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What Are We Trying to Separate?

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ABSTRACT

Heterogeneity both invites and permits separation of components. Coal is heterogeneous at a number of levels. At the simplest level it is a mixture of organic and inorganic phases. The textures and therefore the liberation characteristics of the inorganic components are a response to the sedimentary and metamorphic processes associated with coal formation. Mineral matter occurrence ranges from coarse interbeds and veins to micron sized inclusions and can be selectively associated with specific organic entities. The organic matter in coal is itself heterogeneous at two major levels. At the simplest level, three major groups of macerals are developed (largely as a result of processes in the peat stage). Each group of macerals is both chemically and physically distinctive from the other group and yet is also gradational into the neighboring group. At the finest level of heterogeneity, each maceral is itself a complex mixture of organic compounds. The organic material also contains organically bound elements, such as sulfur and alkalis, which are generally difficult to separate, but can be highly significant in coal utilization. A further level of complexity is introduced by the variation in the chemical and physical properties of all of the macerals (and to a lesser extent the minerals) as a response to rank change.

Separation processes have to take into account the levels of heterogeneity, and to be effective on a specific coal must be appropriate to the variations in properties shown by that coal for each type of heterogeneity. Full characterization of coals at the research, exploration and pilot plant stages is essential if results are to applied to commercial scale processes.

INTRODUCTION

Coal lost its dominance as a fuel to crude-oil sourced hydrocarbons and natural gas because of the low levels of impurities, low cost of production and the ease of handling crude oil, and natural gas. Recent (1973+) large increases in the cost of crude oil have brought about a burst of interest in coal although this has not yet been fully reflected by increased production and use. Coal suffers a price penalty as compared with oil and natural gas since it is a solid, and therefore is less easily handled. Generally, coal also has a higher level of impurities. Price levels for coal tend to be strongly affected by local factors but over the period 1978-1981 the premium commanded by oil over coal in international trade has been approximately a factor of two or three on an energy equivalent basis. That factor forms a margin which is in part the cost of tolerating the extra costs of using coal but also represents a zone of potential clawback of at least some of the costs associated with coal preparation to enhance its properties.

It is probable that coal cleaning will increase in relative importance as coal is substituted for oil. Figure 1 shows how "washed coal" rose sharply and then plateaued as a percentage of the coal mined in N.S.W. over the nineteen years 1960-1979. During that period the total tonnage mined has increased by 274% with the increase going largely to the growing markets of export coking and steaming coal, and domestic steaming coal. The tonnage of coal washed rose steadily throughout the period as did the total amount and the percentage of refuse produced. Similar patterns of change

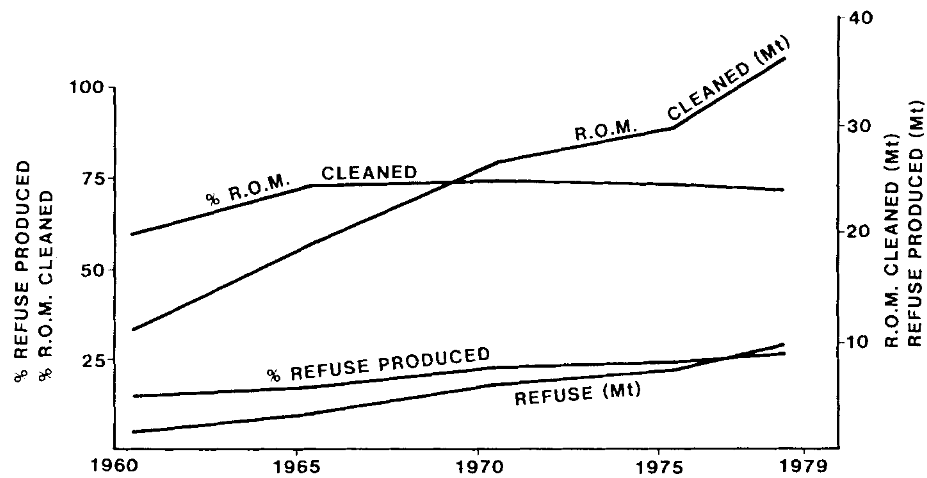

FIGURE 1. Changes in washed coal as a percentage of raw coal from N.S.W., 1975-79. (From data in Edwards, 1981).

seem likely to hold worldwide with the large tonnages of washery waste generating a need for greater use and safe disposal of these wastes.

The ashforming constituents together with most of the other undesirable components are not distributed homogeneously through the coal nor is the carbonaceous material itself homogeneous. This lack of homogeneity allows separation processes to be used to remove at least some impurities and, in some cases, selectivity to remove some carbonaceous material and improve the in-use behavior of coal beyond the changes effected simply by the removal of impurities. The nature of the inhomogeneity of coals varies greatly from one coalfield to another, and within one coalfield, from one seam to another. Some general features hold for all coals and the purpose of this paper is to provide a systematic basis for considering the "as mined" product in the context of coal separation processes.

