

Measurement of coalbed methane isotherms for Australian coals

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ABSTRACT

Australia has large reserves of coalbed methane. In developing these reserves, measurement of adsorption data of methane on coal produces an isotherm that gives the gas storage capacity (volume) as a function of pressure. The gas storage capacity is a function of pressure, temperature, coal rank and coal moisture content. The isotherm enables estimates to be made of the volume of methane liberated from the reservoir as the pressure is reduced during production, and the maximum gas recovery between the critical desorption pressure and the well pressure at shutdown. CSIRO at Lucas Heights has constructed a facility to measure these data with a pressure-volume method. Each point on the isotherm usually takes 3 to 6 hours, coal dependent, so the apparatus is fully automated, with computer controlled addition of methane, pressure measurement and data acquisition. The system and its operation are described.

INTRODUCTION

An increasing Australian interest¹ has developed in the extraction of in-situ methane gas from coal seams where it has remained stored since its formation, adsorbed within the coal under pressure. The attraction of this potential energy source is based on its probable low cost, widespread abundance in Australia's major coal basins, proximity to population centres or existing pipelines and its benefits as a fuel, being clean burning with a high energy output per unit of carbon.

CSIRO Division of Coal and Energy Technology operates research programs on Australia's coals and alternative fuels. An opportunity arose within the alternative fuels program to develop a facility to measure the adsorption isotherms of methane on coals, with combined application to both research and

commercial clients. This grew out of our studies of natural gas storage on high surface area carbons, coupled with significant high pressure gas expertise and the capability of using advanced computer programming techniques to control all facets of the experiments.

EXPERIMENTAL

Equipment

The system, which is based on the paper by Mavor, *et al.*,² and the current recommendations of the GRI, is illustrated in Figure 1, and is capable of analysing four samples simultaneously at the same isothermal temperature. As explained more fully below, this high pressure gas system operates on a pressure-volume method, requiring knowledge of calibrated cell volumes and accurate pressure measurements: successful operation requires it to be leaktight. The following design features were incorporated for both utility and safety. All components are commercial stainless steel high pressure gas fittings, connected with 1/4" diameter, thick-walled, seamless stainless steel tubing, and are used below their service rating which is above maximum gas bottle pressure. There is a pressure relief valve in the system. Metal frit filters prevent coal being expelled from the sample cylinders into the system on pressure release. A feature of the design is that the reference cell volume of 320 cm³ is large compared to the sample cell void volume, ~ 80cm³, to minimise errors attributable to the methane compressibility factor, z , and to enable easy attainment of the maximum methane pressures required. Sample and reference cells are immersed in a 90 litre water bath thermostatted at $30.00 \pm 0.02^\circ\text{C}$, with an over-temperature trip.

System calibration, methane filling and venting operations, and data acquisition are computer controlled to allow un-manned 24 hour operation, because of the long duration of the experiments. Experimental control is centred around an 80386SX IBM compatible computer, with 4 Mbyte RAM, 90 Mbyte hard disk, math co-processor, and various accessories. The program which controls the operation of the system was written 'in-house' in Turbo-Pascal, V 6.0.

Pressure measurements are made with high precision variable capacitance pressure transducers connected to each cell. The transducers provide a 2-wire, 4-20 mA output to process meters which display the pressure and transmit data via a 4-20 mA current loop to a serial converter which allows the five transducers to be accessed from one computer serial port. Valve switching is achieved with a commercial interface card, with 16 TTL compatible output channels, operating solenoid valves via solid state relays. The solenoid valves, equipped with manual override, control the flow of compressed air to remotely operate the main valves, which are air-actuated 2-position, 2-way ball valves. This has the advantages that electrical contacts are removed from the vicinity of the rig and valves go to the 'normally closed' position should power failure occur. The two gas inlet valves and the vent valve have an associated metering valve to control the gas flow rate. The helium and methane gas supply cylinders are located remotely and the gases piped in. Gas released in normal operation or from the pressure relief valve is vented remotely. The system is leak tested with helium before each experiment but should a gas release occur, a methane detector prevents the accumulation of an explosive concentration and provides automatic shutdown of the equipment and gas supply, with appropriate warnings to the operator.

System Operation.

A weighed sample of up to 100 g of powdered coal is placed in a sample cell. The system is evacuated then leak tested with helium to 12 MPa. A series of helium expansions from the previously calibrated volume of the reference cell into the sample cell allows the void volume to be calculated. The system is depressurised and evacuated. With

the sample valve shut, the initial sample cell pressure is recorded. The reference cell pressure is increased to 0.5 MPa with methane and closed. The sample valve is opened to pressurise the sample cell with methane and then closed after 3 seconds. Within the sample cell, the pressure decreases due to methane adsorption onto the coal and this pressure is recorded until equilibrium is reached. Equilibrium is defined as a steady pressure reading ($\Delta P = 0.000$ MPa) for a 40 minute period. A typical adsorption step is illustrated in Figure 2. The reference pressure is further increased and the cycle repeated over 8 to 10 pressure points to required maximum pressure (up to 15 MPa) to characterise the isotherm.

