

A Model for Determination of Methane Flow Parameters in Coal from Desorption Tests

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This paper presents the results of a study of the mechanics of methane gas flow in coal. Desorption tests carried out on cylindrical coal specimens showed that an analytical formulation can be made to model the process of gas flow in coal based on an integration of Darcy's law, Fick's law and Langmuir's relation. The diffusivity term in the flow equation varies directly with local gas content. Permeability tests carried out on the same coal result in permeability constants with values higher than equivalent permeabilities obtained from desorption tests by many orders of magnitude. The study also revealed that permeability is highly dependent on the applied gas pressure and the rock stress.

Introduction

Methane gas is a by-product of the coalification process. In many collieries, methane gas (CH_4) has continually posed serious hazards causing disasters such as explosions, and instantaneous outbursts of coal and gas, or fires. Drainage of methane gas has thus become a natural measure to increase safety in many 'gassy' coal mines throughout the world. Furthermore, with the recognition that proven reserves of petroleum fuels could be exhausted within the foreseeable future, increasing attention has been focused on methane gas drainage from coal seams as an economic source of natural gas production.

Methane emission from coal into mine workings results from various mining processes. The principal source of methane emission into mine workings is the seam being worked and adjacent seams which are penetrated by the fracture system.

Gas emission from the seam being worked mainly takes place at the exposed faces. The rate of gas emission is dependent on the advance rate, seam gas content, and coal permeability. In the vicinity of an active face, the permeability of the seam may increase several fold as a result of fracturing by overstraining.¹⁻³

Methane emission from goaf, adjacent strata and seams accounts for the highest proportion in a district. In some of the Australian mines, this cause accounts for more than 75% of the total emission, and in mines using free-drainage of the mined seam more than 90% gas content in the ventilation comes from this source.

Factors influencing methane emission

Methane emission into mine workings results essentially from pressure differences between seam gas and the

atmosphere of workings. Under normal conditions, the rate of methane emission into mine workings is a function of porosity, permeability and seam gas content. The porosity of coal, comprising macroporosity and microporosity, is influenced by the magnitude of the confining stress. The magnitude of coal porosity may alter drastically following fracturing or stress relaxation associated with mine excavations.

Permeability is basically a function of the porosity and the tortuosity of the interconnected network of void spaces in the coal. It is therefore also dependent on *in-situ* stresses. Studies of the stress dependence of permeability of coal have shown that permeability may decrease under stress by a factor of 1 to 3.⁴⁻⁶

Curl⁷ and Hargraves⁸ have given detailed reviews on the nature and characteristics of methane content in coal seams. Basically, the sorptive capacity of coal increases with coal rank and decreases with increasing moisture content of coal and increasing temperature.

Methane gas present in coal exists in two states: the free gas state and the adsorbed state. Free gas is present in the fractures and macropores of the coal, and its behaviour can be described by Boyle's law and the kinetic theory of gases. In the adsorbed state, gas molecules adhere to the large internal surface area of coal (20 to 200 m^2/g). At normal coal bed pressures, adsorbed gas occupies roughly 90 to 95% of the total gas content in coal, and there is a continual interchange of molecules between the free gas and the adsorbed gas. The content of the adsorbed gas is related to the free gas pressure in equilibrium by experimentally determined isotherms. There are many mathematical models used to describe

the process of methane adsorption, or desorption in coal, and the Langmuir model appears to be favoured by many workers.

The quality of free gas present in the macropore space of coal can be described by Boyle's law:

$$PV = ZnRT \quad [1]$$

where

P is the pressure (kPa)

V is the macropore volume (m^3)

n is the number of moles

R is the universal gas constant (8,30438 kPa/kmol/°K)

T is the absolute temperature (°K), and

Z is the gas deviation factor defined as the ratio of volume actually occupied by a gas at a given pressure and temperature to the volume it would occupy if it behaved ideally. At temperatures existing in most mines, the variation of Z is negligible at low pressures. For methane the ideal gas equation is acceptable at pressures up to 4 000 kPa, but for carbon dioxide, the ideal gas equation is inapplicable at pressures beyond 1 100 kPa.⁹

In terms of coal macroporosity Φ_M , and standard conditions of pressure and temperature, the quantity of free gas per unit volume of coal, C_v (cm^3/cm^3) is computed by:

$$C_v = \Phi_M \frac{P}{P_0} \frac{273}{T} \quad [2]$$

in which

Φ_M is the macroporosity, i.e. fraction of macropore space to total coal volume

P_0 is the standard atmospheric pressure (101,3 kPa)

T is the absolute Kelvin temperature.