COAL TYPE AND RANK

Coal is formed when plant-derived material, usually with some mineral matter, accumulates to form a peat and the peat is then covered and compacted by a cover of younger sedimentary rocks. The range of plant components preserved in the peat and the extent of alteration of these components in the peat determine coal type (White, 1909). The most common methods of expressing type are based on the recognition of three maceral groups (Table 1) following Stopes (1935). Maceral and mineral associations can be expressed in terms of microlithotype analyses (see Stach et al., 1975) and at the

TABLE 1. Maracel Groups*

Group	Origin	Summary of Properties
Exinite	Lipid wax or resin rich tissue	High hydrogen content, tough
Vitrinite	Humified wood or leaf tissue	High aromaticity, brittle if massive
Inertinite	Oxidized vitrinite precursors	High oxygen content. Fusinite is brittle, other varities relatively tough

*For a full list of maracels, see I.C.C.P. (1963, 1971, 1975) and Stach et al. (1975).

scale of visual observation in terms of the lithotypes described by Stopes in 1919. Measurements of bore core or mine sections normally employ brightness logging rather than lithotypes but the concept is similar. The scale relationships of various levels of type differentiation are illusrated schematically in Table 2.

The changes which occur in peat due to burial pressure and the elevated temperatures due to burial acting over time, are essentially metamorphic in character. The extent of these changes in the organic material is a measure of coal rank. Rank is best assessed using properties of the maceral vitrinite, such as reflectance or carbon content. For industrial practice, rank is commonly (and less precisely) assessed using whole-coal analyses. Major changes in both the physical and chemical properties of the macerals occur over the rull rank range.

Coal type and rank are independent variables. The properties of any given coal depend upon both its type and its rank. Thus macerals differ from each other in composition and physical properties at any one rank and each maceral's properties vary systematically with rank (Figure 2).

IMPURITIES IN COAL

Deleterious constituents in coal range in nature from strongly chemically bonded components such as some sulfur and nitrogen through to adventitious mineral matter. The commonly used term inherent mineral matter is rather misleading since it is generally defined in terms of (usually inefficient) separative processes rather than in any genetic sense. The majority of the ash-forming constituents in wood are almost certainly leached out during coalification and the majority non-carbonaceous material is almost certainly of secondary origin and is not therefore inherent. Three main groups of impurities can be distinguished.

TABLE 2. Levels of Heterogeneity in Relation to Coal Preparation

Heterogeneity		Separation method	Scale of thickness
Level	Kind		
Seam	Coal/non-coal	Basic mining process	0.2 m - 100 m +
Ply	Coal/dirt band	Selective mining Crushing and screening Washing	0.5 m - 5 m 0.1 m - 0.5 m 1 mm - 100 mm
Ply	Coal/coal	Selective mining Crushing and screening	0.1 m - 0.5 m 1 mm - 100 mm
Lithotype	Coal/coal or coal/mineral matter	Crushing and screening or cleaning	5 mm - 100 mm
Microlithotype	Various maceral and mineral assemblages	Fine crushing and cleaning	0.5 mm - 5 mm
Macerals and minerals	Maceral/ maceral/mineral	Ultra-fine crushing and cleaning	0.001 mm - 0.05 mm
Sub-microscopic	Within maceral inhomogeneity	Leaching, molecular reorganization	<500 nanometres

