Coal microstructure and secondary mineralization: their effect on methane recovery

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Abstract: Methane production from coal seams rather than porous sandstone reservoirs is now recognized as a valuable and recoverable energy source in Australia. The Bowen Basin of Australia possesses well defined coal seams that contain major methane resources. However, commercial gas production to date has been hampered by the low permeabilities of the coal seams. Recovery of this valuable resource will be assisted by a fundamental understanding of coal microstructures and presence of mineralization, and their influence on the gas flow behaviour through coal.

This paper examines the relationships between coal type, microstructure, secondary mineralization and gas flow behaviour. The study demonstrates that Bowen Basin coals should not be viewed as simply a dual porosity system of micropores which are surrounded by cleats. Instead, studies using scanning electron microscopy show the Bowen Basin coals have a third porosity system comprising a hierarchy of micron-sized fractures and micron-sized cavities at a level between the micropores and the cleat/macropore system, which vary according to coal type.

To determine the influence such microstructures have on the flow of gas through the coal matrix, sorption experiments were carried out on small solid blocks of coal, using a new gravimetric technique. The results demonstrate that a clear distinction exists between diffusivity of dull and bright coals in response to coal microstructure. The gas sorption data suggests that both dull and bright coals can be divided into two categories: coal which have a rapid sorption behaviour and coals which have a slow sorption behaviour.

The results of the sorption experiments indicate that size, continuity, connectivity of the microstructures, and the extent of minerals infilling the fractures and cavities, play a significant contribution to overall permeability, and are likely to play a major rate-limiting factor in the flow of methane through coal at a level between diffusion at the micropore level and laminar flow at the cleat level. The studies indicate that the flow behaviour of gas through coal seams in the Bowen Basin is unlikely to be solely dependent on the cleat system but rather a combination of the cleat, microstructure and secondary mineralization in coal.

Methane primarily resides in the coal as an adsorbed layer on the internal coal surface or as free gas in large pore and fractures. As noted by Harpalani & Schraufnagel (1990*a*), however, a 'Knowledge of conventional gas reservoir modelling is of little value in the case of coalbed methane reservoirs, due to the unique mechanism of gas storage in coalbeds and the unusual flow behaviour of gas in coals'. Present models of methane flow through a coal seam indicate that the absorbed methane after desorption into the gaseous phase must diffuse through the micropore structure of the coal matrix until reaches a cleat (King 1985; Harpalani & Schraufnagel 1990*a*, *b*), followed by Darcy flow through the cleats to a well (Fig. 1).

The relationship of coal seam structure and gas flow behaviour is usually modelled as a dual porosity system of macropores (cleats) surrounding a matrix of micropores (Fig. 2).

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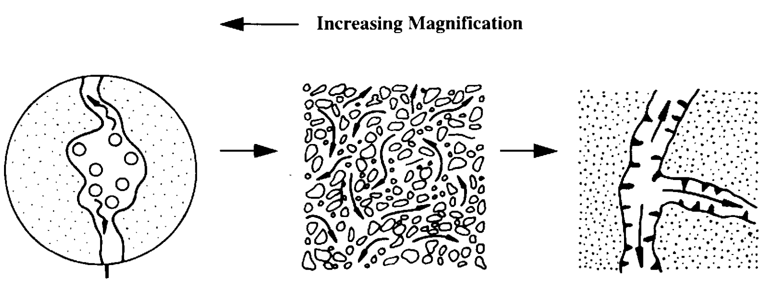

Fig. 1. Current model of methane flow through coal showing desorption, diffusion and Darcy flow.

According to this model, the diffusion is usually modelled using Fick's Law and the free flow is modelled using Darcy's Law. Darcy's Law describes the flow of water through porous media that contains pores of uniform cross-section and of uniform packing, so that pore size, shape, distribution and the connectivity of pores are grouped together under one parameter, permeability. As the cleats are regarded as having a uniform pore geometry (size, shape and spacing) that is representative of coal as a whole, Darcy's Law allows the description of the general behaviour of methane flow through macroscopic structures in coal such as the natural cleat network. The cleats are modelled as defining the size of the matrix blocks, which contain a microporosity, where diffusional flow through the micropores to the cleats is thought to occur. According to this model of gas flow through coals, gas migration is governed by two main factors: (1) the distance methane has to diffuse, which is dependent upon the cleat spacing that delineates the size of the matrix blocks in the coal; (2) the amount of gas flowing through the cleat which is dependent upon the width, length, continuity and permeability of the cleats.

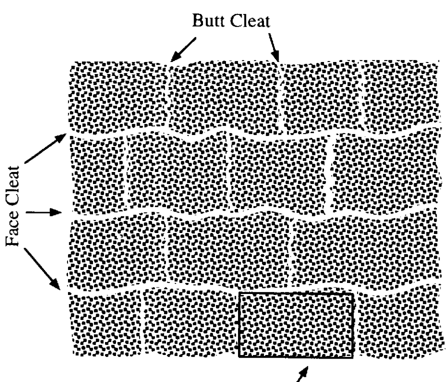

Fig. 2. Dual porosity model of macropores (cleat) and micropores.

This dual porosity model of gas flow (i.e. gas diffusion through the coal matrix block and laminar flow through the cleats) may well apply to predominantly bright coal seams where well defined, open and unmineralized cleats define uniform microporous blocks. Examples of such coals include the Fruitland Seam Fairway coals of the San Juan Basin and the Blue Creek coals of the Black Warrior Basin. Studies using a scanning electron microscope (SEM) show that these coals comprise closely spaced (0.3–1 mm apart), wide (10–60 mm) cleats with narrow (5–20 mm wide) microcleats in between, all of which are open and non-partially mineralized (Fig 3). Typically the nature of these porosities is reflected in their different permeabilities: cleat permeability $\sim$1–50 md; Matrix permeability $\sim 10^{-5}$ to 10^{-9} md.

Given the (micro) structural nature of these coals and the spacing between the cleats and microcleats, it is likely that methane flow through these coals will take place through minute matrix blocks of a size $40\,\mu m \times 40\,\mu m$, suggesting that the distance methane has to diffuse is small.