It is widely known that the free gas makes up only a small fraction (5 to 10%) of the total gas content. The volumetric free gas content, C_v , is proportional to the macroporosity, Φ_M , which is affected by changes in *in-situ* stresses and the moisture content of coal or rock. Φ_M can vary between 0,01 and 0,15 (cm^3/cm^3) for coal, as commonly reported in the literature.

At equilibrium, a unique relationship exists between the quantity of adsorbed gas and the free gas pressure, commonly described by the Langmuir equation:

$$C_p = \frac{A B P}{1 + B P} \quad [3]$$

where

C_p is the quantity of adsorbed gas at pressure P , cm^3/cm^3 of coal, or more commonly m^3/ton of coal. A and B are Langmuir's constants, in units of cm^3/cm^3 (or m^3/t) and kPa^{-1} , respectively. Note that in the case of m^3/t -unit being used, the conversion to cm^3/cm^3 is obtained by multiplication with the coal dry density (t/m^3).

The Langmuir equation describes the mechanics of adsorption or desorption, and its curve is called the sorption isotherm. The constants A and B vary principally with coal rank, temperature and moisture content.

Basic flow equations

The mechanism of methane transmission in coal is a complex combination of a number of different physical pro-

cesses, and for practical purposes, methane gas transmission in coal is normally modelled by Darcy's law and Fick's law.

Darcy's law hold that the flow rate of a fluid through a porous medium is proportional to the potential or pressure gradient:

$$q_D = - \frac{K A_s}{\mu} \frac{\delta P}{\delta x} \quad [4]$$

where A_s is the cross-sectional area perpendicular to the macroscopic velocity vector, K is the permeability of the medium, $\delta P/\delta x$ is the pressure gradient, and μ is the fluid viscosity (N.B. for methane, $\mu = 10,87 \times 10^{-6}$ Ns/ m^2 , 293°K). It is recalled that the Darcy type of flow is a laminar-viscous flow in which the flow paths are either prismatic or streamlined.¹⁰ Darcy's law is essentially empirical and despite many reported theoretical anomalies in the literature of flow in porous media, it remains the main mathematical tool for analysing flow in porous media for a diversity of applications, for instance petroleum engineering, soil science and geotechnical engineering. One particular consequence of Darcy's law is that there should be no gas molecule slippage along the internal wall of the pores. The gas slippage effect, commonly called the Klinkenberg effect, has been accounted for in a recent flow model.¹¹ The Klinkenberg effect is most pronounced at low pressures and can be incorporated in the flow equation by adjustment of permeability values.¹²

$$K = K_\infty \left(1 + \frac{a}{P}\right) \quad [5]$$

where P is the pressure of flow, K_∞ is the permeability at very high pressure, and a is a constant.

Fick's law describing diffusive flow under a concentration gradient can be written as:

$$q_F = - D A_s \frac{\delta C}{\delta x} \quad [6]$$

where C is the species concentration (normally in cm^3/cm^3), A_s is cross-sectional area (cm^2), D is the diffusion coefficient (cm^2/s), and the negative sign indicates that the mass transfer takes place in the direction of decreasing concentration.

Under the form of Equation [6], the diffusion coefficient already takes account of the porosity and tortuosity of flow path in the coal body,¹³ i.e. the diffusion coefficient of methane gas in coal is different from that of methane in air. As a consequence of the dependence on porosity, the diffusion coefficient used in Fick's equation for porous media [6] is a sensitive function of the state of stress and fracturing in the coal body. The characterisation and measurement of the diffusion coefficient is therefore a complex aspect in the physical and mathematical modelling of methane emission from coal. For dilute gas mixtures, the diffusion coefficient is a function of temperature, pressure and is almost independent of composition.¹⁴ Such a relationship for the case of water vapour diffusion in air has been given by Krischer and Rohnalter¹⁵ as:

$$D = D_0 \frac{P_0}{P} \frac{T}{T_0} \quad [7]$$

where D_0 is the diffusion coefficient measured at $T_0 = 273, 15^\circ\text{K}$, and $P_0 = 101,325 \text{ kPa}$.

It is also important to note that the measurement of Fick's coefficient of diffusion in coal has often been made by means of many mathematical models which considered methane transmission in coal to be a combination of two processes in series: diffusion of methane from an 'elemental block' of size 'a', followed by rapid diffusion or Darcy's flow in the macropore system.¹⁶⁻²⁰

Formulation of a unified model

Let us consider that methane transmission in coal takes place under two simultaneous and parallel processes described, i.e. Fick's diffusion and Darcy's laminar flow. Fick's diffusion takes place mainly in the micropore system whereas Darcy's flow occurs only in the macropore channels.

