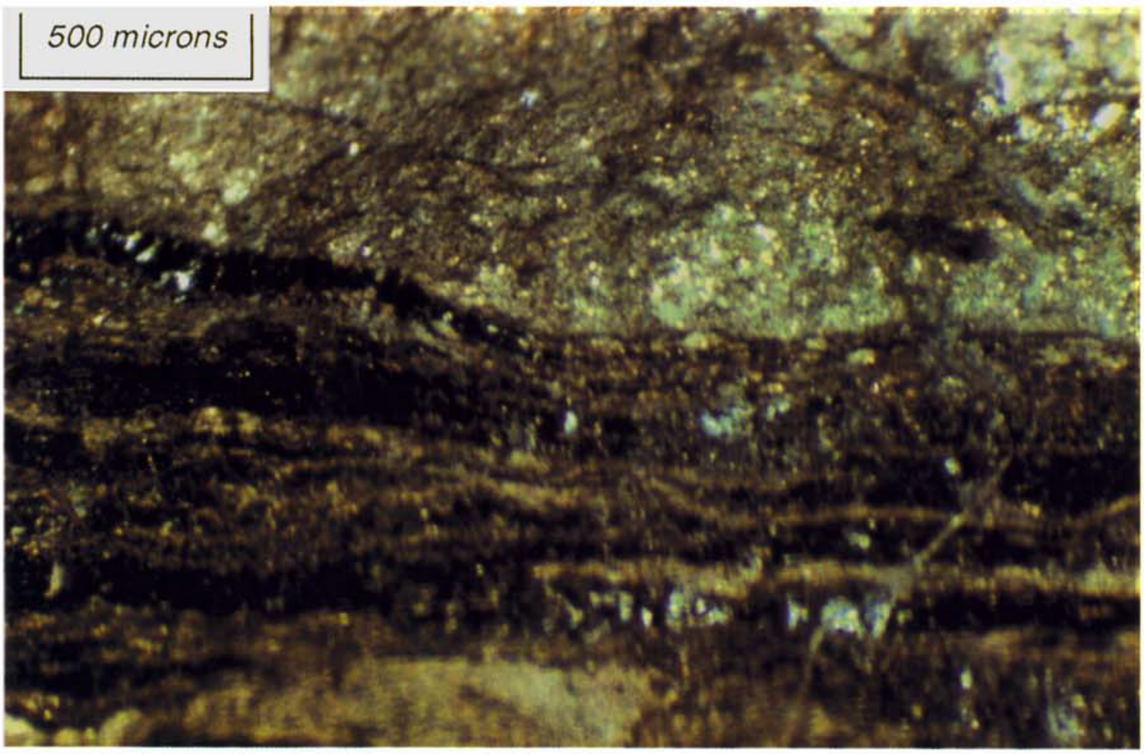


(b)

Figure 1. Microscopic pyrites and marcasite in British coal taken by reflected light.
(a) Discrete marcasite crystalline layer; (b) discrete pyrite grains.

