METHANE DESORPTION CHARACTERISTICS OF SELECTED SOUTH AFRICAN COAL SEAMS.

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A dissertation submitted to the
Faculty of Engineering,
University of the Witwatersrand, Johannesburg,
in fulfilment of the requirement for the
degree of Master of Science in Engineering.

Johannesburg 1988

Declaration:

I declare that this dissertation is my own work. It is being submitted for the degree of Master of Science in Engineering at the University of the Witwatersrand, Johannesburg. It has not been submitted before for any degree or examination at any other University. All of the research was performed at the Mining Department of the University of the Witwatersrand.

E.G. BILLENKAMP

.....E.G.Billenkamp..... day of ...28.th...March.. 1988

SYNOPSIS

The hazard presented by methane gas to underground personnel is especially prominent in collieries. The purpose of this research is to quantify the magnitude of the problem in various South African collieries.

Methane sorption isotherms are constructed for temperatures of 10 , 20 , and 30 C at test pressures of 0,5; 1,0; 2,0; 3,0; 4,0; 6,0; 8,0; and 10,0 MPa. If mine personnel know what the seam temperature and gas pressure is, the methane content per ton can be read off from the isotherms. The samples used for the tests had proximate and Hardgrove Gridability analyses done so that correlation between these factors and methane content could be determined.

ACKNOWLEDGEMENTS

Professor H.R. Phillips for providing the impetus and initial design of the adsorption testing apparatus. Gunther Bakker for painstakingly building the equipment and workshop backup. The Chamber of Mines for accepting Professor Phillip's research proposals and providing the necessary funding. Also I thank Mrs Moore of the Geology library and Mr. D. Ralston of the U.S.B.M. Technology Transfer Group for providing me with the latest publications concerning methane in collieries. My most sincere appreciation goes to my brother, H.E.H. Billenkamp for helping me make my program more user friendly. Finally, I thank Mrs Miles, Mrs Anderson and Mrs Spann for help with the preparation of this text.

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NOMENCLATURE	Symbol
Butane	C_4H_{10}
Carbon dioxide	CO_2
Ethane	C_2H_6
Helium	He
Hydrogen	H_2
Methane	CH_4
Nitrogen	N_2
Oxygen	O_2
Pentane	C_5H_{12}
Propane	C_3H_8
Sulphur hexafluoride	SF_6
Atmospheres	Atm.

	Symbol
Hardgrove grindability index	HGI
Million cubic feet per day	Mcfd.
Percentage by weight	% wt.
Total desorbable methane content	Q_t
Federal Republic of Germany	FRG.
Mining Research and Development Establishment (U.K.)	MRDE
Republic of South Africa.	RJA
United Kingdom	UK
United Soviet Socialist Republic.	USSR
United States of America.	USA
United States Bureau of Mines.	USBM
$9.8687456 \times 10^{-13}$ m2	1 darcy

CHAPTER 1

INTRODUCTION

In many parts of the world coal winning is often inhibited when firedamp, a predominantly methane-rich gas under pressure within coal, is emitted in quantities which exceed the ability of the ventilation to dilute it to within statutory requirements. Methane release has been shown to be related to mining advance rate so the excavation rates attainable by modern excavation techniques may be limited. In extreme cases outbursts, which are violent eruptions of methane and powdered coal, cause mining operations to be halted.

The South African coal mining industry has been fortunate that outbursts have not yet been encountered and relatively few collieries experience severe gas problems. This is attributed to the shallow depth of which most mines operate. The extent of the methane problem in S.A. as well as overseas can be seen in TABLES 1 and 2 which list the principal explosions and are reason for concern.

As these shallow reserves become exhausted and we are forced to work deeper seams with their corresponding higher seam gas pressures, the gas problem will inevitably worsen. However, these problems can be overcome if accurate predictions of the degasification characteristics of the seam and surrounding strata can be made. This enables advance planning of adequate ventilation systems and if necessary, proper gas drainage schemes.

Table 1

Major Coal Mine Explosions in Southern Africa Resulting in the Loss of Ten or More Lives

Date	Mine	Fatalities
1901	New Campbell Colliery, Natal	31
1906	Elandslaagte Colliery Natal	18
1908	Glencoe Colliery, Natal	77
1926	Durban Navigation Colliery, No. 2 Pit, Natal	125
1928	Burnside Colliery, Natal	38
1935	New Marsfield Colliery, Tvl	78
1941	Utrecht Colliery, Natal	15
1943	Natal Navigation C & E Colliery, Natal	78
1944	Vryheid (N) RC & I, Hlobane Colliery, Natal	57
1945	S.A. Coal Estates Navigation Colliery, Transvaal	14
1951	Cornelia Colliery, OFS	26
1956	Schoongezicht Colliery, Tvl	12
1962	Northfield Colliery, Natal	10
1971	HCJ Contractors Colliery, Tvl	28
1972	Wankie No. Colliery, Zimbabwe	427
1974	Albion Colliery	13
1976	Moatize Colliery, Mocambique	99
1981	Newcastle Platberg Colliery	10
1982	Ermelo Mine Services Colliery, Transvaal	11
1983	Vryheid (N) RC & I, Hlobane Colliery, Natal	68
1985	Sasol, Middlebult Colliery, Tvl	30
1987	Ermelo	34

Table 2

Overseas Major Coal Mine Explosions Reported in the Media from January 1984 to August 1985

Date	Mine	Country	Fatalities
1984	Ariakne Mine Mitsui	Japan	83
1984	Strmoster Mine Rembas	Jugoslavia	35
1984	Haisahn Mine Juifang	Taiwan	74
1984	Santan Mine, Urassasga	Brazil	31
1984	Haisahn First Pit Mine, Sanhsia	Taiwan	93
1985	Simon de Forback Mine, Lorraine	France	22
1985	Takashima Mine, Mitsubishi	Japan	11
1985	Minami Oh Yubari Mine, Mitsubishi	Japan	62
1985	Meitian No. 3 Mine, Guangdong	PR China	55
1985	Nianziping Mine, Guangxi	PR China	21

The most significant factors in predicting gas release rates are the methane content and permeability of the coal seam and surrounding strata and the mining methods used. The aim of this study was to investigate the first of these factors and to take note of permeability results available from a parallel research programme.Two test rigs have been designed and commissioned. The first rig is used to develop equilibrium sorption isotherms for selected coal samples. These sorption isotherms are a recognised method for expressing the potential for that coal, to contain gas and providing seam temperature and seam gas pressure are known or can be estimated, then the gas content per tonne mined can be evaluated. A permeability test apparatus has also been designed and commissioned to test the permeability of coal seams and surrounding strata. Thus two of the factors related to gas release could be evaluated.

This dissertation contains a literature search, details of the apparatus used, test results and an analyses of results. It contains basic data, not previously available, regarding South African coals and concludes with a discussion of results and recommendations for future work.