For simplicity, we use notations for the x -coordinate only. Fick's law of diffusion applied to adsorbed gas present in the micropores can be written as:

$$q_F = -D_F \frac{\delta C_a}{\delta x} \quad [8]$$

where

q_F is the rate of diffusing methane across a unit cross-sectional area, or diffusing velocity (cm/s),

D_F is the diffusion coefficient (cm^2/s), already incorporating the effects of micropores and tortuosity,

C_a is the concentration of adsorbed gas per unit volume of coal (cm^3/cm^3). C_a can be estimated from the Langmuir equation [3] in which the constant A is obtained from the customary Langmuir constant by multiplication of the latter with the dry density of coal (in g/cm^3 or t/m^3).

Concurrently with the diffusion process is the Darcy-type flow taking place by free gas molecules in the macropore system, under a pressure gradient:

$$V_x = -\frac{K}{\mu} \frac{\delta P}{\delta x} \quad [9]$$

where

K is the permeability (cm^2)

μ is the methane viscosity ($1,087 \times 10^{-5} \text{ N s}/\text{m}^2$ at 293°K), and

P is the gas pressure (kPa).

The continuity and mass conservation principle can be stated as the change in the free gas density in the pore space of a controlled volume is equal to the net difference between the mass inflow and outflow relative to the controlled volume plus the amount of gas transferred from the adsorbed phase during an infinitesimal time interval. In mathematical notation, this principle can be written as:

$$\frac{-\delta(\rho V_x)}{\delta x} = \Phi_M \cdot \frac{\delta \rho}{\delta t} + \Gamma \quad [10]$$

where

Γ is the rate of gas transfer from the adsorbed phase,

Φ_M is the macroporosity, and

ρ is the density of free gas in the macropores.

Similarly, gas movement in the adsorbed state can be

described by Fick's law and the continuity principle as:

$$\rho_0 D_F \cdot \frac{\delta^2 C_a}{\delta x^2} = \rho_0 \frac{\delta C_a}{\delta t} - \Gamma \quad [11]$$

in which

ρ_0 denotes the density of methane under NTP conditions,

C_a is the adsorbed gas content (cm^3/cm^3) per unit volume of coal,

D_F is the Fick diffusivity of methane in coal, and

Γ is the rate of gas desorption from adsorbed phase to free gas.

Summing equations [10] and [11] we obtain:

$$\rho_0 D_F \cdot \frac{\delta^2 C_a}{\delta x^2} - \frac{\delta(\rho V_x)}{\delta x} = \rho_0 \frac{\delta C_a}{\delta t} + \Phi_M \cdot \frac{\delta \rho}{\delta t} \quad [12]$$

and denoting

$$\lambda = \frac{\delta C_a}{\delta P} = \frac{AB}{(1 + BP)^2} \quad [13]$$

Equation [12] becomes:

$$[\rho_0 D_F \lambda + \rho \frac{K}{\mu}] \cdot \frac{\delta^2 P}{\delta x^2} = [\lambda \rho_0 + \Phi_M \rho c] \frac{\delta P}{\delta t} \quad [14]$$

or

$$D \frac{\delta^2 P}{\delta x^2} = \frac{\delta P}{\delta t} \quad [15]$$

in which

$$c = \frac{1}{\rho} \frac{\delta \rho}{\delta P}, \text{ the gas compressibility factor } \frac{1}{P}$$

and

$$D = \frac{\rho_0 D_F \lambda + \rho K \mu^{-1}}{\lambda \rho_0 + \Phi_M \rho c} \quad [16]$$

D is the total diffusivity, accounting for Fick's diffusion, Darcy's flow, and phase interchange by desorption.

If D is considered to be dependent on pressure, the general form of the methane transport model can be written as:

$$\frac{\delta P}{\delta t} = \frac{\delta}{\delta x} (D_x \frac{\delta P}{\delta x}) + \frac{\delta}{\delta y} (D_y \frac{\delta P}{\delta y}) + \frac{\delta}{\delta z} (D_z \frac{\delta P}{\delta z}) \quad [17]$$

for the general three-dimensional system, assuming that D is isotropic.

Equation [15] can be rewritten for the cylindrical coordinate system as:

$$\frac{\delta P}{\delta t} = \frac{1}{R} \cdot \frac{\delta}{\delta R} (D \cdot R \frac{\delta P}{\delta R}) \quad [18]$$

where $D = D(P)$, i.e. D is a function of gas pressure.

The mathematical model given by [18] was applied to the desorption data on coal samples. Crank's classic solution of heat conduction in solids²¹ was initially used to solve the methane desorption problem [18].