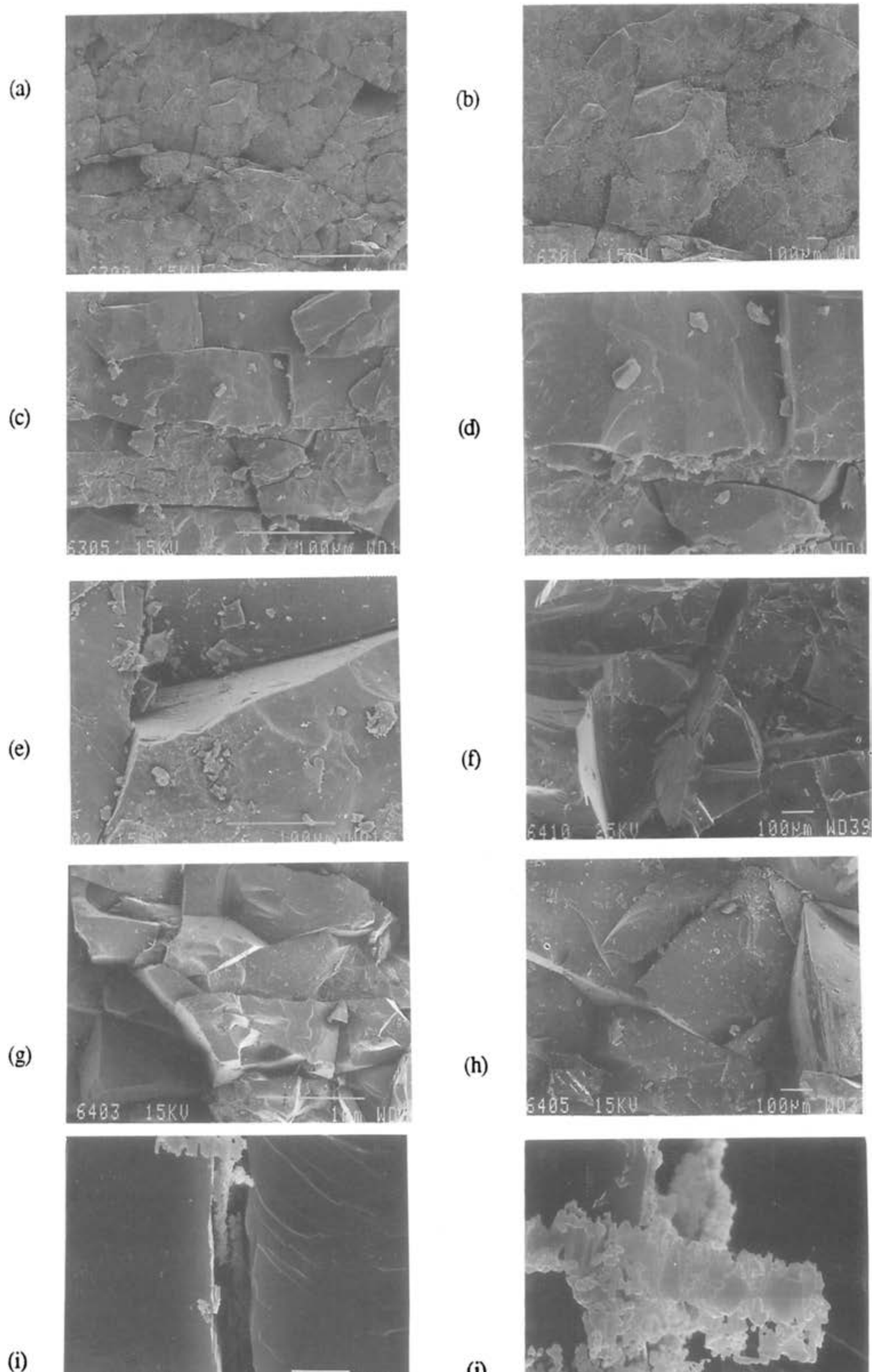

Fig. 3. Australian coals exhibiting closely spaced, wide cleats with attendent narrow microcleats.