2. METHANE IN COAL MINES - A LITERATURE REVIEW

2.1 The Origin of Gas in Coal

It has been estimated that during the formation of 1 ton of coal, up to 1300 m^3 of gas are produced (40). In the early stages of maturation most of the gas escapes through the relatively thin overlying strata and as a result, little gas is found in most low-rank coals. Higher rank coals are usually enclosed by deeper more competent rocks, so more of the gas formed at this later stage, is retained.

Gases, primarily, are a result of the maturation process and introduction of gas from the atmosphere. Thus the sequence of gas found, proceeding from the surface downward, is:

i) A zone of nitrogen (N_2) and carbon dioxide (CO_2),
ii) A zone mainly of nitrogen,
iii) A zone of nitrogen - methane (CH_4),
iv) Finally a zone where methane only occurs.

Carbon dioxide, and occasionally some other gases may come from other sources via faults and intrusions. Higher hydrocarbons such as ethane (C_2H_6), propane (C_3H_8), butane (C_4H_{10}), and pentane (C_5H_{12}) as well as oxygen (O_2), nitrogen, hydrogen (H_2) and helium (He) can occur in small quantities as shown in TABLES 3,4 and 5. (40).

TABLE 3 - Gas produced from plant material

Source	Cellulose	Lignin derivative	Wood
Total gas, ml	150	15	10
Total gas, ml/g substrate	100	10	6.6
Time, mo	3	20	8
Rate, 10^{-6} mole/d	9.9	2.5	4.4
CO_2, pct	5.01	5.00	5.01
CH_4, pct	94.94	94.98	94.99
Other hydrocarbons, pct	0.0496	0.0216	0.0005
CH_4/total hydrocarbons	0.999	0.999	0.999

TABLE 4 - Composition of gas from plant material, peat, and coal, pct

Source	CO_2	CH_4	Other hydrocarbons
Plant material	5.0	94.99	0.01
Peat	96.75	3.23	.02
Coal	1.9	97.8	.3

HYDROCARBON GAS FORMATION DURING DIAGENETIC COALIFICATION

TABLE 5 - Average composition of gas from coalbeds, pct

Coalbed	Pocahontas No. 3[1]	Pittsburgh[2]	Upper Kittanning[3]	B Seam[4]	Lower Hartshorne[5]	Mary Lee[5]	Anthracite[7]
CH_4	94.9	91.1	97.4	87.8	99.2	96.0	98.7
C_2H_6	1.3	.3	.01	.05	.01	.01	.1
C_3H_8	.004	ND	ND	.005	ND	ND	Trace
C_4H_{10}	.002	ND	ND	.001	ND	ND	ND
C_5H_{12}	Trace	ND	ND	ND	ND	ND	ND
CO_2	.3	8.2	.1	12.0	.1	.1	.1
O_2	.2	.2	.2	ND	.1	.1	ND
N_2	3.2	.5	2.3	.1	.6	3.5	1.0
H_2	.02	ND	ND	ND	ND	Trace	ND
He	.04	ND	ND	ND	ND	.3	Trace
Btu/scf	1,030	975	1,037	935	1,056	1,022	1,053

ND Not detected.
[1] Virginia.
[2] Pennsylvania and West Virginia.
[3] Pennsylvania.
[4] Colorado.
[5] Oklahoma.
[6] Alabama.

Commonly these gases in coal are referred to as firedamp. Methane is by far the dominant gas being emitted, so this report deals almost exclusively with the methane aspect of firedamp.

2.2 The Retention of Methane in Coal

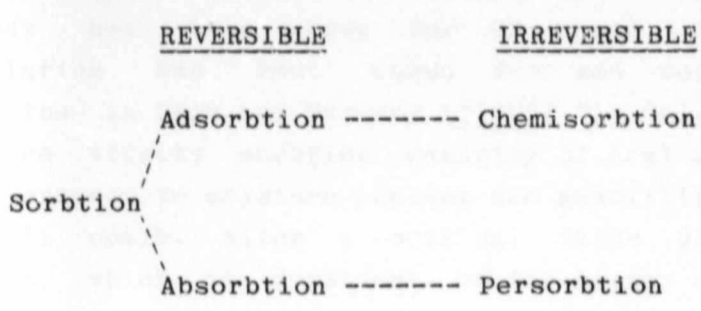
The process of methane retention is called sorbtion which is sub-classified into adsorbtion, absorbtion, persorbtion and chemisorbtion.

i) Adsorbtion is a surface effect whereby one substance is physically held onto the surface of another,

ii) Absorbtion is a more or less uniform penetration of one substance into the molecular structure of another,

iii) Persorbtion is a type of absorbtion of a gas by a solid in which there is the formation of a molecular mixture of the two substances,

iv) Chemisorbtion is a type of adsorbtion where one substance is held onto the surface of the other by chemical forces.



Chemisorbtion and persorbtion are not relevant in methane coal systems and absorbtion does not play a significant role in the flow of methane from coal during mining operations (14). Chemisorption does occur but it is such a powerful bond that evacuation at elevated temperatures is required to break the bond (27). This cannot happen during mining operations. When gas leaves coal it is said to be desorbed.

Methane exists in the adsorbed state or in the free gas state in pores and fractures (FIGURE 1). At low pressures these two states both occur in moderate proportions and the volume of gas contained increases almost in direct proportion to pressure. In the 0,1 - 4 MPa range, 54 - 85% is in the adsorbed state (64). By about 5 MPa almost all gas contained is in the adsorbed state and at higher pressure the rate of gas intake decreases, presumably as monolayer adsorbtion is complete. At about 7 - 10 MPa there is only a slight increase in gas content and some but not all studies have shown a decrease in sorbtion above these pressures. At 10 MPa adsorbed methane can be as densely packed as a liquid (64).

Adsorbative capacity of coal increases with pressure and rank but decreases with temperature and moisture content (1,23,38,41,54,112) (FIGURES 2,3,4,5 and 6). A correlation for fixed carbon, volatiles as well as oxygen content (which is related to rank), and gas content has been shown for US coals. A similar correlation has been shown for gas content and volatiles in USSR and Germany (FIGURE 7). Only adsorbed moisture affects sorbtion capacity of coal and curves with respect to moisture content are generally the same for all coals. After a critical value of moisture content, which is dependent on the oxygen content of the coal, is reached, no further reduction of methane