The Crank-Nicholson form of Equation [18] can be written as:

$$\begin{aligned} P_j^n - P_j^{n-1} &= \frac{\Delta t}{4(\Delta R)^2} [(2j+1)D_{j+1/2}(P_{j+1}^n + P_{j+1}^{n-1}) \\ &- 2j(D_{j+1/2} + D_{j-1/2})(P_j^n + P_j^{n-1}) \\ &+ (2j-1)D_{j-1/2}(P_{j-1}^n + P_{j-1}^{n-1})] \end{aligned} \quad [19]$$

which yields a system of linear equations in P_{j+1}^n , P_j^n , and P_{j-1}^n . Subscript j denotes annular node and superscript n denotes the time $t = n \cdot \Delta t$.

At the centre of the cylinder sample, $j = 0$ and $R = \Delta R/2$, the finite difference scheme can be approximated as:

$$P_0^n - P_0^{n-1} = \frac{2\Delta t}{(\Delta R)^2} [D_{1/2}(P_1^n + P_1^{n-1}) - D_0(P_0^n + P_0^{n-1})] \quad [20]$$

A Crank-Nicholson finite difference scheme coded in BASIC on a microcomputer has been used to obtain a better fit of test results on desorption.

Laboratory studies

Laboratory studies on the desorption of gas from coal cores of 50 to 55 mm diameters were conducted. Specimens were cored out from a sample block of coal from the Bulli seam at West Cliff colliery (New South Wales, Australia), with the axis of the cores at right angles to the bedding planes. The height of the samples varied between 50 and 120 mm. The ends of the samples were polished and sealed using epoxy and aluminium foil to prevent axial flow. Each sample was weighed and all samples were enclosed in high pressure steel cylinders (bomb). The bomb was evacuated for 48 hours and then methane gas pressure was applied to these samples. The methane gas pressure was kept constant over a 5-month period to ensure saturation of the cores. The bomb was then opened and cores were quickly transferred to a temperature-controlled bath. Gas desorbed from each of the cores was measured using the water displacement method. A general scheme of the system is shown in Figures 1-3.

Eleven samples were tested. The gas pressure applied was 7.5 MPa. Gas desorbed from each sample varied between 11 and 13 cm³/g. Desorption tests were carried out until almost no further gas was liberated from the coal cores. The period of desorption tests varied between 850 and 1 000 hours. A typical desorption curve is shown in Figure 5b. The data obtained from the desorption tests were used to determine the diffusivity coefficient, D , of the samples using the concepts developed previously.

It should be noted that a certain time elapsed between the opening of the bomb and the transfer of the core samples to the temperature-controlled bath. This time varied between 90 and 120 hours. The data have not been

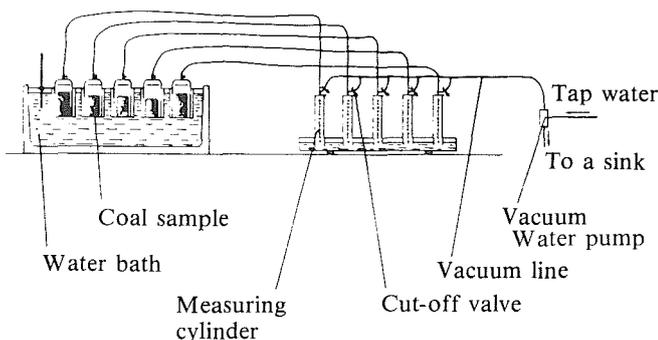


FIGURE 1. Schematic of desorption method

corrected for the gas lost during this period.

Permeability of the cores was also studied using radial techniques at varying pressure. The method used in these studies consisted of drilling a hole in the centre of the core and subjecting the coal sample to an atmosphere of gas, where gas permeates from the sides of the sample into the centrally driven hole. A general arrangement is