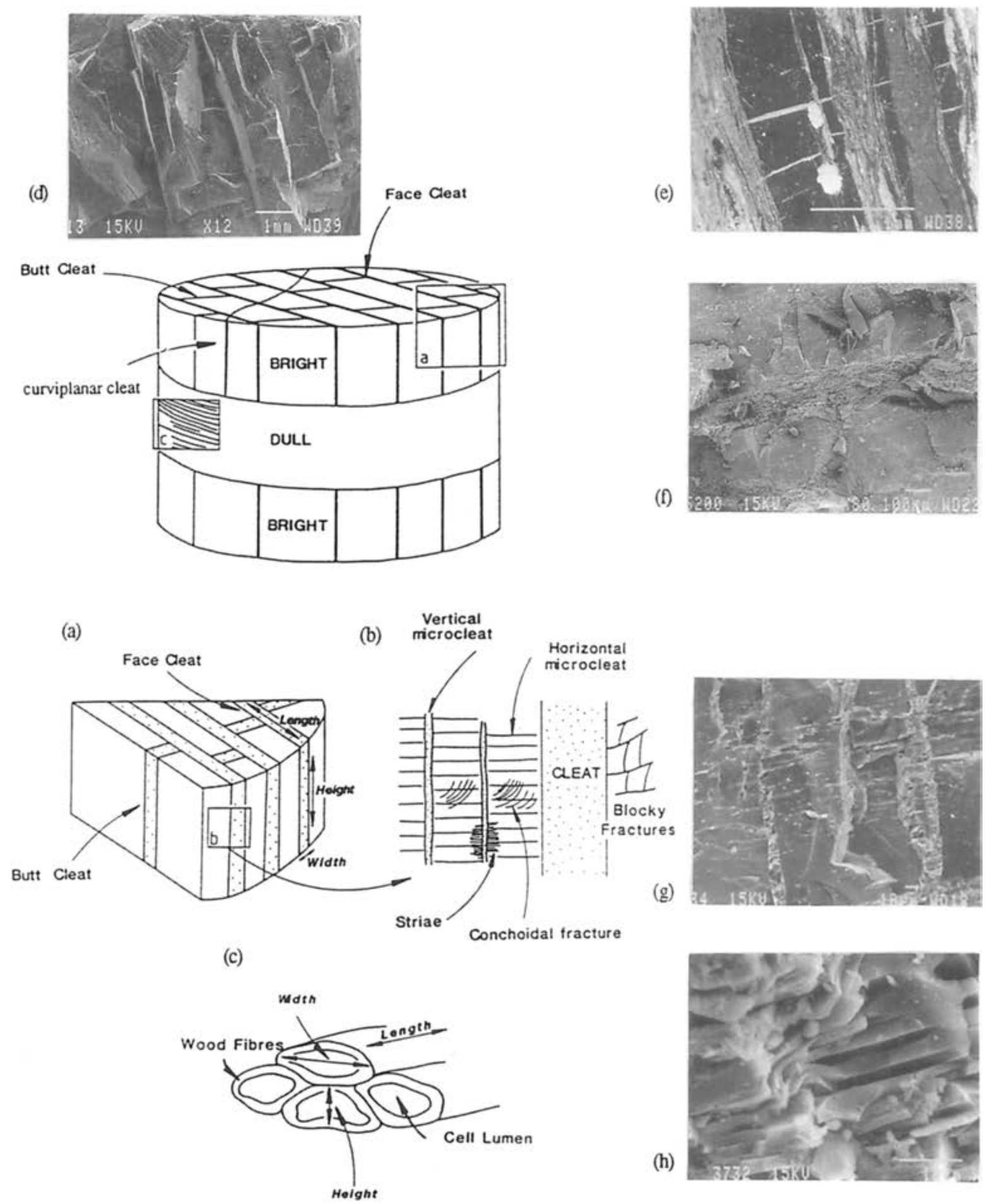

Fig. 4. (**a**)–(**c**) Idealized drawings and (**d**)–(**h**) SEM representations of cleats and microstructures in Australian coals.

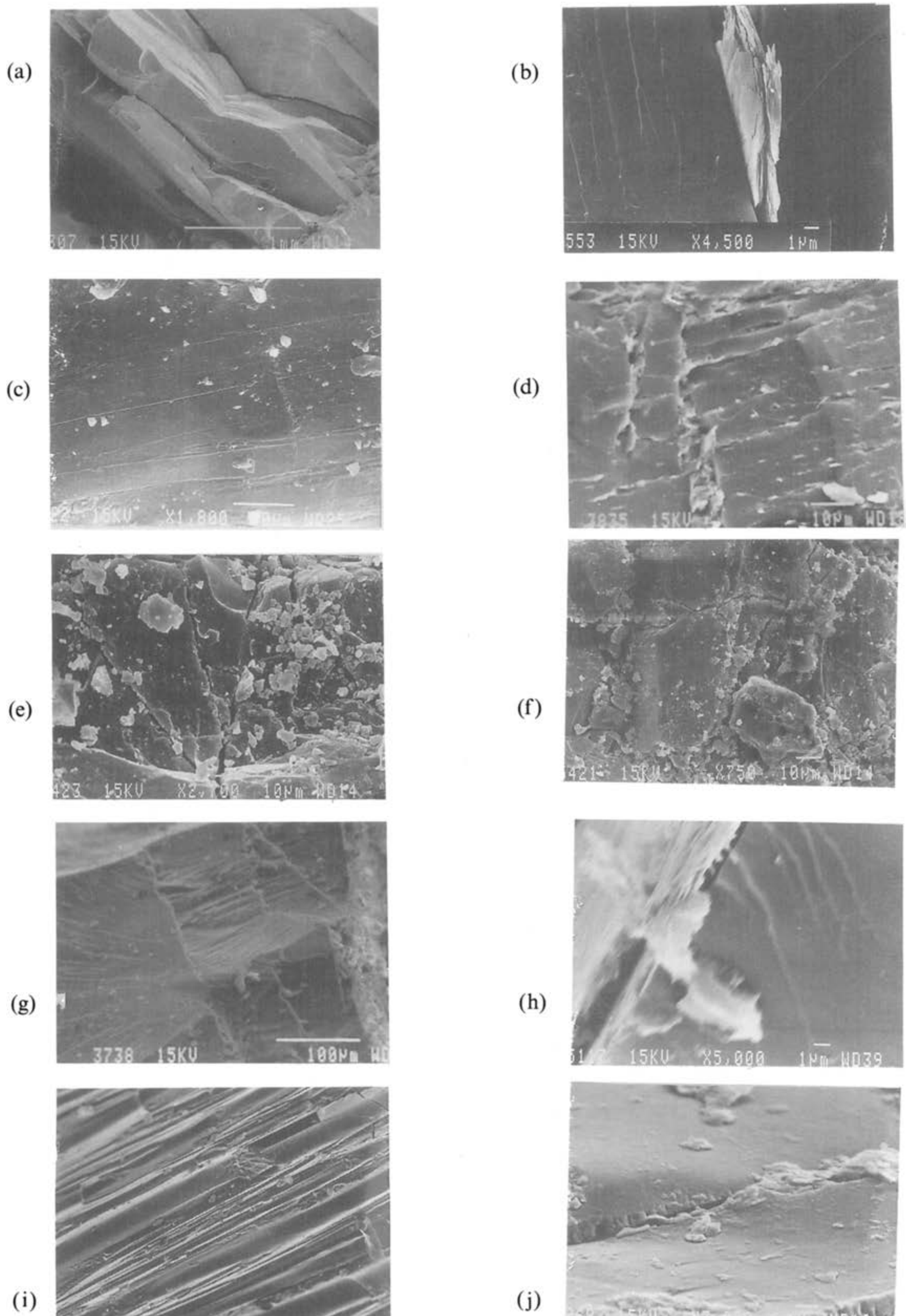

Fig. 5. SEM Photomicrographs exhibiting different styles of microstructures in bright coal.