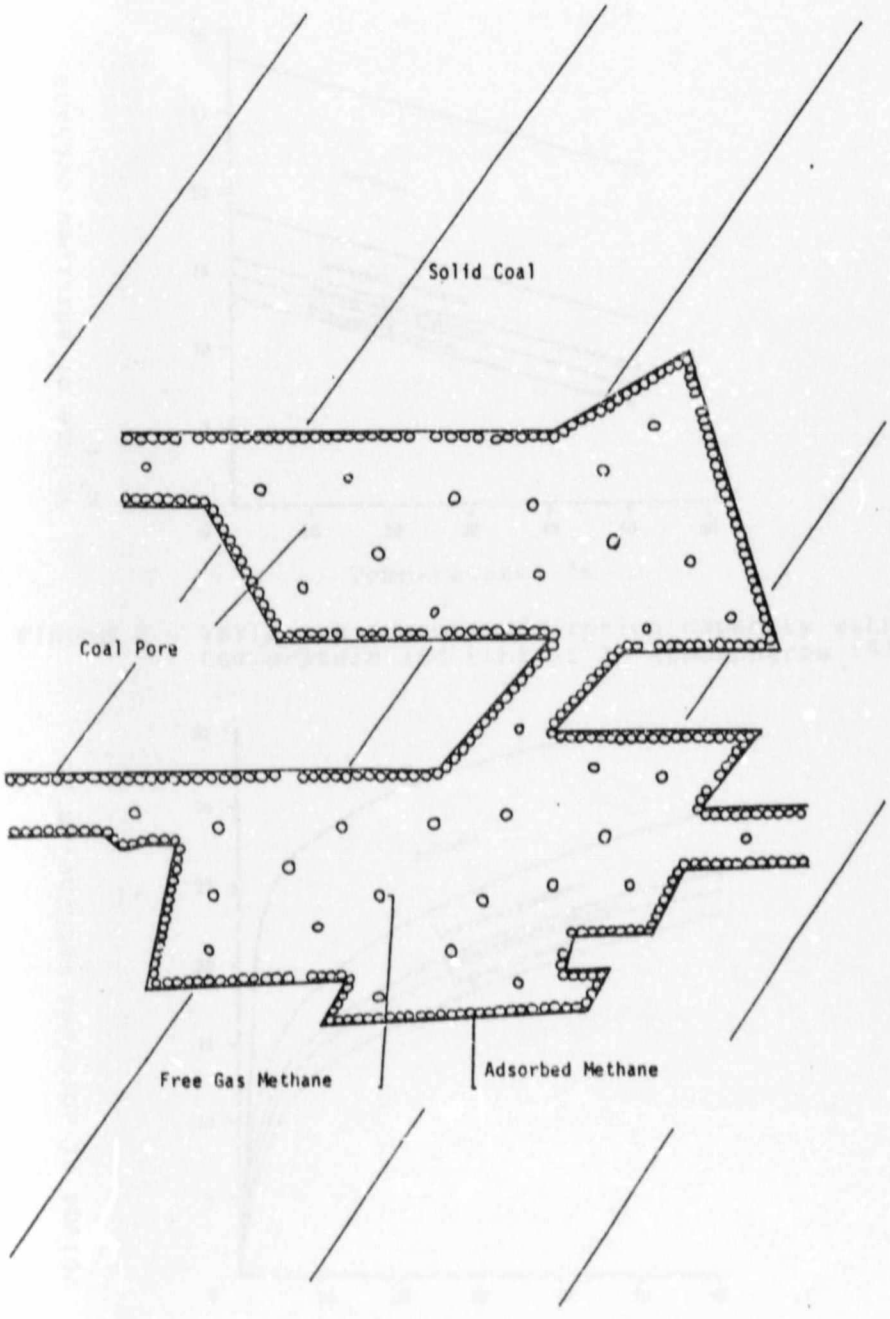


FIGURE 1 Pictorial Representation of Methane in a Coal Pore (54).

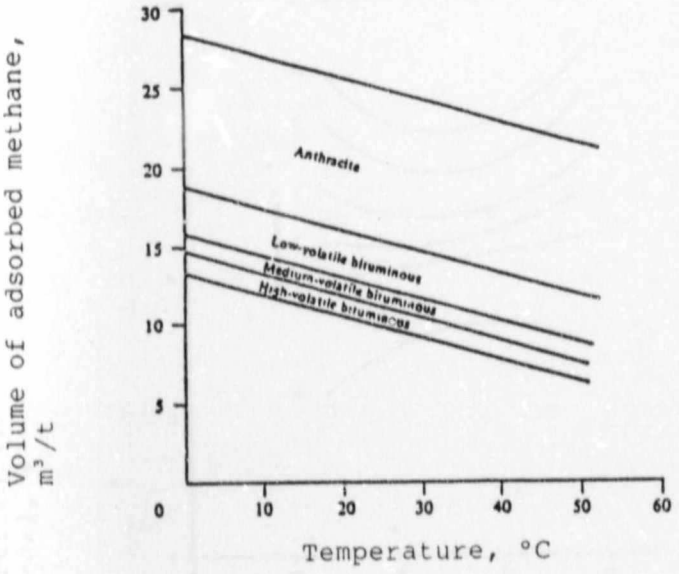


FIGURE 2 Variation of coal adsorption capacity with temperature and rank at 10 atmospheres (41).

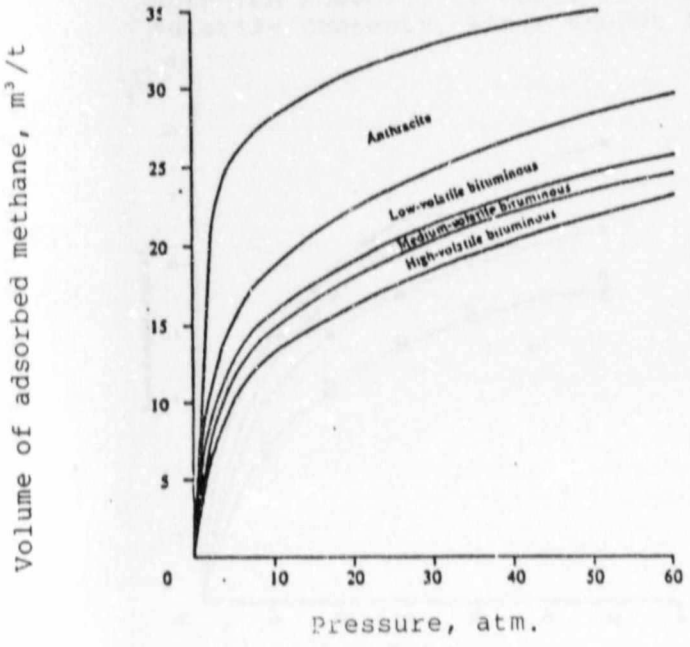


FIGURE 3 Variation of methane adsorption isotherm with coal rank at 0°C (40, 41).

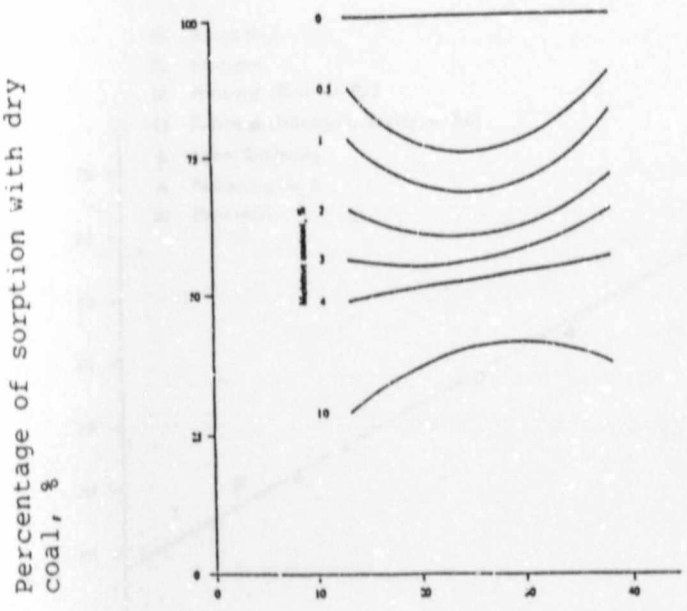


FIGURE 4 The effect of moisture content on the sorption capacity of coals of various volatile contents. After Khodot (14,58)