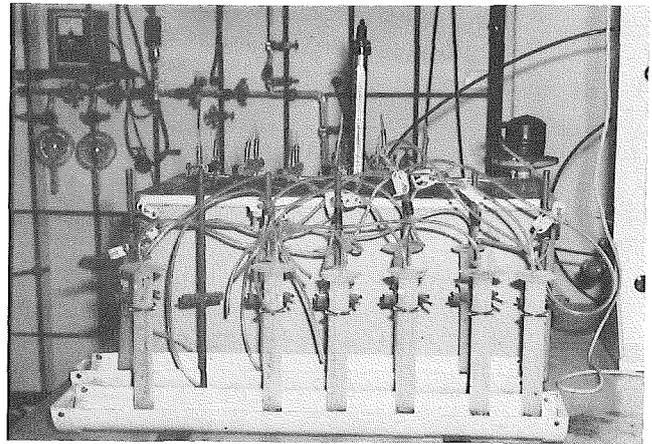


FIGURE 2. Desorption test equipment using water displacement method

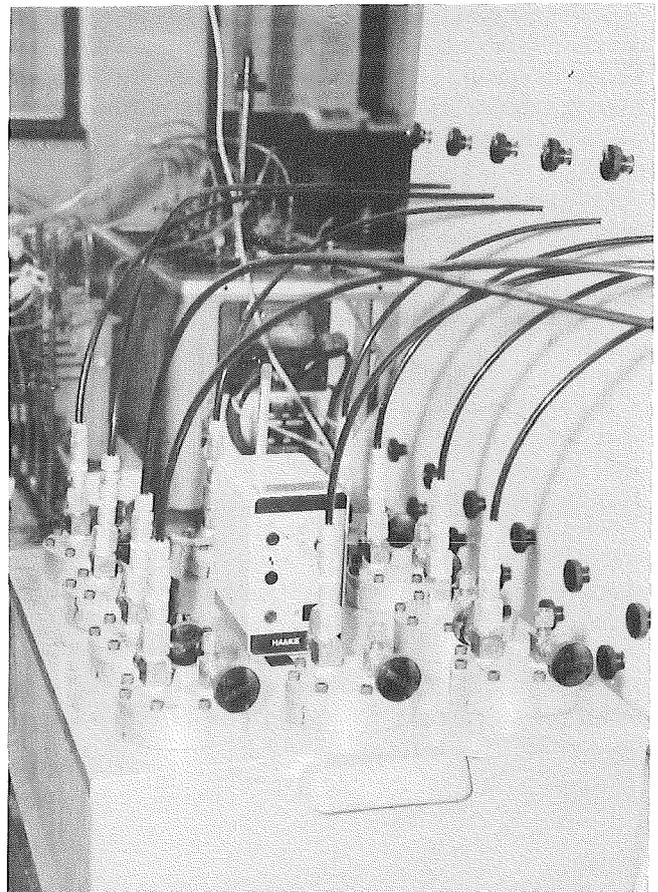


FIGURE 3. A series of bombs in a temperature-controlled water bath

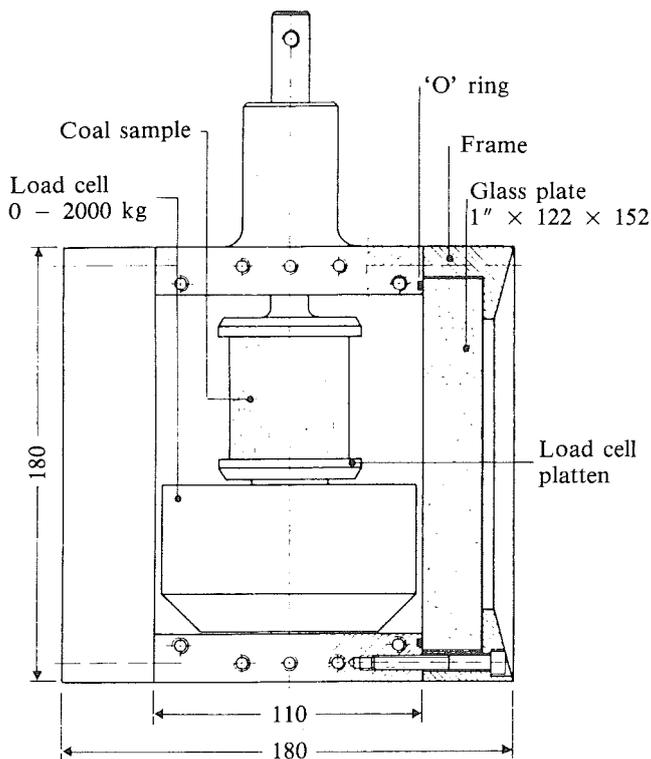


FIGURE 4. High pressure chamber with a coal sample for permeability test.

given in Figure 4. Both the gas pressure and the radius of the central hole drilled in the sample were varied.

Flow measurements were made using an electronic flow meter. Tests were conducted with varying axial stress. The results indicated that stress has a large influence on the gas permeability of coal samples. Permeability dropped by a factor of 2-3 with increase of stress from zero to 10 MPa (Fig. 7).