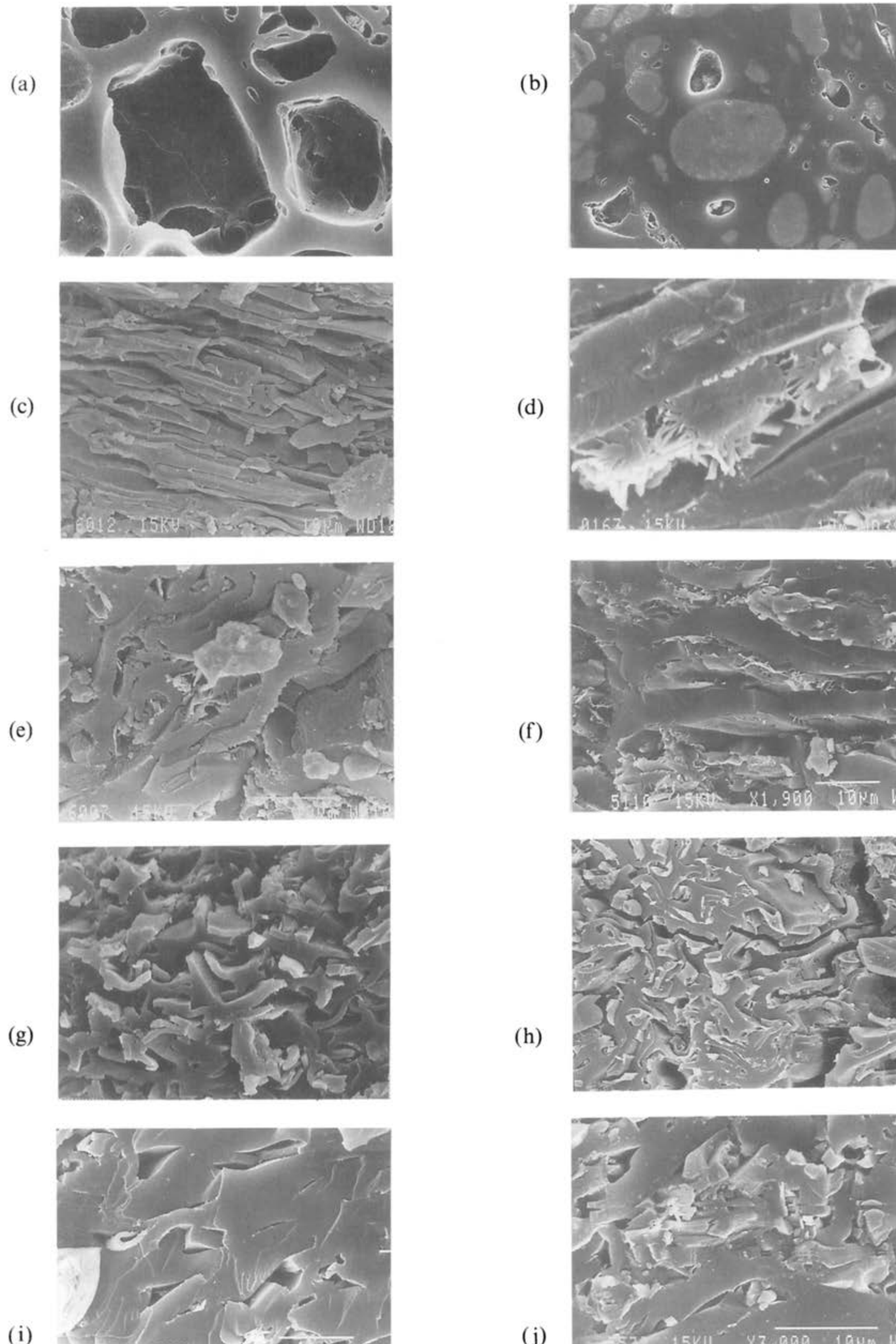

Fig. 6. SEM Photomicrographs exhibiting different styles of phytereal structures.

Structure of Australian coals

The nature of gas flow through Australian coals is perhaps more complex than previously modelled. Examination of coal microstructure using SEM, examples of which are presented here and elsewhere (Gamson & Beamish 1991; Gamson *et al.* 1993), show that Australian coals of the Bowen Basin are not simply a dual porosity system comprising a matrix of micropores that are surrounded by cleats. Instead, these coals are typically compositionally banded, comprising both bright and dull coal types with non-pervasive cleat systems that are commonly restricted to the bright coal layers and terminate abruptly at the bright/dull coal boundary (Figs 4a, e-f). Moreover, a hierarchy of other micron-sized fractures (microfractures) exist in bright coal at a level between the micropores and the cleat system (Figs 4b, g-h; 5a-b). Typically five different types of microfractures can be recognized (Fig. 5): vertical microcleats (5–20 μm wide, 50–500 μm long, and spaced 30–100 μm apart), horizontal microcleats (0.5–2 μm wide, 50–300 μm long and spaced 5–10 μm apart), blocky fractures (1–15 μm wide, 50–200 μm long and spaced less than 100 μm apart), conchoidal fractures (no regularity in spacing) and striae. Striae are the smallest observed microfractures and comprise a number of closely packed parallel laminations or sheet like layers that are 0.1 μm wide, 10–100 μm long and are typically spaced 0.1–0.3 μm.

Dull coals by contrast, tend not to contain microfractures or cleats, and instead typically contain a phyteral porosity of variously sized and shaped microcavities which are associated with the original plant fragments, rather than microfractures. A common component of these phyteral structures are numerous sheet-like structures arranged in a series of stacked layers parallel to bedding which represent remnants of wood fibres. The sheets are typically 2–4 μm thick and are separated by long, cylindrical microcavities of cell lumen that are commonly 2–4 μm high and 10–30 μm wide (Figs 4e, h).

Although the original cavities were probably rectangular in cross-section forming open sieve structures, many of the structures observed in the SEM appear to have been broken and compressed. This has resulted in various morphological structures: needle (fragmented cell walls resulting in networks of pointed, needle-shaped splinters; Figs 6c–d), compressed (cell walls that have been crushed together almost to lines of compression, but are not broken; Figs 6e–f), bogen (cell walls that have been broken and pushed into one another; Figs 6g–h), bogen-compressed (Figs 6i–j) and highly compressed.

Commonly the various microfractures and microcavities are filled with minerals (Figs 5 & 6a–j). The minerals observed infilling such structures include clays (kaolinite, dickite, illite, illite-smectite, vermiculite, chlorite), carbonates (calcite, ankerite, siderite, dawsonite, strontianite) and quartz. SEM examination of the minerals show that they commonly tightly infill the cleats and microstructure space leaving little pore space for water and gas flow. Minerals in bright coals tend to infill cleats and microfractures and occur as either discrete phases or as mixed mineral phases. In contrast, in dull coals minerals occur in the form of either bands/lenses, as fine particles disseminated through the coal, as cavity infill, and/or clay particles interbedded between maceral fragments.

Table 1. *Coal samples measured for their sorption behaviour*

Sample label	Coal group	Seam name	Rank (VR)	Ash content	Surface area (m^2/g, db)	Total porosity	T_{sorp} (minutes)
1A-bright	Fort Cooper	GIRUP	1.66	1.9	261	2.6	148–170
1B-dull	Fort Cooper	GIRUP	1.66	24.5	187	12.4	5–10
2A-bright	Baralaba	"0"	0.81	2.1	237	1.6	288–1288
2B-dull	Baralaba	"0"	0.81	4.2	209	13.7	15–20
3A-bright	Baralaba	"7"	0.88	3.7	277	0.9	415–1309
3B-dull	Baralaba	"7"	0.88	13.8	175	9.0	189–210

Coal microstructure and gas flow behaviour

The presence of micron-sized fractures and cavities in both dull and bright coals at scales below the matrix block and between the micropores and the cleat system suggests that the flow behaviour of gas through these coals is unlikely to be solely dependent on the cleat system, but rather a combination of the cleat, microstructure and extent of mineralization in coal. The size, continuity and connectivity of the microstructures suggests that they contribute significantly to the overall (micro)-permeability, and are likely to have a major role in the flow of methane through coal at a level between diffusion at the micropore level and laminar flow at the cleat level. At present, current models of gas flow do not account for these variations.