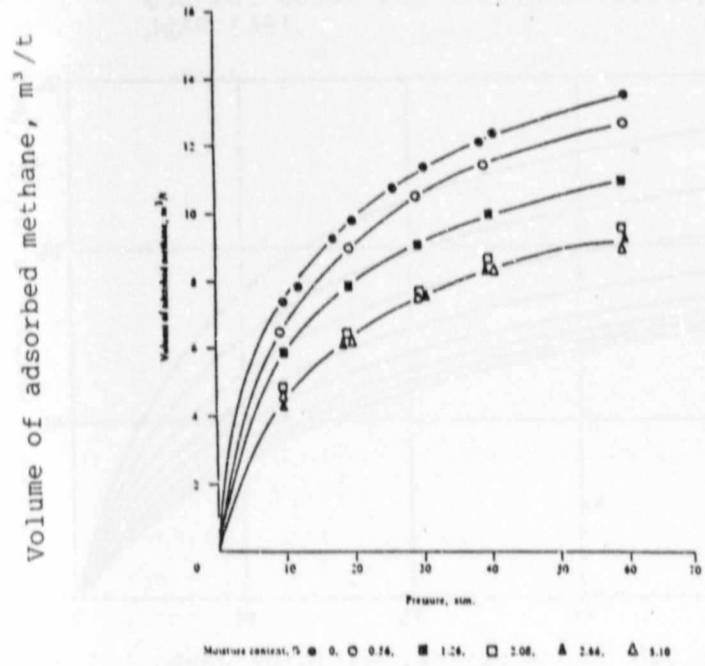


FIGURE 5 Methane adsorption isotherms of Pittsburgh II coal at 30°C at various moisture contents (38).

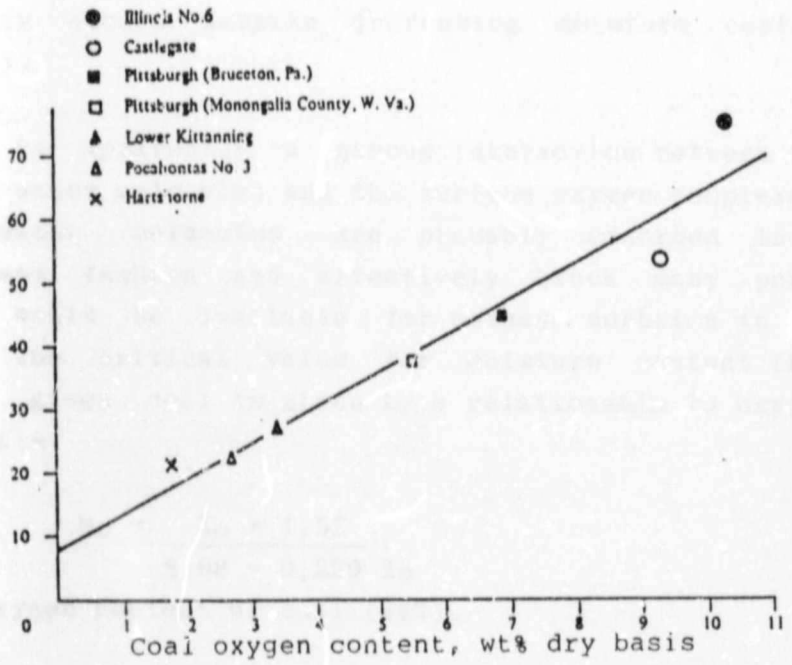


FIGURE 6 Maximum reduction in methane sorption versus coal oxygen content at values of moisture content above the critical values, 10 atm., 30°C (39).

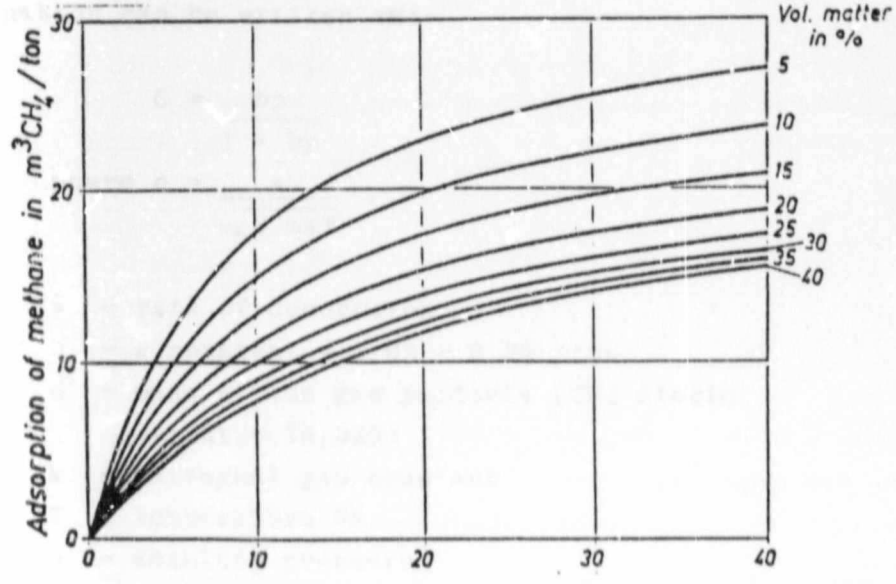


FIGURE 7 Mean sorption isotherms for dry coals with 5-40% volatile matter (Temp. 30°C) (62).

capacity occurs despite increasing moisture content (14,28).

There is apparently a strong interaction between the polar water molecules and the surface oxygen complexes. The water molecules are probably adsorbed in a localised fashion and effectively block many pores which would be available for methane sorbtion in dry coal. The critical value for moisture content (M_c) for a given coal is given by a relationship to oxygen content:-

$$M_c = \frac{X_o + 1,51}{3,98 - 0,229\ X_o}$$

X_o - oxygen content of coal (wt%).

Of the models used to describe adsorbtion onto coal, Langmuir's model is more commonly favoured over the Polyani, Temkin and Freundlich models. Langmuir's equation can be written as:-

$$G = \frac{bp}{1 + bp}$$

$$\text{where } b = \frac{a}{v\sqrt{2\ mkT}}$$

G - rate of desorbtion
b - a constant = 0,03 - 0,23 /ton
m - mass of the gas particle (CH_4 atomic weight = 16,043)
k - universal gas constant
T - temperature ^oK
p - absolute pressure
a - a constant $0 < a < 1$

This equation relates the quantity of gas adsorbed per unit mass of solid to the partial vapour pressure of

the gas, and has been shown to be suitable up to 15 MPa (11).