Results and discussion

The typical results of history matching using Equation [20] are presented in Figure 5a. Figure 5a shows that the analysis involving constant of diffusivity, D , and Crank's heat conduction solution does not result in a good fit of desorption data. The experimental curve has a more gentle change in slope and curvature, whereas the heat conduction solution has a sharp kink at around 85% of the gas depletion quantity. This observation is in fact well known to many researchers.¹²

The fact that the heat conduction solution shows a sharp kink relative to the data indicates that a constant diffusivity models a desorption rate faster than the actual rate recorded from tests. Alternatively, it can be said that gas desorption process tends to slow down with time to produce a gentle desorption curvature prior to an asymptotical total depletion of gas from a coal specimen. Therefore, the diffusivity is not a constant but varies proportionally with the local gas content or gas pressure. The total diffusivity can assume the form:

$$D = D_0 \left(\frac{P}{P_s} \right)^x \quad [21]$$

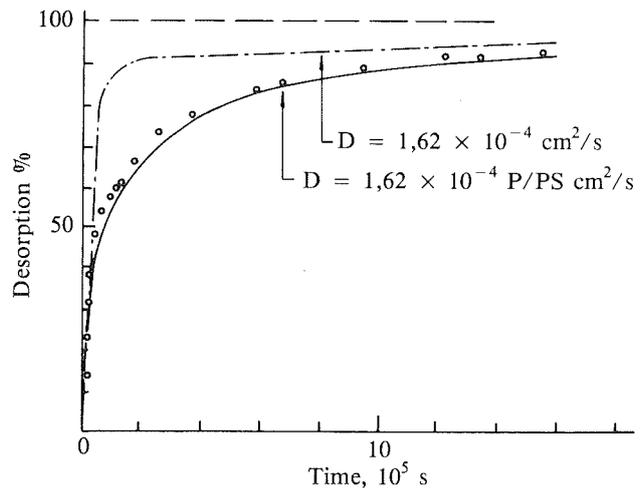


FIGURE 5a. History matching by Crank's analytical solution

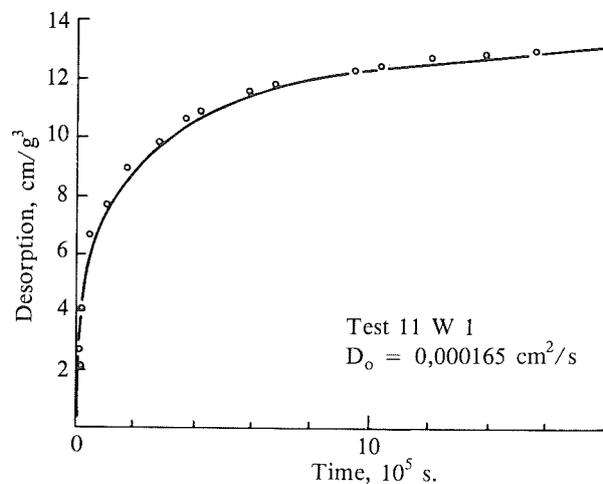


FIGURE 5b. Radial desorption of gas (CH_4) from coal cores, lab. tests and mathematical fit, $D = D_0(P/P_s)$

where D_0 is the diffusivity at the initial or seam gas pressure, P_s , and the exponent x has been set equal to unity for the sake of simplicity.

An optimisation search scheme was devised to back figure the value of D_0 from experimental desorption data. The numerical scheme is a simplified and single-parameter version of the simplex reflection technique, commonly used in geomechanics applications in recent years.²³ It has been found from this study that the one-parameter reflection method gives faster and more reliable convergence than the secant method in the search for an optimum (minimum). The reason for this is that the secant method when applied in the search for a non-zero functional minimum does not converge easily and can readily 'overflow' on the microcomputer. In the present instance, the optimal function is:

$$f = \sum_{i=1}^N [y(CN)_i - y_{Ei}]^2 \quad [22]$$

where

$y(CN)$ is the predicted desorption quantity obtained by the Crank-Nicholson scheme (solved by Gauss-Seidel procedure),

y_{Ei} is the experimentally determined desorption quantity, and

N is the total number of data points.

Figure 6 gives an illustration of the one-parameter reflection technique for the determination of D_0 to obtain a better fit of test results. History matching of test results shows that D_0 ranges from 0,00016 to 0,00017 cm²/sec, and indicates strongly the dependence of total diffusivity on gas pressure.