A sub-sample for proximate analysis for moisture and ash, by thermogravimetry of the coal sample used in measuring the isotherm, enables calculation of the gas content on a dry ash-free basis. The surface area characteristics and pore size distribution of the sample, which are expected to correlate with the gas capacity of the coal, are measured using CO₂ porosimetry.

Calculation of Methane Adsorbed

Calculation of the gas adsorbed is based on the gas equation:

$$PV = nRTz$$

and the following equation can be derived for the volume of gas adsorbed at each pressure step per mass of dry ash-free coal:

$$V_{\text{ads}} = (T_0 / (T_{\text{iso}} P_0 m_c)) (V_r (P_r / z) - V_c (P_s / z - P_{s_i} / z))$$

cm³ g⁻¹ (or m³ tonne⁻¹) at STP, where the isothermal temperature is 30°C and where the variables and constants for both equations are described in Table 1. The volumes of gas adsorbed at each pressure step are added to provide the cumulative gas volume adsorbed at each cell equilibrium pressure and are then plotted to provide the isotherm.

RESULTS AND DISCUSSION

Results for a sample of coal from the Bulli seam from Tower mine in New South Wales are listed in Table 2 and the isotherm is shown in Figure 3 which gives the gas storage

capacity (volume) as a function of pressure. The information available from the isotherm is illustrated by the following example. For gas desorption to take place within a well, the pressure must be at or below the critical desorption pressure², which is the pressure on the isotherm corresponding to the gas content present in the seam. This can be estimated from desorption canister data (e.g. a gas content of 15.6 m³ tonne⁻¹ would give a critical desorption pressure of 6.8 MPa, Figure 3.). In the initial stages of production when the downwell pressure exceeds 6.8 MPa, water and gas from solution in water will leave the seams to be pumped to the surface. Once the pressure drops below the critical desorption pressure the methane can desorb, and if the permeability of the deposit is high enough, will flow through the cleat system to the well for extraction. The residual gas content after production depends on the well pressure at shutdown but if this pressure can be reduced to 0.7 MPa, this corresponds to a residual gas content of 6.93 m³ tonne⁻¹. The difference between the two gas contents reflects the recoverable gas given ideal conditions, and is for this example 8.7 m³ tonne⁻¹ coal in place, representing 56% of the gas in place.

These calculations are made on a dry ash-free basis. The ash and water content for the seam or seams of interest must be taken into account when calculating recoverable gas. Although a proximate analysis is made on the sample used to obtain the isotherm, this is not necessarily representative of the downwell condition. More extensive modelling of the isotherm to the Langmuir equation is possible² but will not be undertaken here. Other features that can be studied using adsorption isotherms include the influence on the gas content of the coal moisture content, as well as the effect of coal rank and the well temperature.

Therefore, the isotherm estimates the volume of methane liberated from the reservoir as the pressure is reduced during production, and the maximum gas recovery between the

critical desorption pressure and the well pressure at shutdown. Both the shape of the isotherm and the maximum gas content influence the volume of gas recoverable from a well. Maximum production is achieved where the isotherm shape is flattest at low pressures and where the gas content is highest at the critical desorption pressure.

CONCLUSION

A facility has been developed to measure coalbed methane isotherms. Such isotherms can be used to estimate the amount of gas recoverable from a producing well between the critical desorption pressure and the well shutdown pressure, and to answer other appropriate research questions.