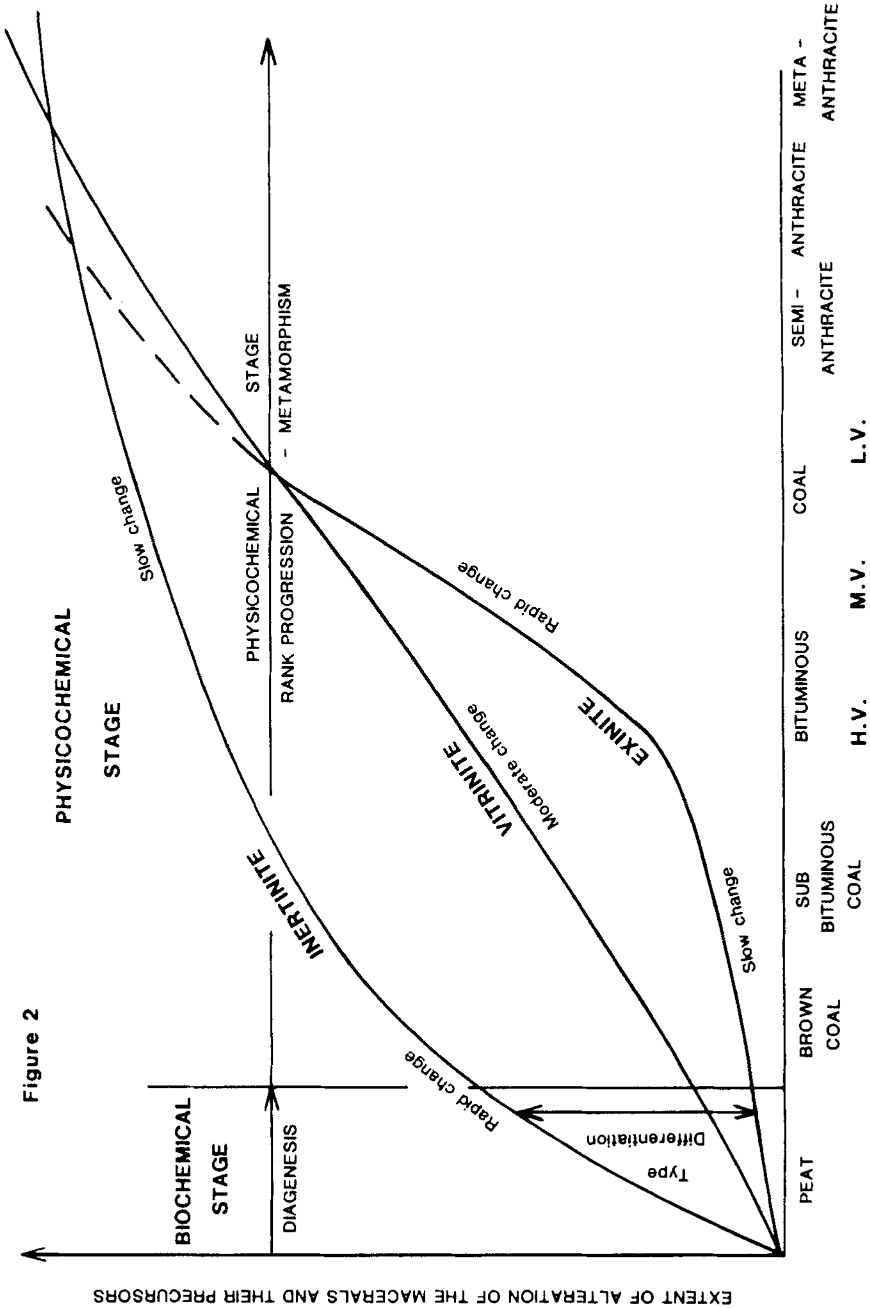

FIGURE 2. Type differentiation and rank change in the major maceral groups. Modified from Schopf, 1948, using data from Smith and Cook, 1980, with acknowledgements to A. J. Kantsler.

Strongly Chemically-bonded Elements

The most important of these are organically bound sulfur and nitrogen compounds which are present within the macerals. The nitrogen is dominantly of primary origin. Some of the sulfur is also primary in origin but the majority is clearly a result of reaction between the peat and sulfate-bearing vadose (mobile) water. Variations between macerals in the amounts of strongly bonded elements are relatively poorly known and understood. Removal of these elements can only be achieved if a major chemical restructuring of the coal (such as gasification or solvent extraction) is undertaken. G. C. Smith (pers. comm.) suggests that relatively little differentiation is present in brown coals so that the impurity levels of the macerals are similar. He suggests that mobilization and differentiation probably takes place before the hard brown coal stage with formation of some of the minerals which are normally considered to be syngenetic (formed at the same time as the peat).

Adsorbed and Weakly Bonded Groups

Some coals especially those of low rank contain ash-forming components in pore water, adsorbed on the surface of the coal or weakly bonded ions. In rare cases, such as the Morwell seam in Victoria, salts in solution form the majority of the ash-forming constituents with the SiO_2 percentage in the ash being typically less than 5% (total ash yield about 4% on a dry basis). As with chemically bonded impurities, the opportunities for removal of the solution, adsorbed and weakly bonded elements are limited.

<u>Mineral Matter</u>

Mineral matter has three chief modes of origin:

1. <u>Epiclastic minerals (minerals derived from outside the peat swamp)</u>

These consist of chiefly clay and quartz washed or blown into the peat swamp during its formation. The minerals range in grain size from clay size (in some cases as small as 0.001 mm) to cobble size (up to 0.1 m) but the majority lie within the silt size range. The minerals may be dispersed through or between macerals or be concentrate into beds (dirt bands).

2. <u>Syngenetic and pene-syngenetic minerals</u>

Minerals form within the peat during and immediately after its deposition from solutions of inorganic components moving through the peat. Deposition occurs in response to changes in pH, Eh (redox potential) conditions of the system. Some of the most important minerals formed at this stage are pyrite and siderite but, in some coals, significant amounts of other minerals such as clay, chalcedonic quartz and apatite originate in this manner. Most syngenetic minerals are intimately intergrown with the coal macerals and in many cases partly replace the organic matter. For example, pyrite occurs as small specks, framboids and more rarely as massive nodules which are dominantly replacive after vitrinite (Figure 3) but may also occur within the lumens of inertinite or as pyrite petrifactions replacing the coal maceral (Figure 4). Only the massive occurrences (Figure 5) of these minterals (typically nodules) are amenable to normal separative processes. Indeed examination of