To understand the relationship between coal type, microstructure and gas flow behaviour, sorption experiments were carried out on selected samples that have been examined in the SEM for their microstructure. Using a new gravimetric technique developed by Beamish *et al.* (1991), sorption experiments were carried out on small (1 g), solid blocks of dried coal rather than crushed samples, using a microbalance. This technique has been developed to test small samples and allows a closer understanding of the influence coal microstructure has on the diffusivity of coal, which controls the gas flow rate through the coal matrix at a level including the cleats. This new approach to gas sorption studies contrasts with previous studies which have measured coal sorption using a bulk crushed sample to measure a coal's maximum gas storage capacity, but tells us little about the time it takes for methane to flow through a solid coal, i.e. the coal's diffusivity.

The individual pieces of dull and bright coal were desorbed from 1.1 mpa to atmospheric pressure, and the amount of gas released measured with time. To understand the effects of microstructure on sorption, parallel samples were analysed in the SEM.

In total thirty coal samples were used to study sorption behaviour and coal microstructure. Of those samples, six are reported on here as representing typical sorption behaviour

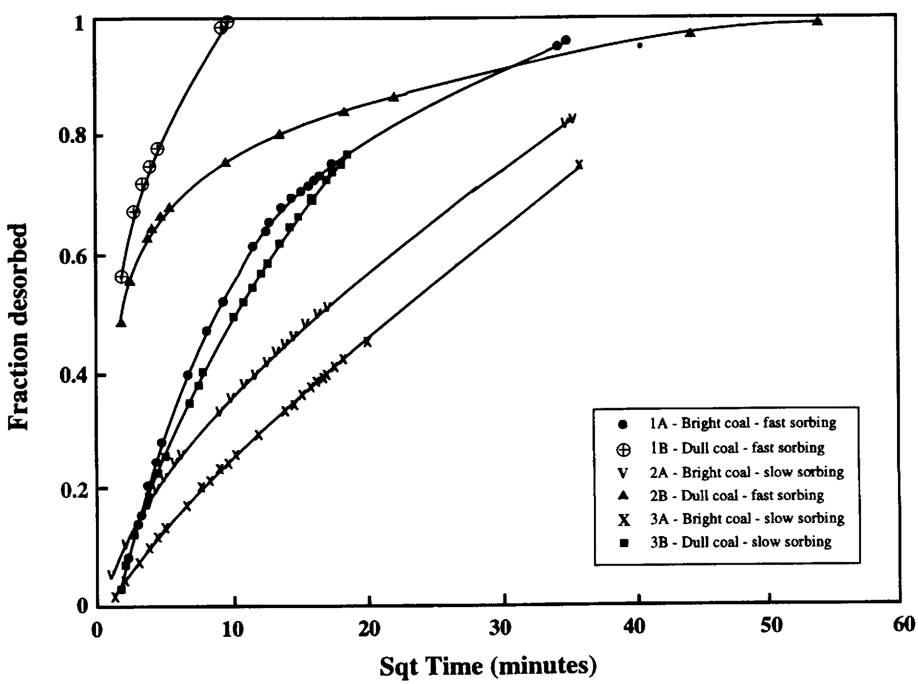

Fig. 7. Experimental desorption results from six coal samples.

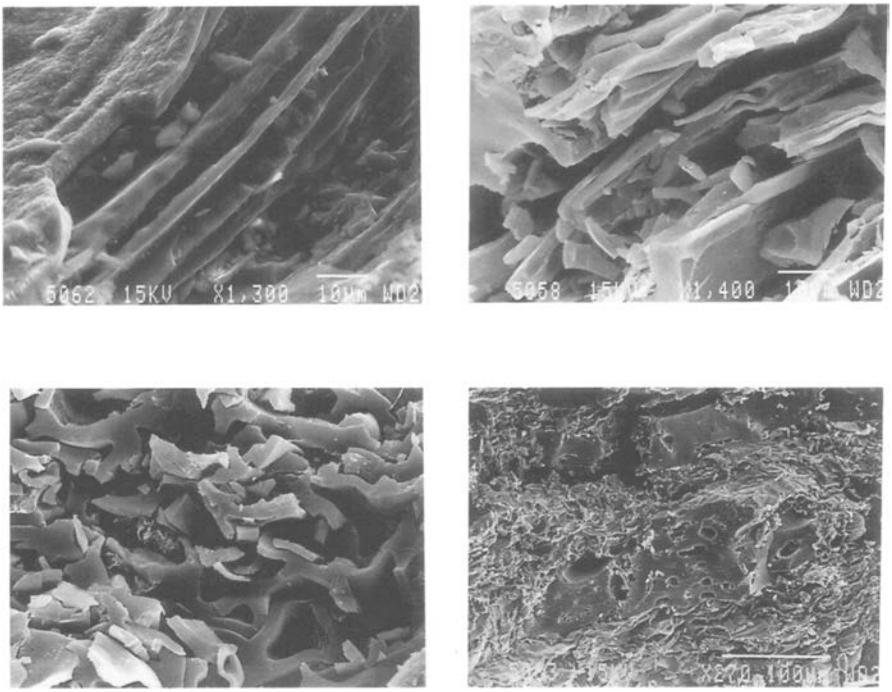

Fig. 8. SEM Photomicrographs of dull, fast sorbing coal.