The Brunauer, Emmett and Telle (BET) equation is an extension of Langmuir's equation in that it considers multilayer adsorbtion. However, a second layer does not become apparent at pressures less than 5 MPa, while pressures greater than 4 MPa are not common in coal mining.

The occurance of methane has been assessed for US bituminous coals and is found to seldom exceed 55 m^3/t even at very high pressures (FIGURE 8).

2.3 Determination of In-Situ Methane Content

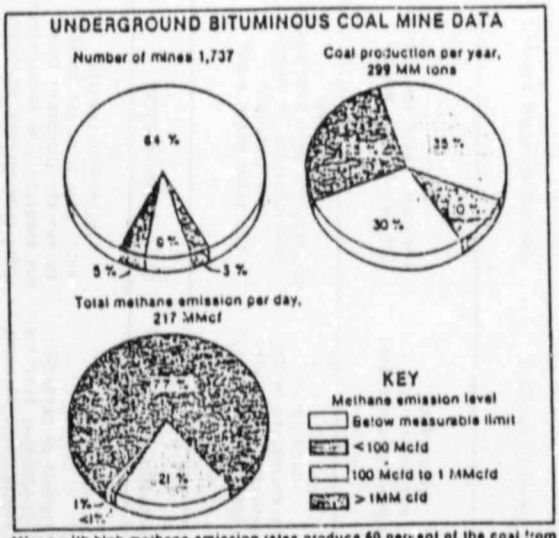
Methods for determining methane content of coal are broadly classified as direct or indirect. The direct method measures gas content of a sample in a laboratory or field experiment. The indirect method calculates methane content from gas pressure and use of the relevant adsorbtion isotherm for the coal in question. (TABLE 6).

2.3.1 Direct Methods

Gas content is measured using either:-

i) A desorbtion meter (FIGURE 9) (5). Here the size of flask is important as pressure rise must not be large enough to impede desorbtion.

   For 10 g of chippings sample, V flask = 1 litre.



Mines with high methane emission rates produce 60 percent of the coal from U.S. underground bituminous coal mines. (72)

FIGURE 8

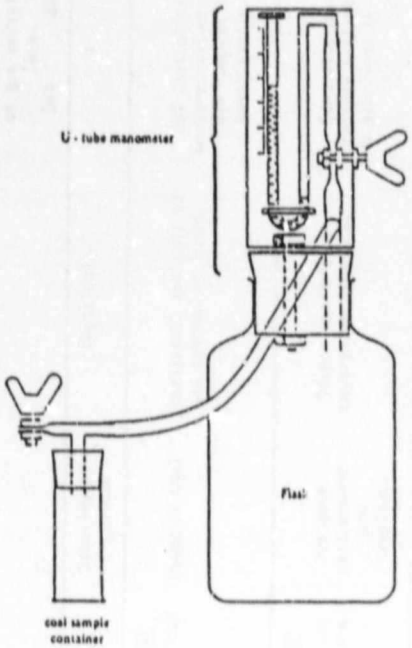


FIGURE 9 A desorption meter (5)

Table 6 METHODS OF ESTIMATION OF GAS CONTENT OF COAL SEAMS

METHOD	REQUIREMENTS	METHODS OF INTERPRETATION	ACCURACY	COUNTRY	METHOD OF MEASUREMENT	APPLICABILITY
DIRECT	Cores	Requires knowledge of gas emission laws	Depending upon time lost, core quality and applicability of gas emission laws. 50% 20%	U.K. U.S.A. Australia	Volumetric gas emission at atmospheric pressure followed by crushing.	Good from surface boreholes and doubtful from underground boreholes, fractured cores, etc. Degree of fracturing influences results.
DIRECT	Samoling of fractions	Empirical	?	Poland	Volumetric gas emission	Existing mines
INDIRECT Statistical	Lumps of coal	Statistical analysis of gas content of lumps	20% Statistically measured values from face samoles are consistently.	U.K.	Sample (30-40 mm size), collection at the face and estimation of gas contents.	For seams under ming, face samoling, applicability to highly variable and high rank coals not proved.
INDIRECT Adsorption Isotherms	Pressure measurement and sampling.	Direct reading for adsorption isotherms	Depending upon pressure measurements and sampling. 10 - 20%	Poland USSR Australia Germany	Volumetric techniques and gravimetric techniques.	For seams under mining and seams lying [illegible] or below.
INDIRECT Chemical Analysis	Proximate analysis of	Empirical	30 - 100%	USSR U.S.A. Poland	Chemical analysis	New and existing mines.
INDIRECT	Exhaust ventillation sampling.	Subtraction techniques	20%	Australia	CH_4 or CO_2 gas analysis (%)	Existing mines
INDIRECT	Sampling of exhaust mine air	Empirical	50%	Germany U.S.A.	Volumetric gas emission	Existing mines - general make of [illegible].

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INDIRECT	Sampling of exhaust mine air	Empirical	50%	Germany U.S.A.	Volumetric gas emission	Existing mines - general make of gas.

For a solid core a minimum of 100 g is required but preferably 1 - 2 Kg should be used.

ii) A test tube and level meter (FIGURE 10) (5). h_0 is the level read immediately after the coal has been introduced into the upper chamber and h_1 is the final level.

iii) Inverted cylinder (FIGURE 11) (4,5,16,18).

iv) Gravimetric procedures (15,47,57). Here the sample of coal is placed in a pre-cleaned and pre-weighed container and allowed to desorb whilst the weight is being measured. The change in weight is proportional to the amount of gas desorbed.

The site of sampling is important so as to be representative of the coal seam being studied with due consideration for over- and under-worked seams. The sample must preferably come from drill chipping or a core from outside the zone of emission, or if not, then at least from an active coal face which has not been in contact with air for more than 48 hours. However, to use such face coal requires chipping out coal for 2 - 6 m into the face so this is usually not practical. The most recent models of this zone of emission have been developed by CERCHAR (FIGURES 12,13,14,15,16,17 and 18.)

a) A certain amount of gas is lost 'Q_1' during the time from when drilling intersects the seam until it is enclosed in a sealed container. If drill flushing is by air or mist, time 't' starts when drilling intersects the seam, if by water then 't' starts at half the time taken to remove the core (as it is about then that the hydrostatic head approximately equals seam gas pressure). There are

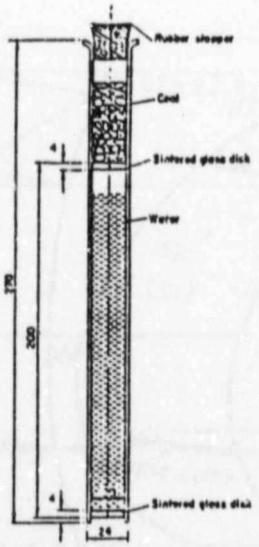


FIGURE 10 Glass test tube for determination of Q2 (5).