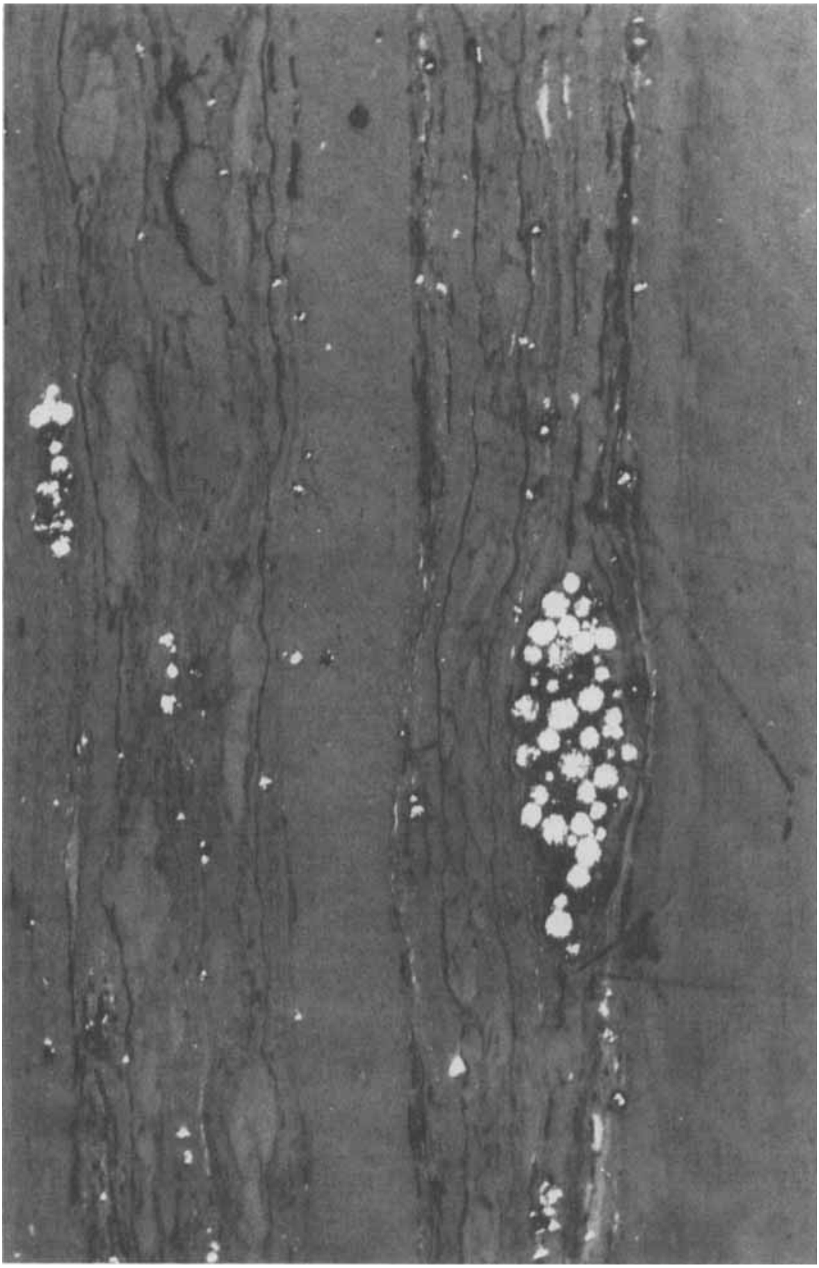

FIGURE 3. Framboidal pyrite associated with vitrinite in a Cherokee coal, Kansas, U.S.A. Reflected light, oil immersion, field width 0.7 mm.

FIGURE 4. Pyrite occurring within cell lumens in fusinite in the same coal as that shown in Figure 3. Reflected light, oil immersion, field width 0.5 mm.