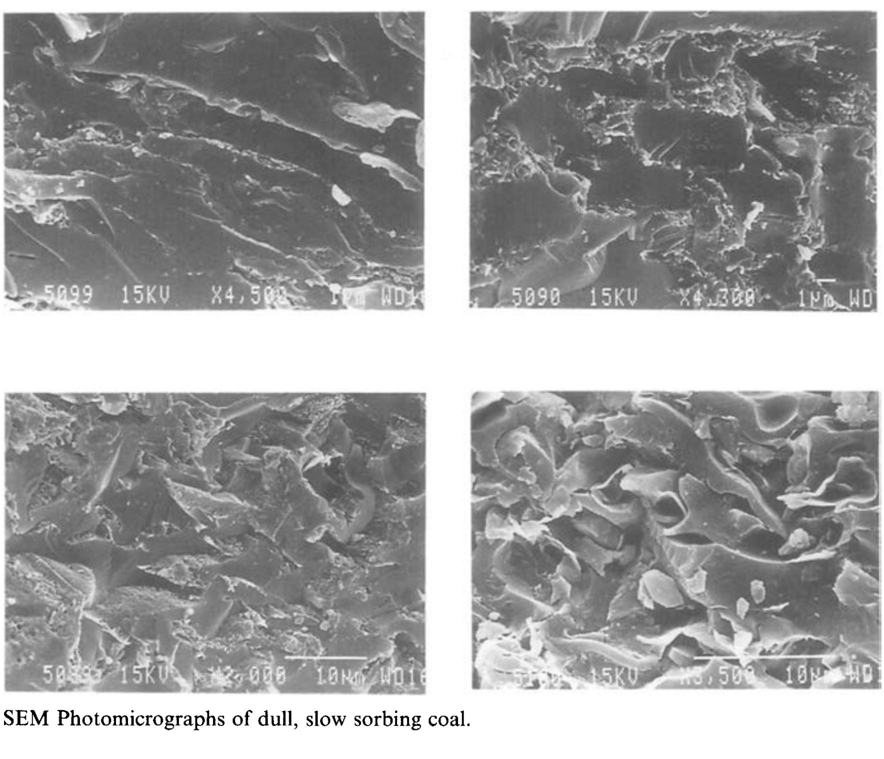

Fig. 9. SEM Photomicrographs of dull, slow sorbing coal.

of dull and bright coal (Table 1). The samples consist of three sets of dull and bright coals that were selected from the same seam and the same depth. To reduce the influence of the effect that sample size has on diffusivity, all the samples were cut into similar sized cubic blocks.

Sorption behaviour of dull and bright coal

The desorption rates of the six samples are shown in Fig. 7, which plots the fraction of gas desorbed versus the square root of time. The desorption data for the six samples (Fig. 7) show that the rate at which methane desorbs from a solid coal differs between coal samples, and coal samples of different coal type. The results show that both the dull and bright coals show two distinct fields of behaviour: fast sorption and slow sorption.

Typically the faster desorbing dull coals have a one-stage sorption process that is characterized by a rapid release of the total gas (Fig. 7). In contrast, the slower desorbing dull coals are characterized by a two-stage sorption process: a first stage during which there is a rapid release of gas, followed by a second stage where sorption is much slower (Fig. 7). Similarly the bright coals are characterized by two types of sorption behaviour. The faster desorbing bright coal is characterized by a two-stage sorption process: a first stage during which there is a rapid release of gas, followed by a second stage where sorption is much slower (Fig. 7). In contrast, the much slower desorbing bright coals are characterized by a one-stage sorption process.

In terms of the time it takes or the dull and bright coals to desorb 63% of their total gas (T_{sorp}) the differences in the rate of sorption are marked and vary from a low of 5 minutes to a high of ~22 hours. Typically T_{sorp} for the various coal classes are:

- Dull coal-fast sorbing: 5–50 minutes
- Dull coal-slow sorbing: 150–250 minutes
- Bright coal-fast sorbing: 150–250 minutes
- Bright coal-slow sorbing: 300–1300 minutes

These differences in T_{sorp} suggest that proportions of these coal classes in any seam profile will govern the rate at which methane will be released. The behaviour of these coal classes can be explained in terms of microstructure and extent of mineralization.

The fast sorbing dull coals contain a high proportion of thick inertinite bands (80–90%) which are predominantly composed of open, unmineralized wood fibres that are either: (1) unbroken, (2) broken and pushed into one another, and/or (3) broken and compressed (Fig 8). In contrast, the slow sorbing dull coals contain a lower proportion of inertinite bands (as little as 50%) and a higher proportion of vitrinite bands. The inertinite bands are predominantly composed of wood fibres with closed, mineralized cell lumens that are either: (1) tightly compressed, and/or (2) broken and tightly pushed into one another. In addition, the bands of vitrinite contain micro-fractures which are tightly infilled with minerals (Fig. 9).

The fast sorbing bright coals are characterized by a well defined cleat network. The cleats are moderately infilled with minerals although pore space between the mineral grains is apparent. Microcleats, blocky fractures and smaller microfractures are common in the faster sorbing coals and are typically open and unmineralized (Fig. 10). In contrast, the slow sorbing coals bright coals are characterized by tightly infilled face and butt cleats, tightly infilled microcleats and other microfractures, and/or no microfractures (Fig. 11).

The differences in sorption behaviour shown by the two coal classes can best be explained in terms of the macropore and micropore components of the coal. The simplest form of comparison of the desorption data is to apply a spherical unipore model. Of the six coals however, only three of the coals, 1B-dull, 2A- and 3A-bright fit the unipore spherical model. To explain this, the release of methane in such coals may be considered a one-stage sorption process. In the dull coal, 1B, only macropore sorption occurs due to the domination of macropores. In contrast, the one-stage sorption process in the bright coals, 2A and 3A, fit a unipore spherical model due to the domination of micropore diffusion.

The three other coals, 1A-bright, 2B- and 3B-dull, however, do not fit a unipore spherical model due to a distinct curvature in the desorption curve (Fig. 7). To explain this phenomenon, the sorption behaviour is divided into a microsphere (micropore) component which is surrounded by a macrosphere (macropore) component (Ruckenstein *et al.* 1971). The release of methane in these coals may be considered a two stage sorption process, where sorption in the

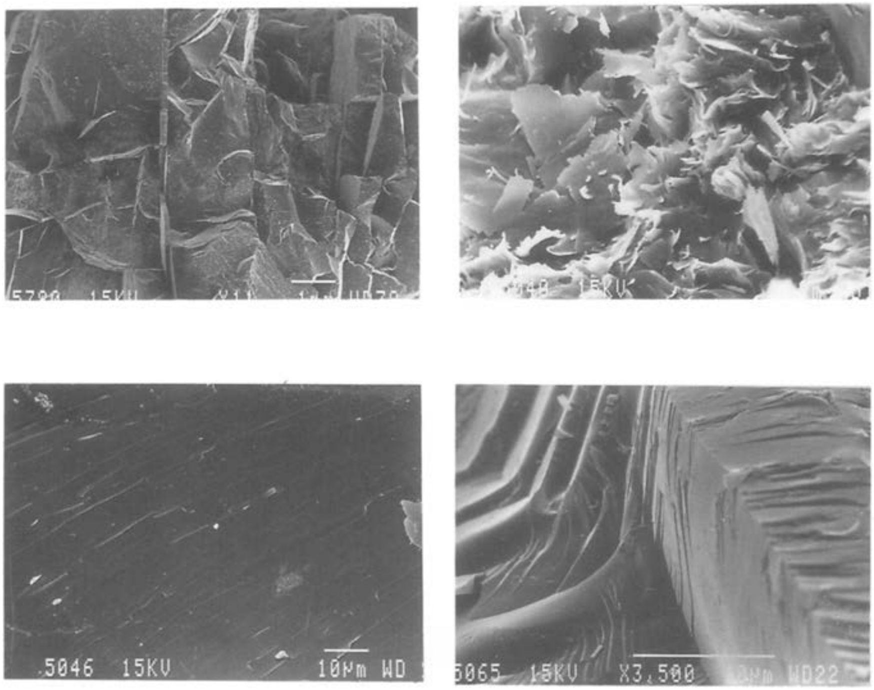

Fig. 10. SEM Photomicrographs of bright, fast sorbing coal.