REFERENCES

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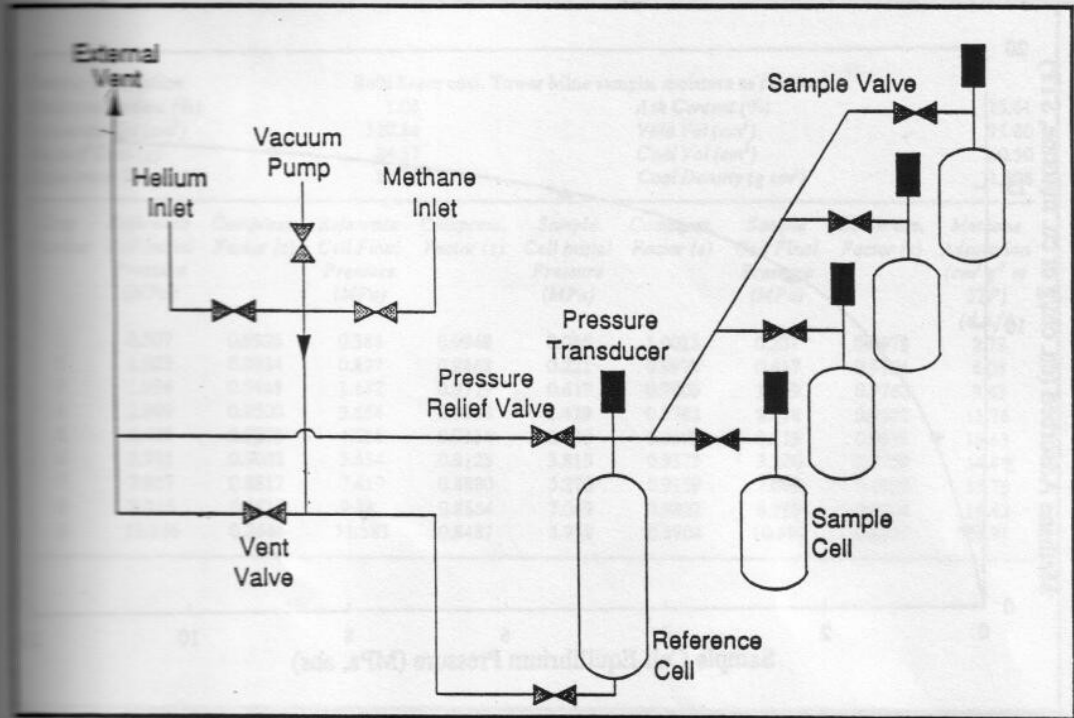