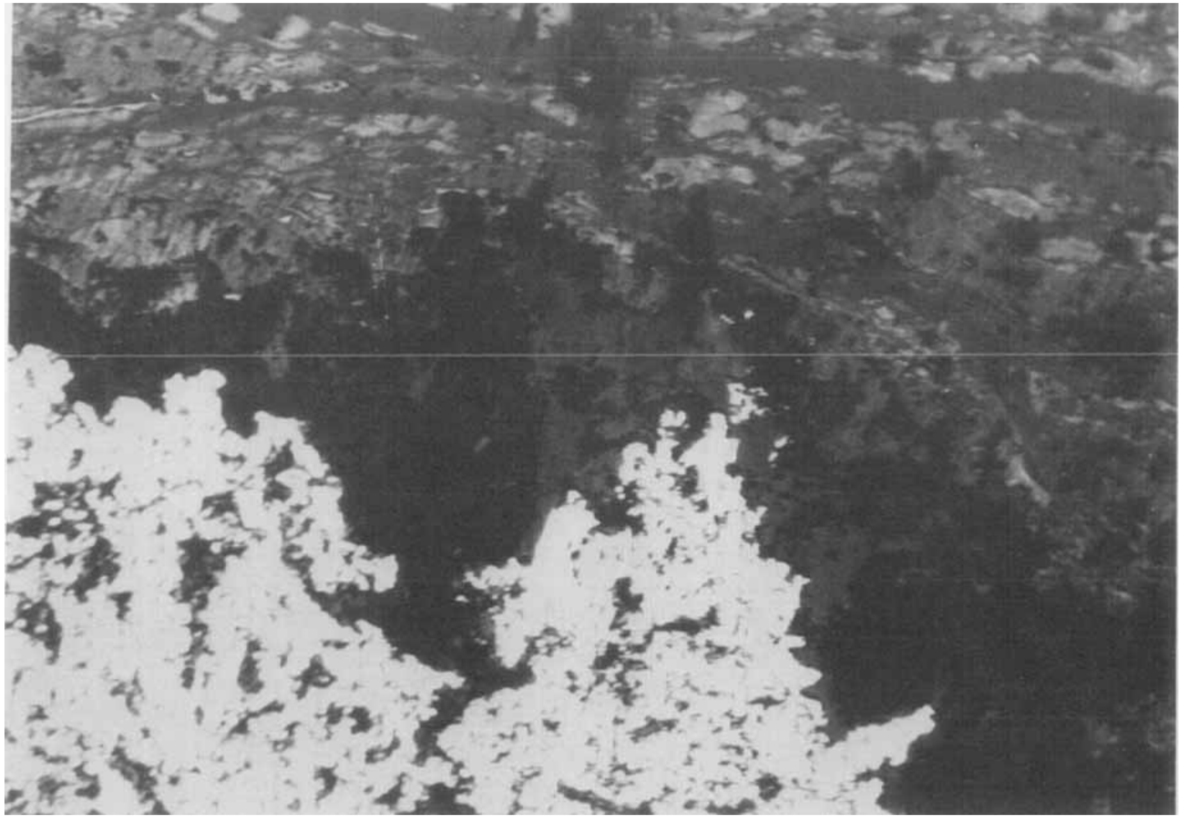

FIGURE 5. Pyrite replacing siderite nodule which is in turn replacing the coal microlithotype vitrinertite I. Tongarra Seam, Huntley Colliery, N.S.W. Reflected light, oil immersion, field width 0.5 mm.

residues from the laboratory determination of pyritic sulfur commonly shows the presence of at least some disseminated pyrite (this is of course then erroneously recorded as organic sulfur). Thus even on a laboratory scale and using reagents which cannot be used in plant practice, complete removal of a reactive phase such as pyrite is seldom possible. The fine specks of chalcedonic quartz shown in Figure 6 present a much greater cleaning problem. Syngenetic minerals are widely dispersed through most seams but tend to show significant systematic variations such that some separation can be attempted.

3. Epigenetic minerals

These minerals deposited within a coal after significant burial of the peat. Most of the mineralization is of vein type but the veins may be complex and in part replacive. The major epigenetic minerals are pyrite and calcite but a wide variety of rare minerals including sphalerite, millerite, douranite and laumositite has been reported. Epigenetic minerals are relatively amenable to separation. They are typically concentrated near the roof of the seam (less commonly the floor), and occur along cleats, thus being preferentially exposed during breakage of the coal. Mineralization tends to be concentrated laterally as well as vertically further aiding separation.

Physical methods of beneficiation are almost entirely restricted to the removal of mineral matter, with the efficiency being strongly dependent upon the mode of origin. Epiclastic and epigenetic minerals are, in general, the easiest to separate. Syngenetic minerals pose greater problems. The separation of any of the occurrences of mineral matter is dependent upon there being a lack of uniformity of