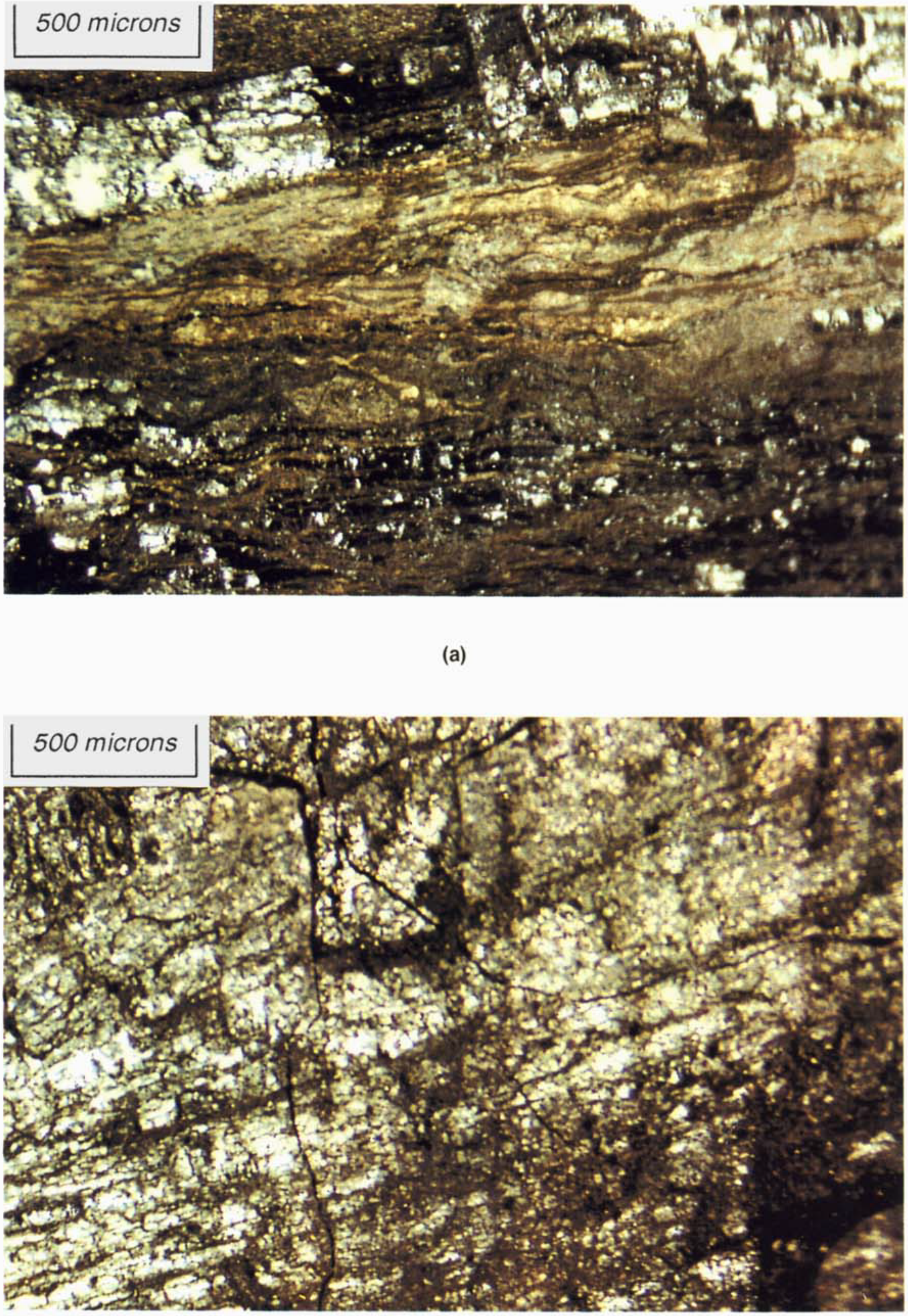


(a)



(b)

Figure 2. Microscopic pyrites in British coal taken by reflected light.
(a) Discrete pyrite corn; (b) pyrite cavity fillings and discrete pyrite grains.



(a)

(b)

Figure 3. Microscopic ashes and vitrain in British coal taken by reflected light.

(a) Ash cavity fillings and discrete ash layers; (b) British vitrain coal.

Every paramagnetic particle experiences always such a force balance as

$$\frac{4}{3}\pi b^3 \rho_p \frac{dU_p}{dt} = F_m + F_d + F_g$$

When contrasted with the vertically downward airstream, the net gravitational force is employed in capturing the mineral particles with the recently designed vertically upward test rig. Significant advantages will accrue from conducting experiments in a vertically upward airstream since larger capturing force will be of great significance in improving capture of impurities.

Experimental

The settling velocity in air of a 100 μm particle is 1 m/s for pyrite and 0.35 m/s for coal. In a vertically upward airstream of 1.5 m/s, the pyrite would travel at a velocity of 0.5 m/s, while the coal would be traveling at almost twice its settling velocity, over 1 m/s. Significant advantages were thus expected to accrue from conducting tests in a vertically upward airstream experimental rig given the resulting enhanced probability of capture of slower moving pyrite. The recently designed rig is shown in Figure 4. Pulverized coal particles were fluidized in a vertical vibrating fluidized-bed aerosol generator and injected into the Bell-mouth inlet of a vertical copper pipe (101.6 mm ID) where they mixed with upward