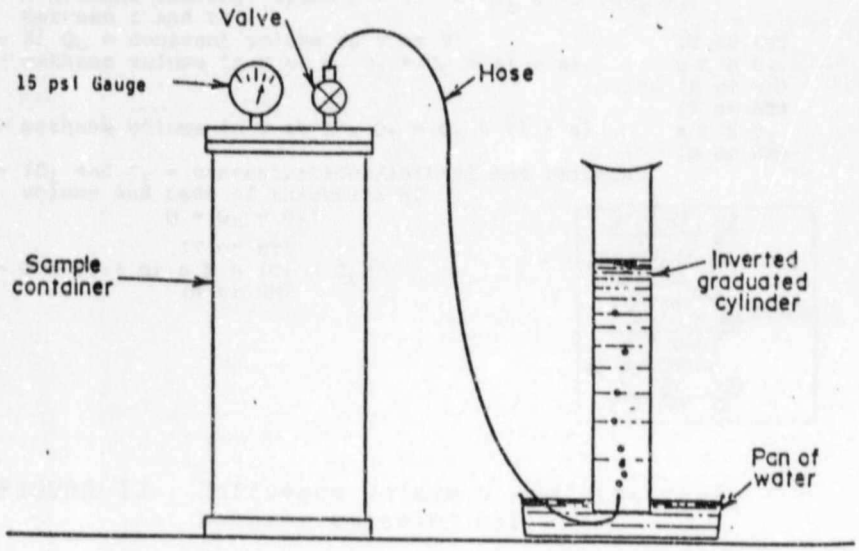
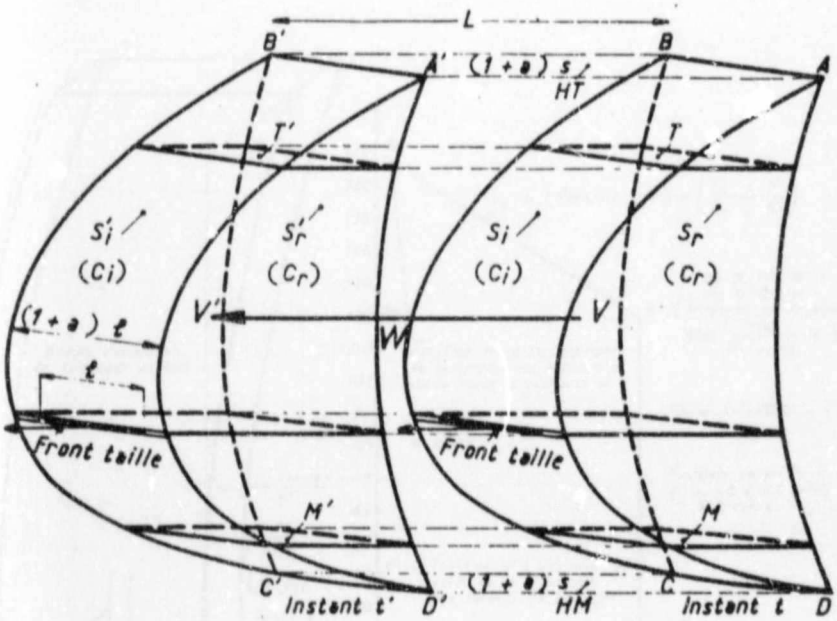


FIGURE 11 Sample container. Gas emission is measured by displacement of water (45).



- hypothesis V' = V (form, volume, concentrations) by migration L
- surface swept by face between t and t' = s = ℓ L
- methane emission Q between moments t and t' = methane leaving volume W = (A' B' S_i C' D' CD_r S_r) between t and t'
- Si Q_o = constant volume in V or V' (T or HT) s Σ h C_i (H or HM)
- methane volume in W at t, $Q_t = Q_o + (1 + a)$ (T or HT) s Σ h C_r (M or HM)
- methane volume in W at t', $Q_t = Q_o + (1 + a)$
- (C_i and C_r = concentrations/initial and residual volume and beds of thickness h)

$$Q = Q_t = Q_{t'}$$

$$Q = (1 + a)\ s \sum_{(M \text{ or } HM)}^{(T \text{ or } HT)} h\ (C_i = C_r)$$

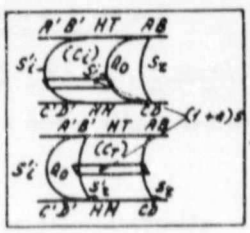


FIGURE 12 Influence volume V and stationary methane emission (31)

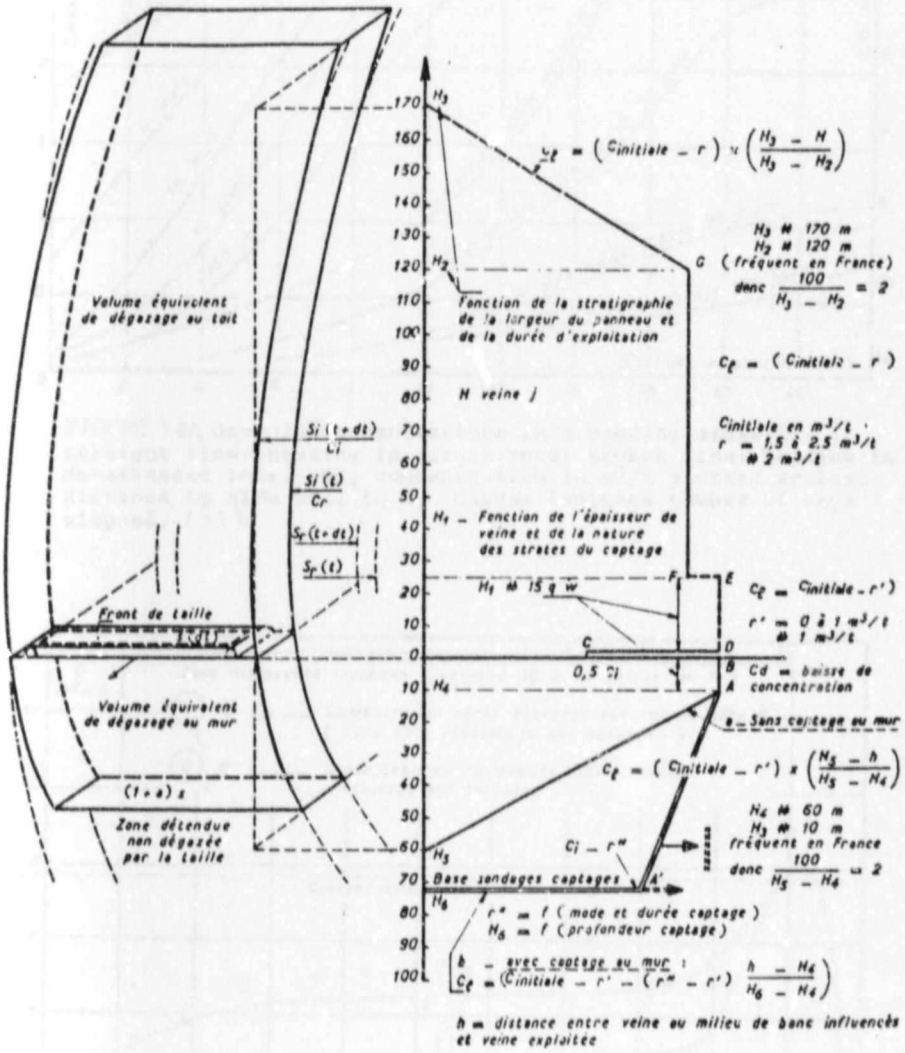


FIGURE 13 Equivalent emission volume (left); emission intensity. Left hand diagram (from top): equivalent emission volume in roof; coalface; equivalent emission volume in floor; de-stressed zone not drained by face. Right hand d'agram: Cá = drop in concentration; a = without floor drainage; r" = f (type and duration of drainage); H_6 = f (depth of drainage); b = with floor drainage; h = distance between seam or affected beds and worked seam. (31)