In methane transport modelling, empirical or semi-

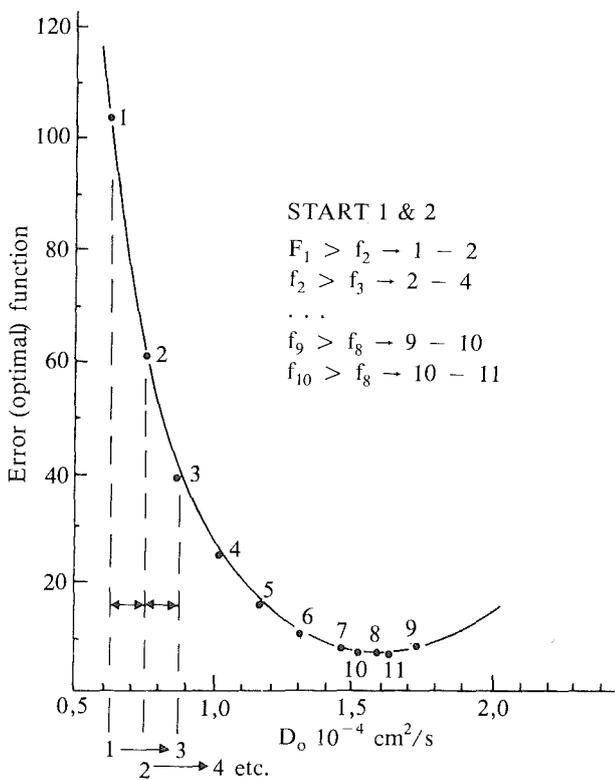


FIGURE 6. Principle of the reflection technique to determine D_0 by iterations

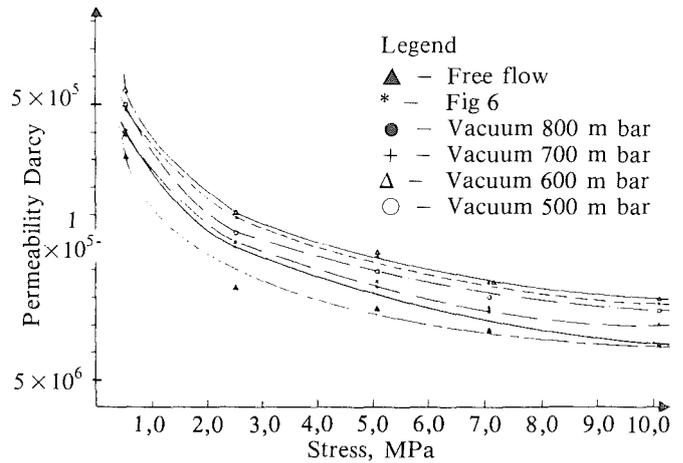


FIGURE 7. Effect of stress and vacuum on permeability of cylindrical coal samples, 0 50 mm L = 50 mm, sample 2 MB, West Cliff Colliery, gas CH₄, pressure 6 bar abs

empirical formulae are customarily used. Modelling of methane transport in real life, however, is often compounded by the presence of water and other gases (such as carbon dioxide) in the coal body, as well as the interaction between induced excavation stresses and gas permeation or diffusion properties.

The proposed model simplifies some of these aspects. The particular feature of this model is that it combines Darcy's laminar flow with Fick's diffusion and phase interchange taking place at any time during the flow of methane in the coal body. In this respect, the present model justifiably by-passes two common sources of disagreement between proponents of various popular simulators in the past. The first source is the choice between Darcy's law and Fick's law, and the second involves the concept of elemental blocks. In the 'elemental block' concept, methane transport in coal is considered as a combination of two processes in series, diffusion of methane from an elemental block of size 'a', followed by Darcy's flow in the macropore system.^{1,3,16,17,19} The propor-

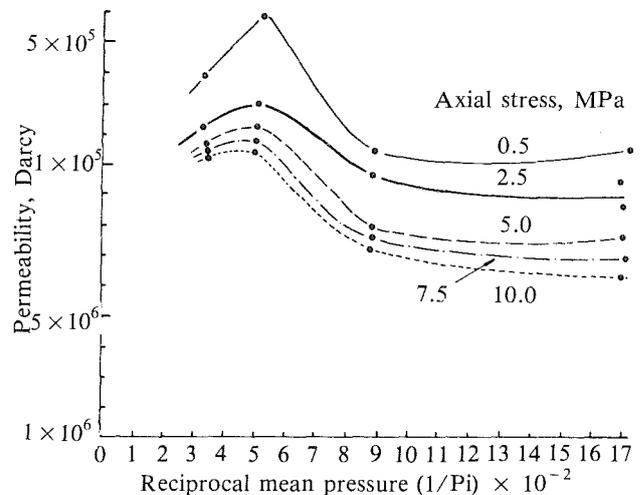


FIGURE 8. Permeability of coal sample (sample No. 2 - MB) as a function of reciprocal of mean abs. gas pressure (gas CH₄)

tionality constant (i.e. diffusion coefficient) provided by these models thus appears in the form of D/a^2 , resulting in disagreement in the choice of block size 'a', and rendering the models unattractive to modern computer modelling techniques.