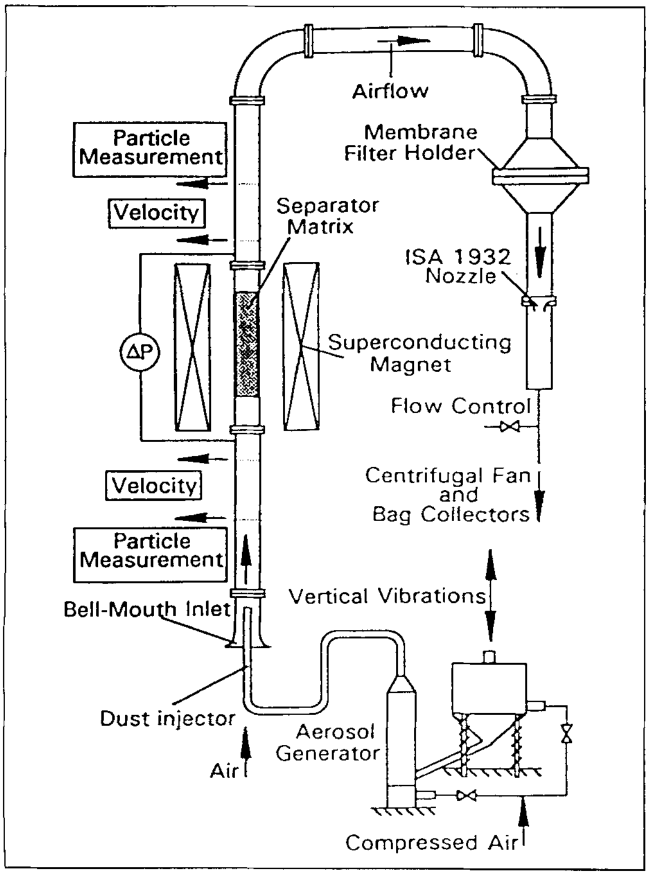


Figure 4. Dry magnetic desulfurization test rig for coal cleaning.

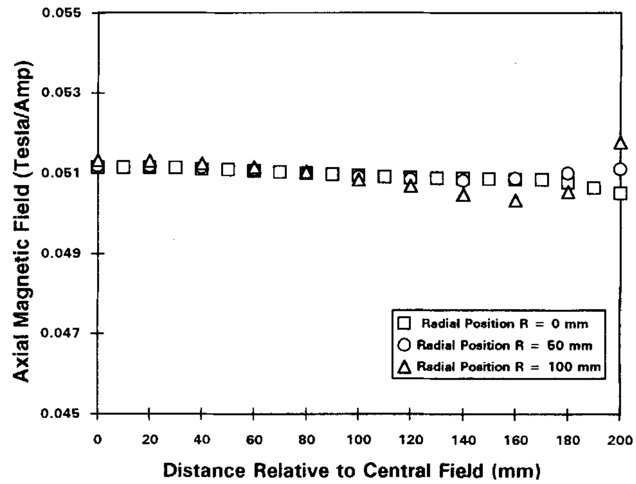


Figure 5. Distribution of axial magnetic field over axis in the 264 mm bore of superconducting magnet.

air flow drawn in by a centrifugal fan drawing room air/coal mixture through the superconducting magnet and separator matrix. The separator is located in the 264 mm bore of the superconducting magnet of a maximum rated magnetic field 4 Tesla. The tails cleaned were recovered in a Whatman glass microfiber GF/D filter with 2.7 μm retention, and the clean air then vented to the atmosphere. Pyrites and other paramagnetic minerals in coal were captured on the wires of separator matrix, forming the fraction called Mags.

The main part of the rig consists of a superconducting solenoid magnet with field homogeneous to within $\pm$ 2.0 % in 200 mm dia. $\times$ 400 mm long vol. Figure 5 shows the radial distribution of the axial component of measured magnetic field in vertical direction. Particle-size analysis of the feed coal and the Mags was obtained using a Malvern 2600 Particle Sizer. The feed coal was jaw crushed, milled, and air classified to 20–105 μm range with a mean particle size (by mass) 49.73 μm. The particle sizes were selected to closely simulate those specified by the Central Electricity Generating Board, UK, for its pulverized fuel coal which is 75% < 75 μm and 100% < 100 μm. Samples of feed coal and tails were analyzed for moisture, ash, total sulfur, pyritic sulfur, sulfate sulfur, organic sulfur, carbon and hydrogen contents, and calorific values. These were determined by British Standard methods, BS1016. Table 1 shows the chemical analysis result of a typical feed coal from Coalfield Farm North (Middle Lount).