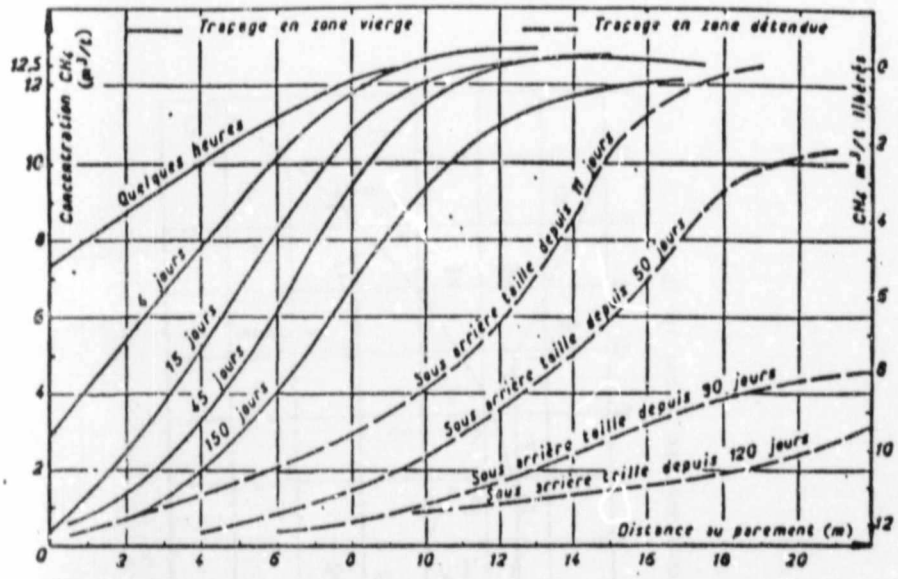


FIGURE 14A Drop in concentrations in a heading sidewall. Straight line: heading in virgin zone; broken line: heading in de-stressed zone. CH_4 concentration in m^3/t plotted against distance to side wall in m. Curves indicate number of days elapsed. (31)

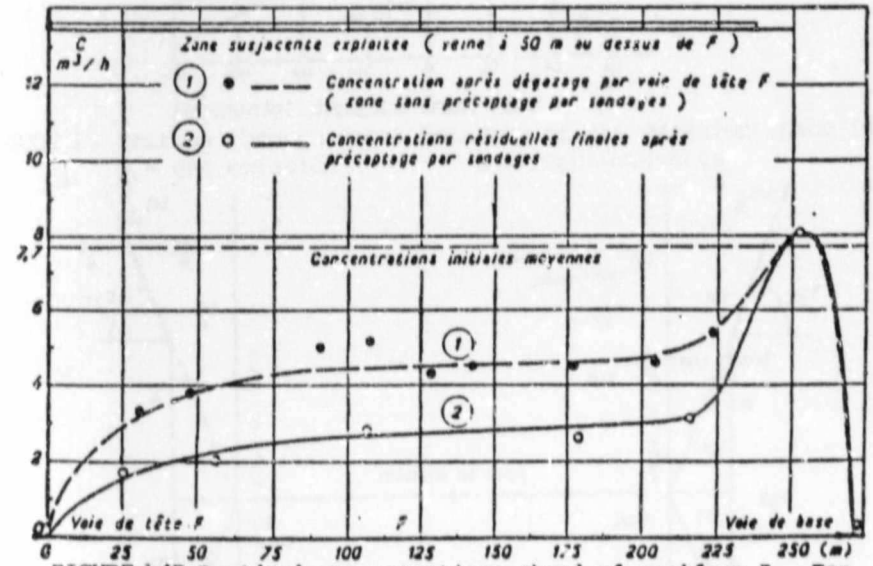


FIGURE 14B Residual concentrations ahead of coalface B. Top line: overlying zone worked out (seam 50 m above F); (1) concentration after drainage by top road F (zone without pre-drainage by boreholes; (2) final residual concentrations after pre-drainage by boreholes. Concentration in m^3/h plotted against face length from top road to bottom road. (31)

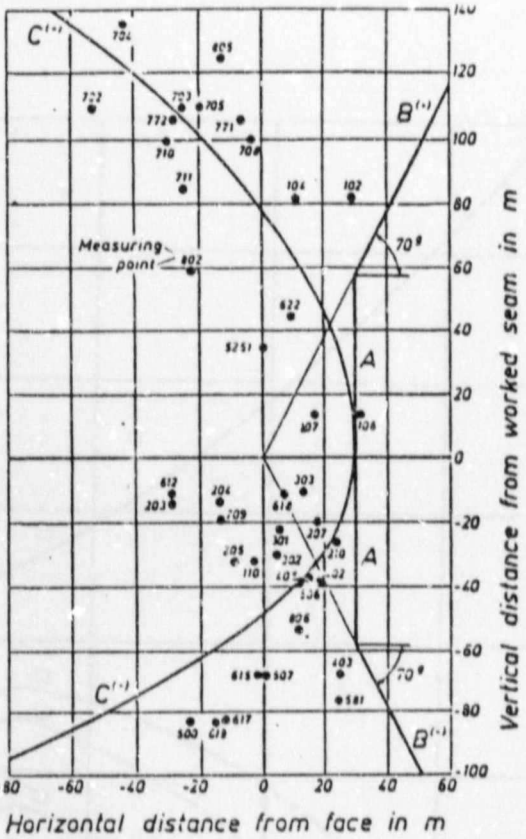


FIGURE 15 Section through front boundary of gas emission space (62)
C = gas emission B = rock movements

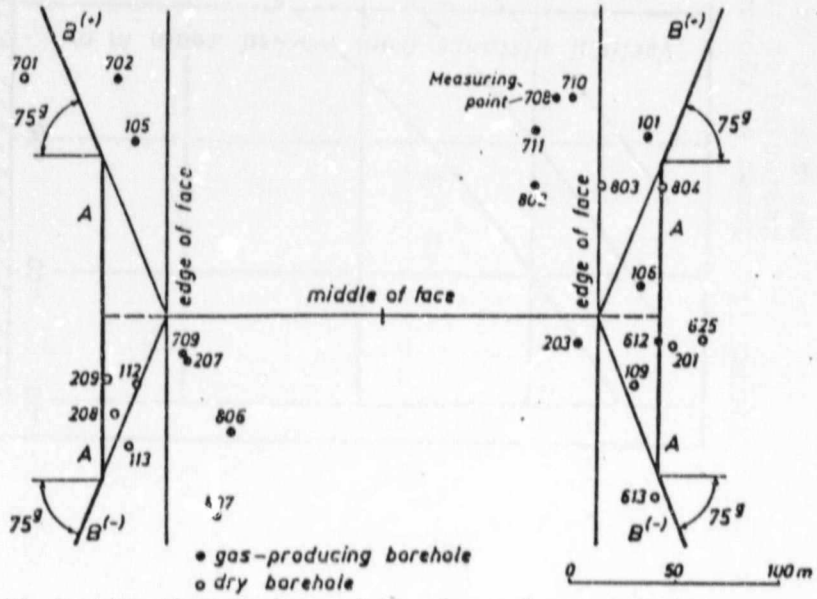


FIGURE 16 Section through lateral boundary of gas emission space (62)