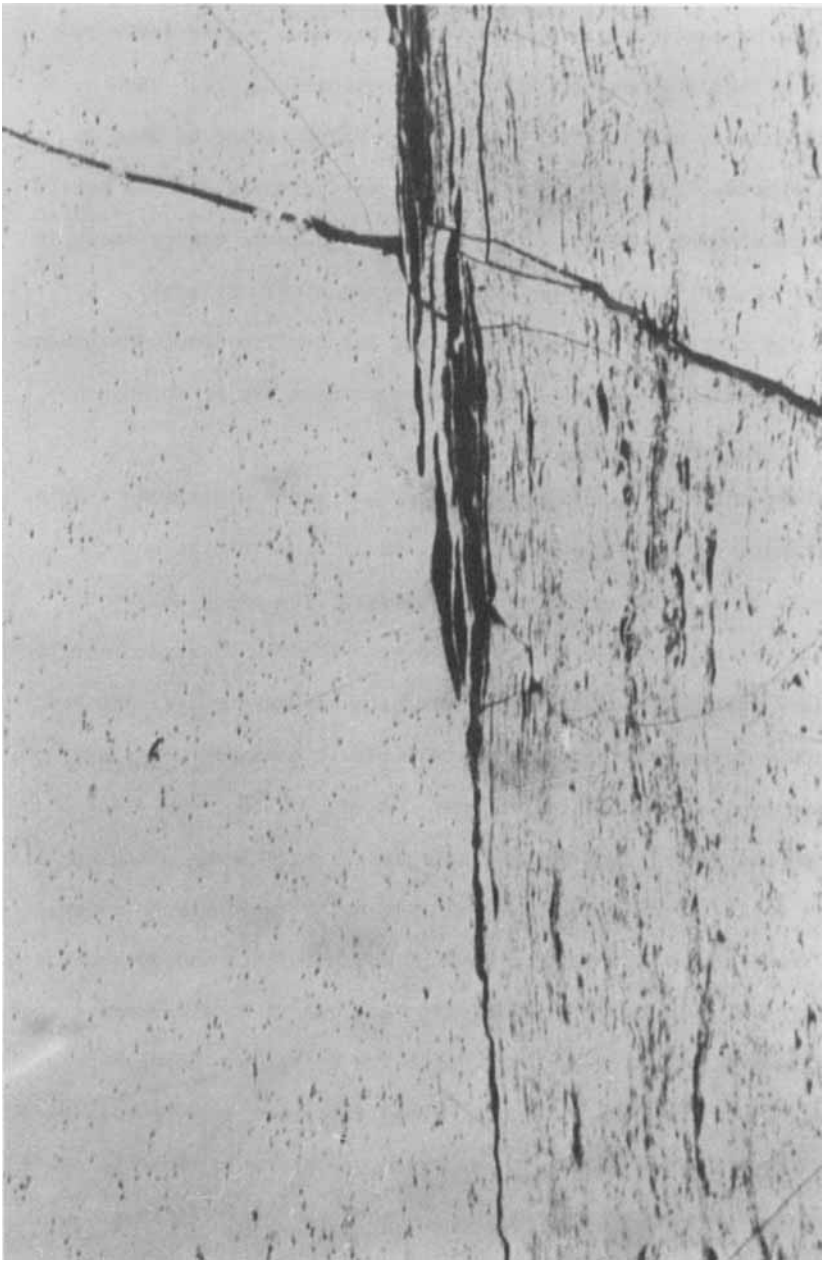

FIGURE 6. Clay and chalcedonic quartz disseminated in vitrinite. Tongarra Seam, Huntley Colliery, N.S.W. Reflected light, oil immersion, field width 0.5 mm.

distribution. The next section examines the levels of this heterogeneity and the ways in which advantage can be taken of the heterogeneity within coals. Attempts to go beyond the classical gravity, flotation and screening methods of separation are even more dependent upon this kind of knowledge.

LEVELS OF HETEROGENEITY

Some of the levels of heterogeneity are set out in Table 2 with an estimate of the scale relations of these levels expressed in terms of thickness. Thickness is used because the bedded nature of coals means that each unit has a lateral extent which is normally greatly in excess of its vertical extent. Heterogeneity persists down below the maceral level since each of the macerals represents a complex mixture of dominantly polymeric compounds. These most fundamental levels of heterogeneity are normally of significance in relation to processes such as carbonization, liquefaction or gasification but could become of greater importance in more general separation processes. Certainly it is essential to recognize that even the macerals are not uniform chemical compounds but are themselves complex inhomogeneous mixtures.

Coal and Non-coal Strata

Coals range from having sharp upper and lower contacts to having transitional contacts. Sharp contacts are desirable in order to minimize quality and quality control problems and, in the case of underground mining, a sharp roof contact generally eases the problem of contamination by roof rocks. Some contamination with adjacent strata will always occur due to unexpected geological phenomena such as floor rolls, washouts or clastic dykes, accidental mining of the floor or roof and roof falls. Figure 7 shows typical overburden strata at a mine in N.S.W. High wall stability is good and the immediate roof has a sharp boundary with the coal.

Plies

Plies within a seam may be separated by a dirt band or defined by a change in coal type. Smith and Cook (1976) have pointed out the general tendency for vitrinite content to decrease up section within coal seams and have observed that a similar tendency occurs with most although not all plies. Separation along ply boundaries is generally easy so that selective preparation is generally possible if the grindability of the various plies differs (Burstlein, 1954). Separation of dirt bands (Figure 8) is close to the lower size limit at which very limited amounts of coaly material are associated with the potential reject material. Below this level potential rejects generally comprise a mixture of free mineral matter and coal/mineral matter composites (middlings). Figure 9 shows the ply sequence in a relatively massive low vitrinite coal of Triassic age from Callide in Queensland.

Lithotype

At the lithotype level considerable variations in both petrographic type and mineral content are present (Figure 10). Grindability is also variable but unfortunately mineral matter has at least two main modes of occurrence; one with the relatively friable

FIGURE 7. Overburden strata at the Costain Opencut, Liddell, N.S.W. Part of the seam (approx. 3 m) is exposed at the base of the high wall (40 m in height).

vitrinite rich coal, and a second with the tougher inertinite rich coal. The balance between these modes is highly variable, and with some coals potential exists for improving coal cleaning operations at relatively low cost by examining carefully the characteristics of the various size fractions and designing the plant to match these characteristics. Figure 8 shows a number of lithotypes which contain varying amounts of a number of different minerals.

Microlithotype

Similar considerations apply at the microlithotype level, but most microlithotypes are only liberated in the finer sizes of a normal

FIGURE 8. Thick, composite dirt band in the Liddell Seam, N.S.W. Lithotype banding can be seen in the coal plies above and below the dirt band. The lens cap is 65 mm in diameter.

grind or if very fine grinding of the whole coal is undertaken. Figures 3 to 6 illustrate some associations of mineral matter with a range of microlithotypes.

Macerals

At the level of macerals, Figures 3, 4 and 6 indicate that locking of mineral matter within individual occurrences of macerals