Results and Discussion

Tests were carried out to investigate the effects of applied magnetic field, matrix loading, and coal flow flux on coal cleanability by isolating operating parameters individually.

Table 1. Analysis of Feed Coal (Coalfield Farm North) on a Dry Basis

Moisture	Ash	Total Sulfur	Pyritic Sulfur	Sulfate Sulfur	Organic Sulfur	Calorific Value
5.7%	9.16%	2.89%	1.79%	0.32%	0.78%	26,732 kJ/kg

Table 2. Parametric Effect of Applied Magnetic Field on Pulverized Coal Cleanability*

Magnetic field, Tesla	1.0	2.0	3.0	4.0
Mags capture, %	5.22	5.74	5.78	6.80
Coal recovery, %	94.78	94.26	94.22	93.20
Pressure drop, mbar	12.0	12.3	12.3	14.0
Pyritic sulfur reduction, %	31.84	45.25	60.34	69.27
Sulfate sulfur reduction, %	15.63	21.88	21.88	21.88
Total sulfur reduction, %	42.56	44.29	49.83	56.71
Ash reduction, %	13.97	28.49	20.85	33.19
Heating value recovery, %	96.90	98.63	97.76	96.96
Mean particle size of Mags, μm	46.48	41.84	53.71	49.97

*Feed coal: Coalfield Farm North, 50 g; coal flow flux: 8.24 $g/m^2 \cdot s$, superficial gas velocity: 1.50 m/s; matrix length: 250 mm; matrix material: 0.710 mm circular stainless wires.

The chemical analysis results on dry basis tests conducted are given in Tables 2, 3, and 4, which display that excellent coal desulfurization and good ash removal could be achieved by the dry, high gradient magnetic cleaning process for pulverized coal.

Coal recovery decreases with increasing applied magnetic fields and a correspondingly increasing Mags capture on the matrix shown in Table 2. Accumulation of Mags captured causes the pressure drop across the matrix separator to increase. As the applied magnetic fields increase, the tractive magnetic force is able to increasingly capture more of the weakly magnetic composite particles of organic coal and minerals due to incomplete liberation, giving rise to increase of organic coal fraction in Mags capture resulting in slightly lower coal recovery. The results in Table 2 show further that the heating value recoveries for all the four tests are slightly higher than the coal recovery. This is the result of the removal of very low calorific value inorganic impurities and the retention of very high calorific value organic carbon and hydrogen. The heating value recoveries do not reduce with strengthening the applied magnetic fields, although the coal recoveries do so while the Mags capture increases. With the increase in the Mags capture as the applied magnetic field is strengthened, there is correspondingly increasing pyritic sulfur reduction, sulfate sulfur reduction, total sulfur reduction, and ash reduction. This is to be expected, because increasing only the applied magnetic field gives rise to greater retention magnetic force on the paramagnetic mineral particles over the other fixed competing forces and hence improves impurities removal efficiency. Generally speaking, a positive change of the applied magnetic field creates several effects. Firstly, a similar increase of the magnetic field in the vicinity of the matrix wire is produced due to strengthening of both the applied magnetic field itself and the induced field resulting from an increase in the stainless wire magnetization (provided its saturation value has not yet been reached). Secondly, the magnetization of the paramagnetic mineral particles is raised linearly with the applied magnetic field.