The drop in the value of D with the drop in pressure is of fundamental interest. The results of laboratory tests indicated that at higher gas pressure, the Klinkenberg effect is no less dominant than the non-laminar flow. The results of the mathematical simulation showed this effect. Laboratory studies also indicated a drop in permeability with drop in gas pressure (Figure 8). The drop in K is by a factor of 2–4. The total diffusivity D determined in the desorption test is given by Equation [16]:

$$D = \frac{\rho_0 D_F \lambda + \rho K \mu^{-1}}{\lambda \rho_0 + \Phi_M \rho c} \quad [16]$$

where

$c = \frac{1}{\rho} \frac{\delta P}{\delta P}$, the gas compressibility factor ($= 1/P$) and

$$\lambda = \frac{AB}{(1 + BP)^2}$$

D is the total diffusivity (cm^2/s), accounting for Fick's diffusion, Darcy's flow, and phase interchange by desorption. In the laboratory tests used to determine permeability under stress and at mean gas pressure P , the conditions of flow are those of Darcy's flow, possibly without Fick's component and Langmuir's phase interchange. The total diffusivity D can thus be related to the equivalent permeability as:

$$D = \frac{K}{\Phi_M \mu c} = \frac{KP}{\Phi_M \mu} \quad [23]$$

Also

$$D = D_0 \left[\frac{P}{P_s} \right] \quad [24]$$

Equating [23] and [24] we get:

$$K = D_0 \frac{\Phi_M \mu}{P_s} \quad [25]$$

showing that the variation of K with gas pressure P is identical to that of D_0 with P . It is recalled that D_0 is the initial value of total diffusivity D being proportional to the gas pressure (Eq. [23]). The dependence of D_0 on P (or P_s), however, has not been investigated in the study reported here, due mainly to the long duration required to pressurise coal specimens and to conduct desorption tests. Permeability tests carried out on coal specimens subjected to varying axial stress has revealed that the permeability of methane gas through coal is directly proportional to the applied gas pressure P_s , and independent of stress change (see Table 1). The tests also showed that for a consistent value of P_s , the value of K determined is not influenced by the applied suction.

Let us now return to Equation [16], and its simplified form for calculating equivalent permeability (Eq. [25]). Substituting the following parameter values in [25]:

$$\mu = 1,087 \times 10^{-5} \text{ Ns/m}^2, \Phi_M = 0,01 \text{ 1 Darcy} = 10^{-8} \text{ cm}^2,$$

$P_s = 6500000 \text{ Pa}$, and $D_0 = 1,6 \times 10^{-4} \text{ cm}^2/\text{s}$ we obtain:

$K = 2,67 \times 10^{-9} \text{ Darcy}$ which is many orders of magnitude smaller than the permeability measured from permeability tests (Table 1).

TABLE 1
Permeability results

Gas pressure (bar)	Mean K (Darcy $\times 10^{-5}$)	K/K_0	P/P_s
6 (P_s)	0,8	1	1
11	0,9	1,07	1,83
19	2,78	3,30	3,17
29	2,26	2,69	4,83

Note: Permeability is average of all values determined at different stress levels

This apparent anomaly could be explained in terms of different flow mechanisms taking place in the two different tests. In desorption tests, diffusion is more predominant, whereas in permeability tests, Darcy's flow is the main mechanism and diffusive flow as well as Langmuir's phase interchange are completely suppressed.

Conclusion

History matching of diffusion test results has been made on a number of cylindrical coal specimens, and the study reveals the following:

1. The diffusivity of methane in coal under unstressed condition is directly proportional to local gas pressure. Curve fitting of desorption data using analytical formulation has to use total diffusivity as a direct function of local gas pressure.
2. Permeability of methane gas through coal as determined directly from permeability tests increases as the applied gas pressure increases.
3. The values of equivalent permeability, obtained from desorption test data, are smaller than those determined by permeability tests by many orders of magnitude.

There are, however, a few other points that must be considered. The Langmuir adsorption equation used in the present model does not fit the experimental data at high pressures. For methane it tends to give higher values, while for carbon dioxide it gives lower values.

It is also possible that this may also affect the history matching results.

Acknowledgements

The work described in this paper was partly sponsored by the National Energy Research Development and Demonstration Council (NERDDC) of the Federal Government of Australia under Research Grant No. 574 provided to Kembla Coal & Coke Ltd.

The authors also wish to place a record to the encouragement received from the management of Kembla Coal & Coke and for the permission to publish this work.

The provision of space and facilities for the fabrica-

tion of equipment and conduct of laboratory tests by the Department of Civil and Mining Engineering of the University of Wollongong is gratefully acknowledged. Thanks are also due to Mr W. Tomczyk who carried out laboratory tests very diligently.

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