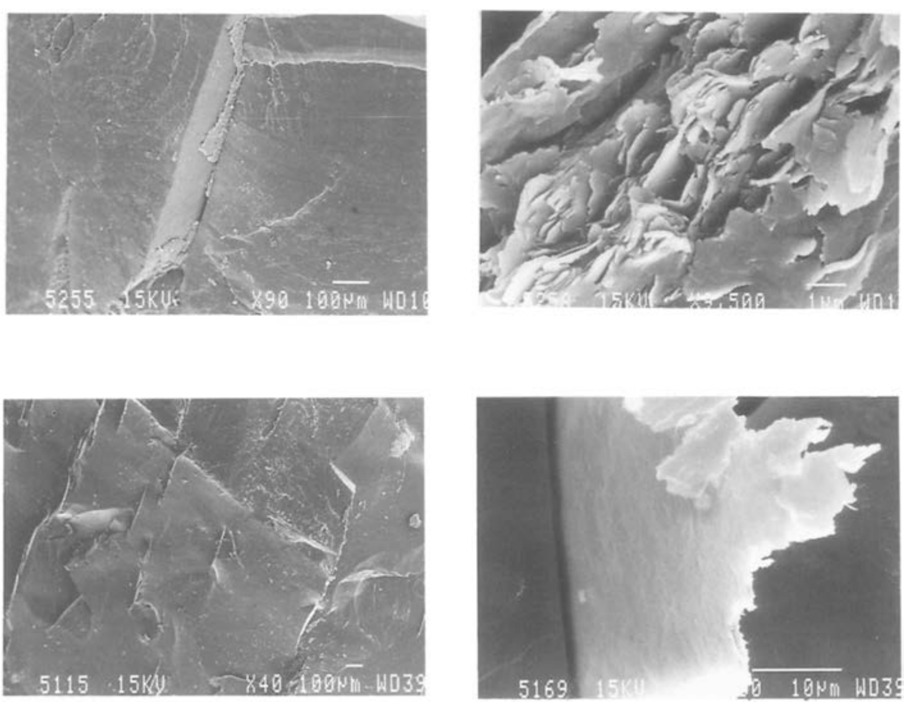

Fig. 11. SEM Photomicrographs of bright, slow sorbing coal.

macropores is much faster than in the micropores so that equilibrium is essentially achieved in the macropores before any appreciable release by the micropores is observed. Consequently a first stage is observed during which only macropore sorption occurs, followed by a much slower second stage during which macropore sorption is at equilibrium, and only micropore sorption occurs.

Gas flow through coals

A combination of scanning electron microscopy and sorption testing using a microbalance has shown that the time taken for methane to travel through the coal matrix varies according to microstructure and is reflected in significant variations in T_{sorp}.

Scanning electron microcopy has shown, that at a scale between the micropores and the cleat system, Australian coals contain a range of microfractures in the bright coals and a range of microcavities in the dull coal, of various pore shapes and sizes which vary according to the degree of mineralization. Consequently, it is likely that methane flow through the matrix will take place not through pores of similar geometry as previously suggested by a dual porosity model, but rather through a complicated network of interconnected microfractures and microcavities, of varying size, shape and cross-section, and minerals filling pore space.

According to current models of gas flow through coals (Fig 1), however, the flow of methane is dependent upon the effective permeability of the coal, i.e. the cleat network. Assuming the diffusion of methane begins and presumably finishes at the micropore level where the majority of gas is stored as an adsorbed layer (and where micropores 4–20 angstroms in diameter are joined by minute passages), as previous models suggest, then methane flow from the micropore system to the cleats in Australian coals must rely upon the effectiveness of the microstructure system to transport the methane.

The various microfractures and microcavities, and the different sorption behaviours exhibited by the dull and bright coals, suggest that microstructures play a rate-limiting role between diffusion at the micropore level and flow at the macropore or cleat level. In terms of modelling, this suggests that additional steps may be involved in the flow of methane through Australian coals (Fig. 12). Four steps are proposed (Fig. 12):

- Step 1. Diffusion from and through the micropores to microfractures in the bright coal and microcavities in the dull coal.
- Step 2. Diffusion and/or flow of methane through microfractures and microcavities partly blocked by diagenetic minerals. Methane flow would be dependent on the size and connectivity of the pore space between the mineral infilling.
- Step 3. Flow through open, unmineralized microfractures in the bright coal and microcavities in the dull coal.
- Step 4. Gas movement through cleats and joints to the well base. Where cleats are generally infilled by minerals, and mostly this infill forms a tight seal, gas movement will be either completely blocked or be by diffusion.

These additional steps are not presented so as to complicate the issue by incorporating more tiers into the gas production process. Instead the extra steps show that microstructures are likely to play an important and probably significant rate-limiting role at scales before gas transport at the cleat or fracture level.

Importantly, the effectiveness of methane flow through the microstructures, as opposed to the cleats, would be ultimately influenced by several microscopic considerations, which include the shape and size of microstructures, microstructure distribution (density, orientation and continuity), connectivity of the microstructures and the cleat system, the amount of fracture infilling with secondary minerals, clay dispersed through the organic matrix, and the change in stress conditions after stimulation. Each of these microscopic considerations will have a different effect on the quantity, rate and direction of gas flow through the coal. For Australian coals the term micropermeability is introduced and refers to the conductivity of the microstructures.