Table 3. Parametric Effect of Matrix Loading on Pulverized Coal Cleanability*

Coal Feed g	Pres. Drop mbar	Pyritic Sulfur Reduction %	Sulfate Sulfur Reduction %	Total Sulfur Reduction %	Ash Reduction %
69.01	11.2	57.54	18.75	43.94	30.57
100.56	11.5	50.28	15.63	42.21	26.20
135.16	11.9	46.93	21.88	35.64	23.14
168.35	12.2	43.02	21.88	34.26	24.02
208.25	12.7	42.46	18.75	34.26	23.47
245.50	12.9	37.99	25.00	33.91	26.53
278.78	13.2	36.87	21.88	32.53	19.43
310.40	13.5	35.75	21.88	32.18	14.96

*Feed coal: Coalfied Farm North; coal flow flux: 8.24 $g/m^2 \cdot s$; superficial gas velocity: 1.50 m/s; magnetic field: 3.0 Telsa; matrix length: 250 mm; matrix material: 0.710 mm circular stainless wires.
Mags capture: 1.75%; coal recovery: 98.25%; heating value recovery: 98.56%; mean particle size of Mags: 42.89 μm.

Table 4. Parametric Effect of Coal Flow Flux on Pulverized Coal Cleanability

Coal flow rate, g/min	0.906	1.10	4.00	6.43	23.66	73.16
Coal flow flux, $kg/m^2 \cdot h$	6.73	8.17	29.66	47.66	175.5	542.5
Mags capture, %	4.28	3.78	4.37	5.28	9.13	16.55
Coal recovery, %	95.72	96.22	95.63	94.72	90.87	83.45
Pressure drop, mbar	11.8	13.4	11.6	12.6	11.4	12.7
Pyritic sulfur reduction, %	56.42	55.87	57.54	56.98	59.22	53.63
Sulfate sulfur reduction, %	21.88	18.75	18.75	21.88	18.75	21.88
Total sulfur reduction, %	41.87	34.60	37.02	48.79	45.67	44.29
Ash reduction, %	32.31	24.67	23.58	24.89	16.92	13.97
Heating value recovery, %	98.83	96.25	95.66	95.17	91.37	83.99
Mean particle size of Mags, μm	53.48	49.86	42.89	46.25	37.61	40.06

Feed coal : Coalfield Farm North, 50 g; magnetic field: 3.0 Tesla; superficial gas velocity: 1.50 m/s; matrix length: 250 mm; matrix material: 0.710 mm circular stainless wires.

Obviously, the percentages of total sulfur reduction, sulfate sulfur reduction, and ash reduction are less than that for pyritic sulfur. The reason that total sulfur reduction is lower than pyritic sulfur is that the dry magnetic separation beneficiation does not clean organic sulfur, which is chemically bonded to the coal structure. The reason that sulfate sulfur reduction and ash reduction are less than that of pyritic sulfur may be owing to their lower densities and/or magnetic susceptibilities. It is interesting to note that the mean particle size of the Mags captured increase with the applied magnetic fields. As the magnetic field increases, the magnetic force is able to increasingly capture more of weakly paramagnetic composite particles of organic coal and inorganic minerals and these particles are sometimes of larger particle size.

Matrix loadability was also investigated to determine its effect on pulverized coal cleanability in the vertically upward airstream test rig. Matrix loadability is a measure of the amount of the paramagnetic mineral impurity load the matrix can accumulate before a significant loss of its capturing efficiency. These tests were carried out under identical stable operating conditions except for varying feed coal loadings. These requirements lead to the weights of tails cleaned must be determined at intervals during the test run. Table 3 lists parametric effect of matrix loading on pulverized coal cleanability. As expected, the impurities reductions decrease for increasing feed. The pressure drop across the matrix separator increases linearly for increasing feed as the buildup of mineral particles on the matrix wires increases because the mass of the Mags captured increases with increasing feed. It too exhibits that all the mineral impurities reductions decreases with feed coal increase. It may suggest that the Mags particles are "bouncing-off" from the matrix with the accumulation of Mags captured. It too implies that the magnetic force exerted by the matrix wire on a suspended paramagnetic composite particle, which is to be seized and retained, decreases with buildup of captured Mags, that is, the matrix wire becomes less effective. With reference to the previous results published by Lua and Boucher (1990), the suspension flow alternation from a vertically downwards direction to a vertically upward airstream appears to have increased matrix loadability, permitting longer cycle times and/or heavier throughputs, while basically maintaining sulfur and ash reductions. The result may be retention on matrix wires increases and the probability of "bouncing-off" decreases in a vertically upward airstream due to the lower pyrite particle velocity.