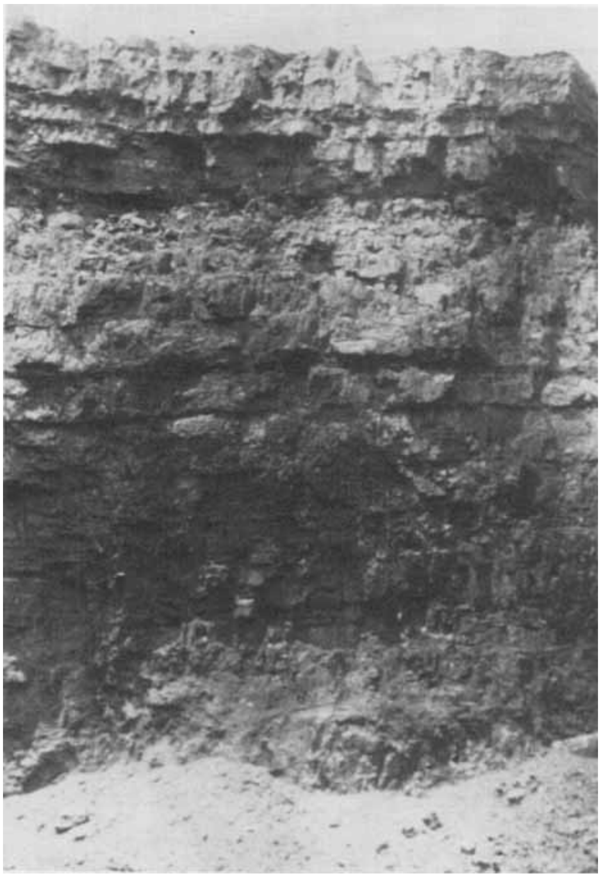

FIGURE 9. Ply sequence in the Main Seam, Callide, Qld. The section is 17 m in thickness and averages only 30% vitrinite on a mineral free basis. Few dirt bands are present and the level of compositional variation is low.

is a common phenomenon. Figure 11 shows an occurrence of calcite of epigenetic origin. Some intergrowth of the calcite with the vitrinite is evident.

Current commercial plant practice generally does not effect separation at either the microlithotype or the maceral level. Unless such a separation is made, significant amounts of coal will be

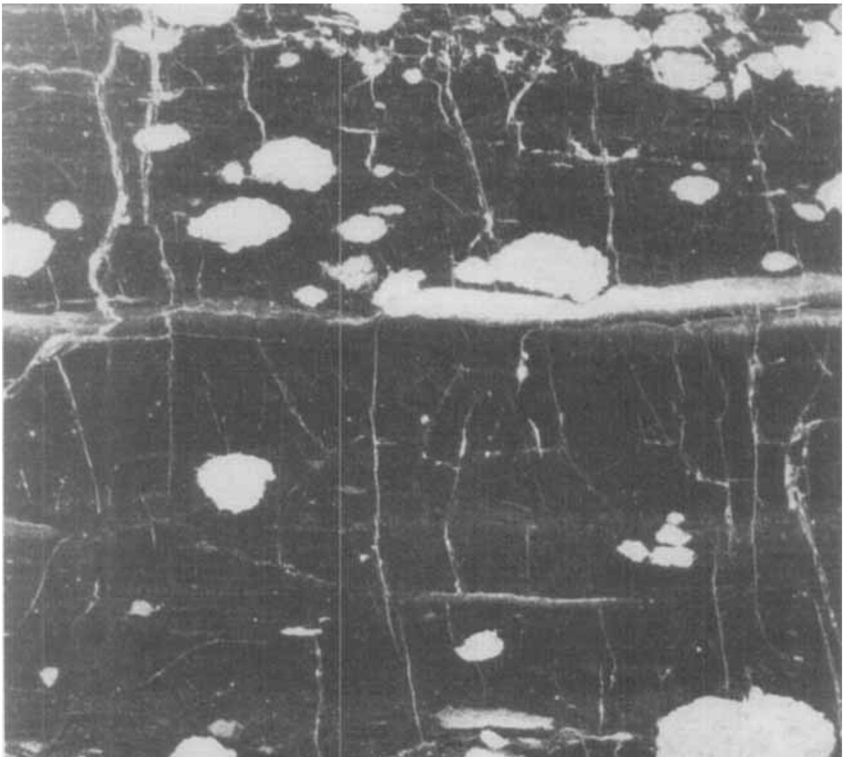

FIGURE 10. A range of lithotypes contains elongate lens of clay and oval siderite nodules. Bulli Seam, N.S.W. The incipient fracture pattern indicates the probable breakage behaviour of the coal with the vitrinite rich coal (dark and massive layers) being more prone to breakage. Polished block, field heights approx. 80 mm.