Implications of microstructures on gas flow models

In terms of gas flow modelling, such variations in coal seams have important implications for present models of gas flow through coal. Currently, models subdivide matrix blocks where diffusion dominates from that involved

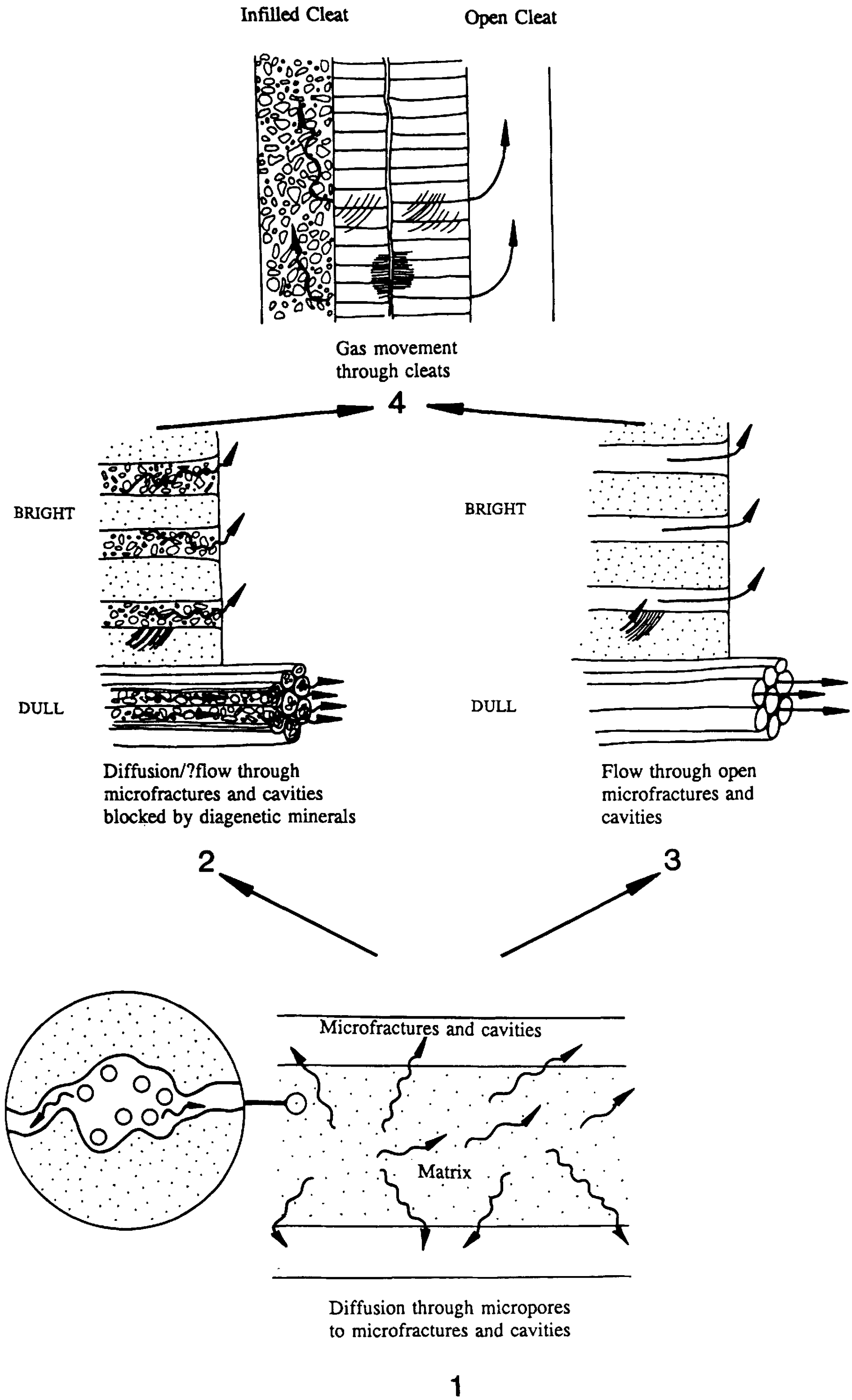

Fig. 12. Enhanced desorption model for Australian coals proposing four distinct steps of methane migration.

with laminar flow, on the cleat spacing. Because the effects of microstructure on diffusivity are considered to be real, the presence of open and continuous microstructures in coal suggests that laminar flow is likely to begin at levels smaller than that identified at present by the spacing of cleats. To accommodate the effects of microstructure into models of gas flow, it is necessary to redefine the effective block size within which diffusion dominates.

In bright coals, where there is a hierarchy of open, continuous and connecting microfractures between the cleats, the effective block size is probably not that being defined at present by the cleat spacing, but somewhere between the cleats and the microfractures. In contrast, where cleats and microfractures are infilled with minerals and the pore space for laminar flow is reduced, the effective block size may be larger than previously assumed, and may be more effectively defined by other more widely spaced, open fractures (joints) in the bright coals. Ultimately this could reflect fracture sets beyond the core.

In dull coals however, where there are usually no cleats, it is more difficult to define the effective block size for gas flow. The microstructural and sorption results indicate that where dull coals contain open, unmineralized structures, and macroporous flow dominates, the effective block size in these coals correlates to the size of the phyterals, which could be in the order of 10 μm. However, where microstructures in the dull coals are heavily mineralized it is probable that the block size relates to a larger composite dimension.

These differences in diffusivity of a coal have important implications for gas drainage. Although a general relationship between coal rank and gas content is well documented (Kim 1977), and that bright coals have a greater storage capacity than dull coals of equivalent rank (Beamish & Gamson 1993), it does not follow, however, that bright coals offer a greater potential for methane flow (i.e. a higher diffusivity) than dull coals of equivalent rank. Instead, it has been shown that dull coals generally have a higher diffusivity than bright coals due to the greater macropore porosity, and associated small block sizes for diffusion, than bright coals of equivalent rank. This is important, for it suggests that areas of low rank coal with low gas contents, due to their low storage capacity, may offer a better gas flow rate than higher rank coals with higher gas contents due to differences in diffusivity between the coals. Equally significant, it also suggests that coals with high permeability may not offer higher gas flow rates than coals with lower permeability due to low diffusivity.

Conclusions

Scanning electron microscopy has shown that Australian coals should not be viewed as simply a dual porosity system of micropores which are surrounded by cleats, and instead, viewed as having a third porosity system comprising a hierarchy of micron-sized fractures and micron-sized cavities at a level between the micropores and the cleat/macropore system, which vary according to coal type.

Examination of the sorption behaviour of these coals suggests that bright and dull coals exhibit distinct fields of behaviour: fast sorption and slow sorption. The behaviour of these classes indicate that the size, continuity, connectivity of the microstructures, and the extent of minerals infilling the fractures and cavities play a significant contribution to overall permeability, and are likely to play a major rate limiting factor in the flow of methane through coal at a level between diffusion at the micropore level and laminar flow at the cleat level. A new model is presented to account for these variations in coal type, microstructure and mineralization.

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