Finally, the low percentage of Mags capture arise from the high coal recovery figure, that is, less diamagnetic coal has been trapped to appear as Mags.

The quantities of pulverized coal treated giving good demineralization are very important to plant scale-up. The matrix loadability tests have been discussed above, and another important operational condition which is of economic significance is coal flow rate or coal flow flux. High coal flow flux is crucial to practical application to reduce cleaning cycle times, of course. All the experimental results are presented in Table 4 for the parametric effect of coal flow flux on pulverized coal cleanability for a constant total feed, 50 g. The pressure drops across the matrix separator remained almost unchanged. The paramagnetic impurities can be removed from the nonmagnetic coal in dry magnetic separator. It should be fully appreciated that the impurities must be liberated from the coal. As coal flow rate/flux increases, more and more coal particles may agglomerate together, and the fluidized-bed aerosol generator is unable to produce good monodispersed pulverized coal aerosols at very high turndown ratio. As expected, the weight of the Mags captured increases with coal flow flux and coal recovery decreases with coal flow flux. The relevant heating value recovery, therefore, reduces as coal flow flux rises. It is interesting to note that there is no significant deterioration in the efficiency of desulfurization, however, ash removal reduces as shown in Table 4. This ash reduction deteriorating phenomenon may be due to the small retention force on ash producing minerals and also to matrix loadability, a large amount of Mags captured on the wires.

Conclusion

These experimental tests indicated that the dry magnetic desulfurization process was able to remove up to 69% of the pyritic sulfur and 33% of the ash content from a typical pulverized coal in a vertically upward airstream rig, and the heating value recovery is 97%. Increasing magnetic fields increases the percentage of reductions of pyritic sulfur, total sulfur, and ash. The penalty for the improved sulfur removals was the reduced heating value recoveries. The effectiveness of dry magnetic desulfurization depends on the matrix loadability. As the Mags captured accumulate on the wires of the matrix, its capture efficiency proportionally deteriorated. The matrix loadability in a vertically upward airstream appeared to be greater than in the vertically downward flow rig. Very high coal flow flux depressed slightly the magnetic cleaning efficiency, but this may be due to limitations of the aerosol generator at high flow rates.

Separator Scale-Up Plant

The coal feed treated with good cleanability was in the 50-100 g range at low velocities (≤ 1.5 m/s). It is important to recognize what rate of coal throughout can be achieved on a scaled-up plant to assess economic feasibility and technical practicality.

The pipe bore in these experiments was 101.6 mm and the separator length was 250 mm. Large superconducting magnets running in persistent mode that have recently been built have bore sizes up to 3 m (and greater) with similar fields as that used for this present investigation. The volume scale-up for this bore is 3×10^3. In addition, for tight experimental control, less than half the length of the magnet was used. Using the total magnet length, a further volume increase by 2.5 is possible for the typical magnet length : diameter ratios. This total volume scale-up of 7.5×10^3 means that the 50–100 g test quantities of coal feed scale to 375–750 kg plant coal feeds between cleaning operations. Some comments are no doubt desirable on the practicality of both superconducting magnets and a reciprocating matrix changing system in practice. Concerning matrix changing, a delightfully simple system has been devised and tested in which the matrix is moved in or out of the field in a few seconds by large forces generated by passing a pulse of microcurrent through the matrix in the high magnetic field. Cycle times between 1 and 3 min might therefore be considered acceptable giving upper and lower estimates of system capacity of 45–75 tons per hour. Such throughputs at low air velocity imply high coal : air ratios. Although no tests with such high ratios have been conducted, magnetic or aerodynamic interference between particles will become significant only when the mean particle separation is of the order of a few particle diameters. This implies that coal : air ratio will not be a limitation. Table 4 shows that very high coal flux slightly depressed the magnetic cleaning efficiency, which is believed due to limitations of the aerosol generator at such high flow rates.

Although the laboratory scale superconducting magnet requires liquid helium top-up every few days, modern large-bore magnets with superinsulation are routinely used in the china clay industry and in hospitals and require top-up at six monthly intervals or more. Secondly, with the approach of high-temperature superconductors much simpler and cheaper liquid-nitrogen cooled magnets can be anticipated.

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