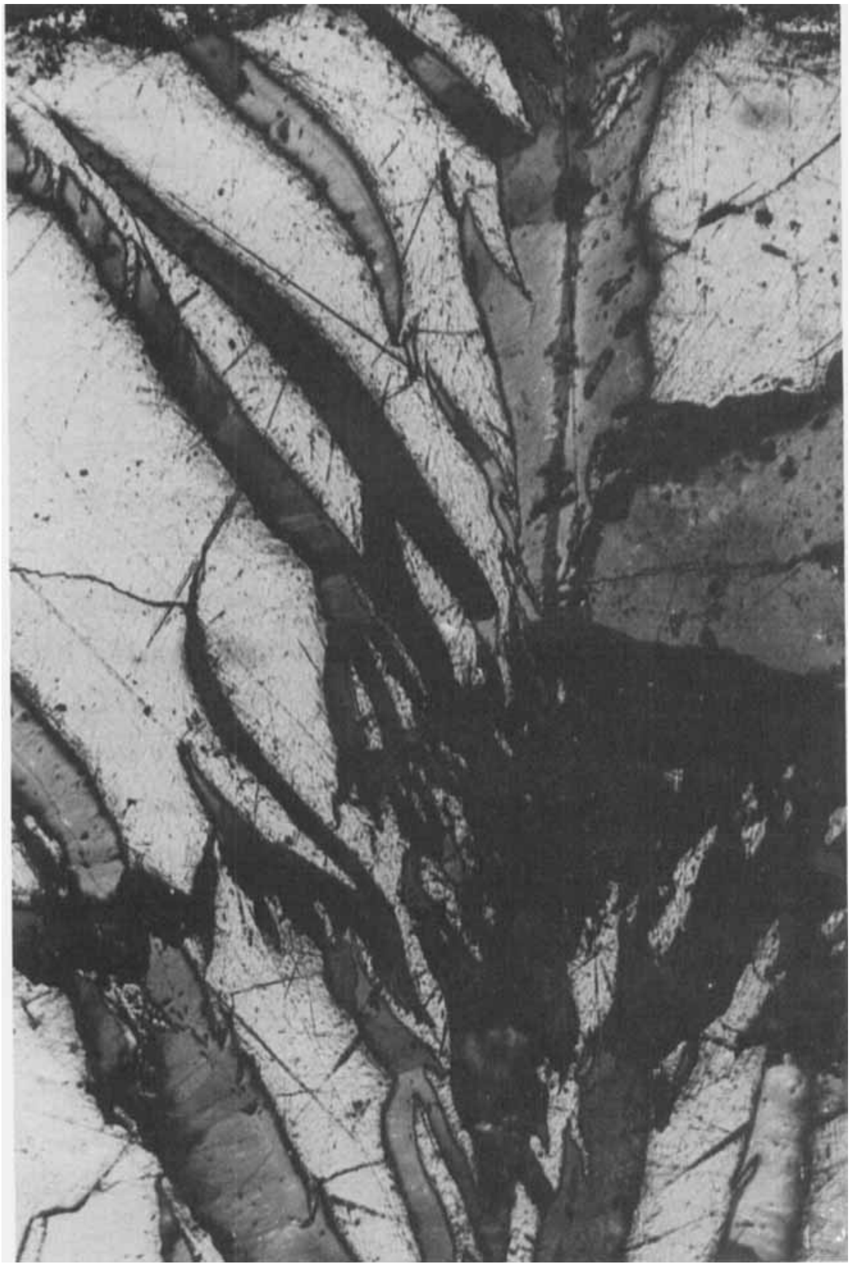

FIGURE 11. Calcite of epigenetic origin occurring as veins within vitrinite. Coal from Singleton Coal Measures, Goulburn Calley No. 1 borehold, N.S.W. Reflected light, oil immersion, field width 0.5 mm.

rejected and significant amounts of mineral matter will report in the product coal. Nor should the problem of achieving effective separation at an economic cost be underestimated. One steel plant in Australia had so much trouble with its fine coal washery that it elected to shut it down and "transfer that part of the washery circuit to the blast furnace."

ASSOCIATION OF MINERALS AND MACERALS

Whilst relatively little is known of the maceral association of impurities other than mineral matter, a great deal of potentially useful information exists on mineral/maceral association.

Attrital maceral assemblages generally have a relatively high content of epiclastic minerals. (Some coals of Tertiary age are usually in this respect in that they contain very little mineral matter. For example, a one metre section in the middle part of the seam at Stockton in New Zealand typically has an ash yield well below 0.5%.) Thus, the inertinite rich lithotypes found especially in the upper parts of seams tend to have relatively high amounts of mineral matter intimately associated with the coal macerals. Cook and Johnson (1975) have suggested that this may be the result of peat ablation in relatively oxidizing environments giving an unfavourable balance between the rate of accumulation of organic material and of mineral matter.

Vitrinite rich coal in some cases has a very low content of mineral matter. A number of authors including Cook (1975) and Shibaoka and Smyth (1975) have noted that many seams deposited in areas of rapid subsidence have both a high vitrinite content and a high mineral matter content present as discrete dirt bands. Vitrinite is also the maceral which is most susceptible to replacement. Thus pyrite and siderite show a preferential association with vitrinite although both minerals will also form in the lumens of fusinite and semifusinite. Chalcedonic quartz, clay and opatite are other minerals which can occur replacing vitrinite. Liberation of mineral matter which is present as a replacing phase is generally difficult to achieve. Significant but variable associations occur and considerable detailed characterization is required for each seam.

SUMMARY

The need for coal cleaning processes will increase markedly as attempts are made to substitute coal for oil and natural gas. A high level of efficiency is desirable both to achieve this aim and to improve resource utilization. Separation processes are possible because coal is inhomogeneous. The heterogeneity is structured and processes which take this structure into account can achieve higher efficiencies at lower costs. The ease of liberation of mineral matter depends upon its mode of occurrence, which is strongly influenced by the mode of origin. Considerable potential exists for improving coal cleaning plants by ensuring that they take maximum advantage of the segregation which occurs during breakage. Together with the systematic characteristics in distribution and association of impurities in coal, a high level of variability exists between seams. Thus, the improvement in the general knowledge of what is to be separated must be complemented by a detailed knowledge of the specific characteristics of each seam which is to be the subject

of a separation process. Since within-seam variation is also important, at least the major characteristics should be mapped prior to mining. Advances beyond the classical separation techniques in commercial use will require a much greater knowledge of the distribution and association of impurities at the maceral level than is generally available at present.

ACKNOWLEDGEMENTS

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