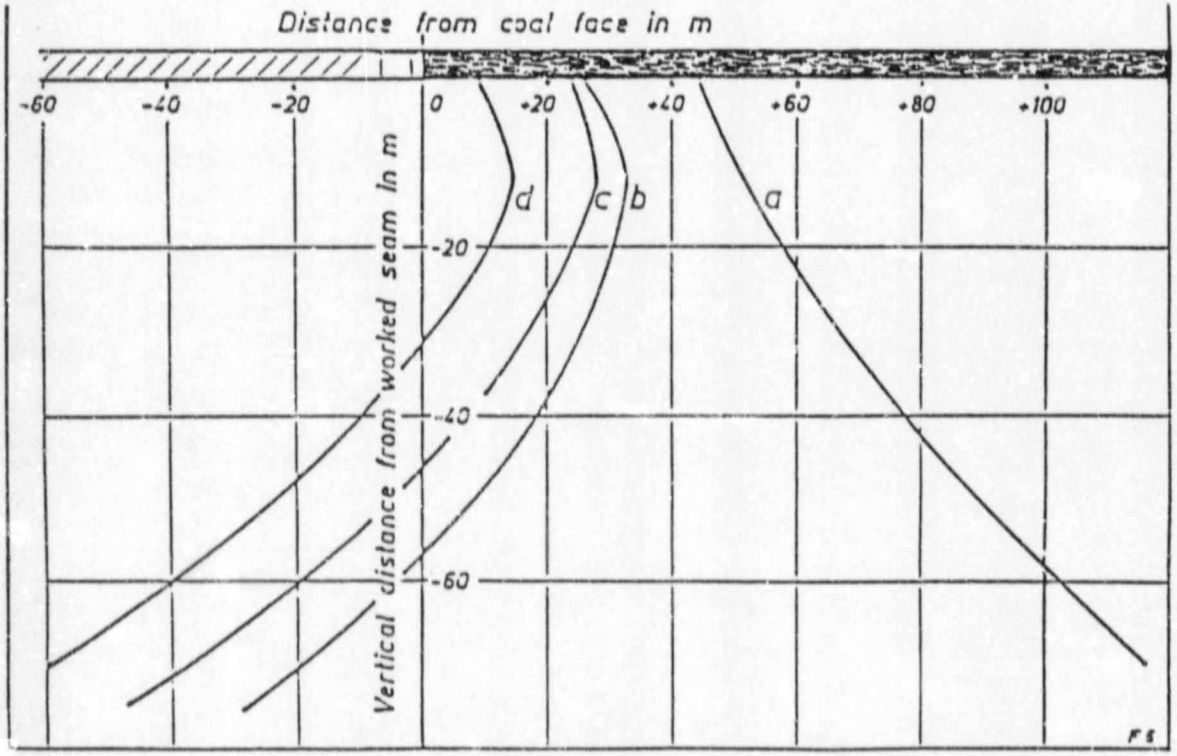


FIGURE 17 Zones of typical gas pressure in the floor of workings. Beginning of increase (a), beginning of maximum (b), end of maximum (c) of gas pressure and beginning of gas emmission (d). (62).

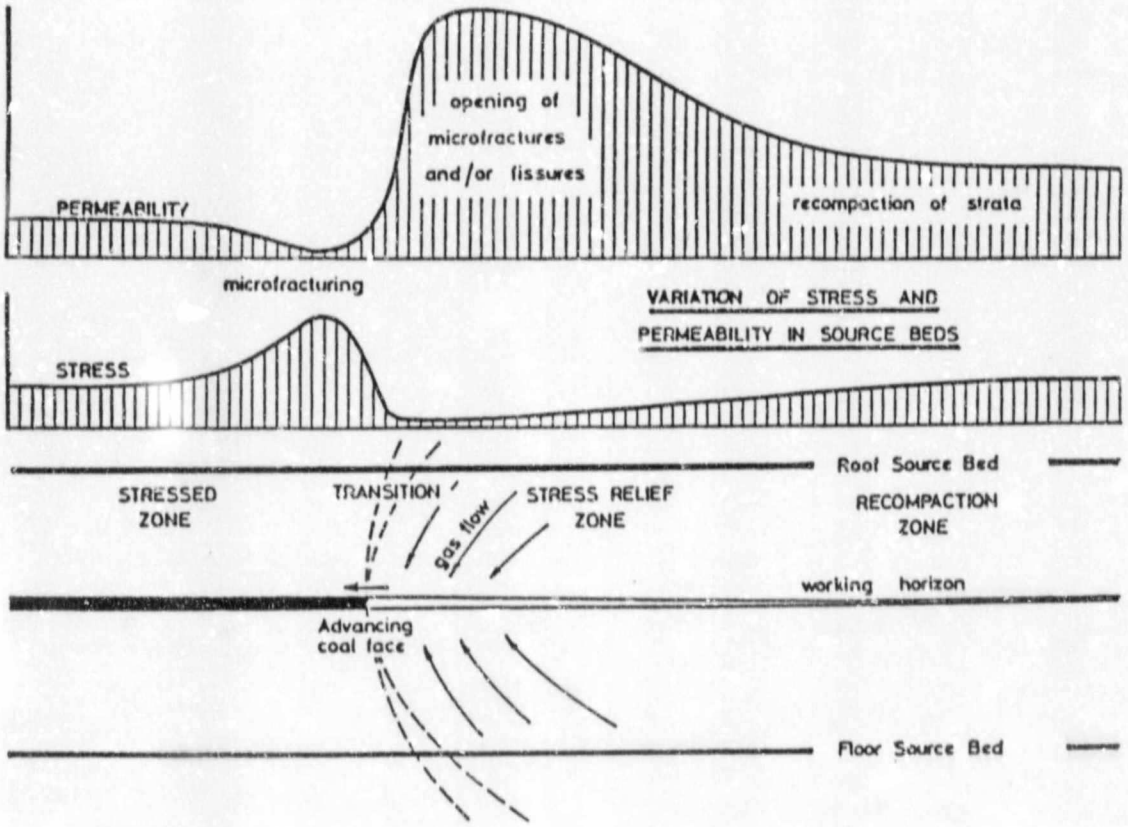


FIG. 18 — The variations of stress and permeability in strata above and below an advancing longwall coal face. (54)