Figure 1. Schematic Diagram of coalbed methane rig

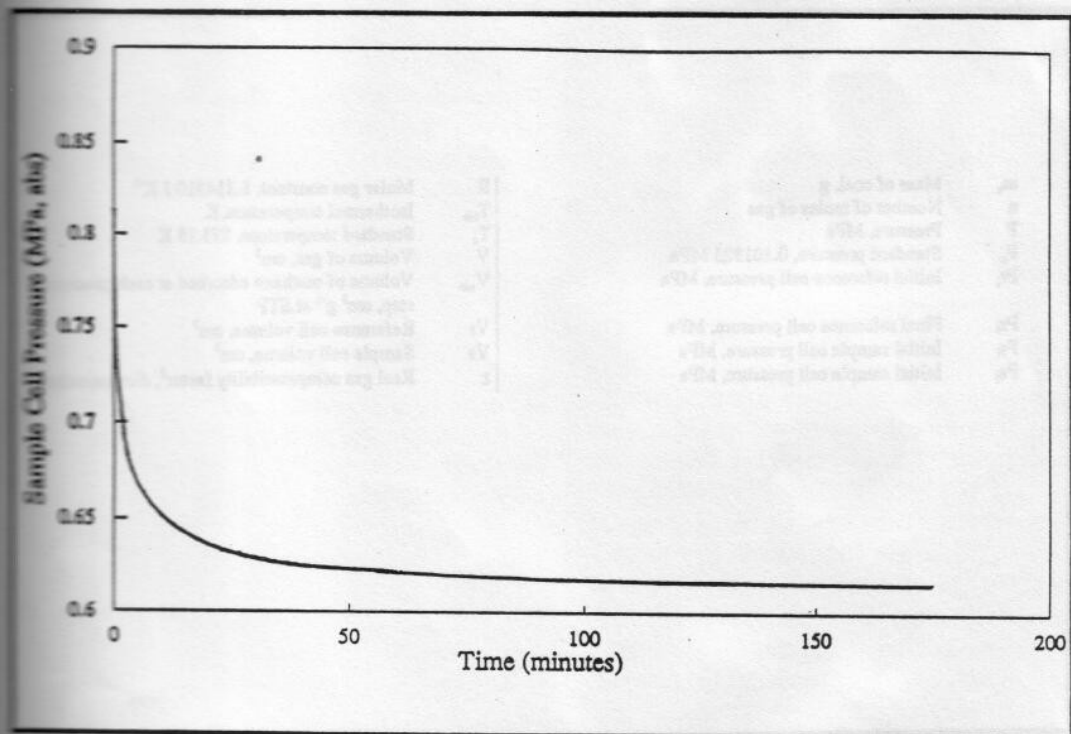


Figure 2. Adsorption step for methane on Bulli Seam coal

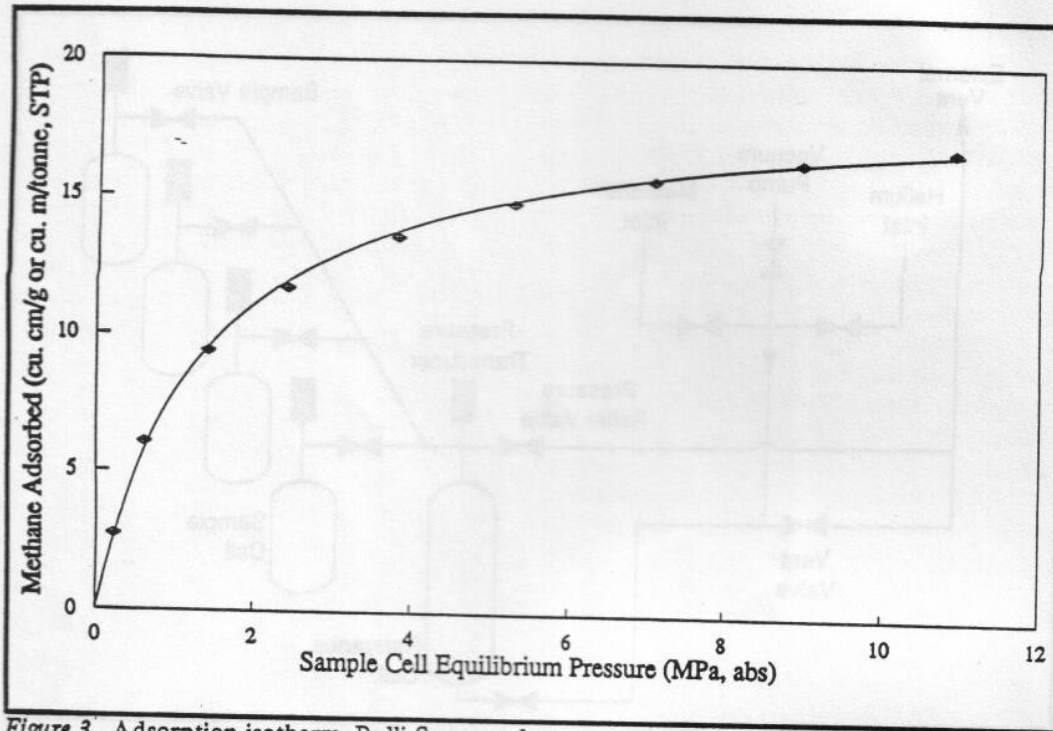


Figure 3. Adsorption isotherm, Bulli Seam coal

m_c	Mass of coal, g	R	Molar gas constant, $8.314510 \text{ J K}^{-1}$
n	Number of moles of gas	T_{iso}	Isothermal temperature, K
P	Pressure, MPa	T_s	Standard temperature, 273.15 K
P_s	Standard pressure, 0.101325 MPa	V	Volume of gas, cm^3
P_{r_i}	Initial reference cell pressure, MPa	V_{ads}	Volume of methane adsorbed at each pressure step, $\text{cm}^3 \text{ g}^{-1}$ at STP
P_{r_f}	Final reference cell pressure, MPa	V_r	Reference cell volume, cm^3
P_{s_i}	Initial sample cell pressure, MPa	V_s	Sample cell volume, cm^3
P_{s_f}	Initial sample cell pressure, MPa	z	Real gas compressibility factor ² , dimensionless

Table 1. Nomenclature

Sample Description		Bulli Seam coal, Tower Mine sample, moisture as received.							
Moisture Content (%)	1.08	Ash Content (%)							15.61
Reference Vol (cm^3)	320.84	Void Vol (cm^3)							95.80
Mass of Coal (g)	84.57	Coal Vol (cm^3)							60.50
Experiment Temp ($^{\circ}\text{C}$)	30.00	Coal Density (g cm^{-3})							1.398

Step Number	Reference Cell Initial Pressure (MPa)	Compress. Factor (z)	Reference Cell Final Pressure (MPa)	Compress. Factor (z)	Sample Cell Initial Pressure (MPa)	Compress. Factor (z)	Sample Cell Final Pressure (MPa)	Compress. Factor (z)	Methane Adsorption ($\text{cm}^3 \text{g}^{-1}$ at STP) (d.a.f.)
1	0.507	0.9926	0.386	0.9948	0.035	1.0013	0.221	0.9978	2.72
2	1.023	0.9834	0.827	0.9868	0.221	0.9978	0.617	0.9906	6.05
3	1.996	0.9665	1.682	0.9719	0.617	0.9906	1.429	0.9762	9.43
4	2.999	0.9500	2.654	0.9556	1.429	0.9762	2.438	0.9592	11.76
5	4.495	0.9270	4.058	0.9336	2.438	0.9592	3.813	0.9373	13.63
6	5.981	0.9062	5.534	0.9123	3.813	0.9373	5.270	0.9159	14.86
7	7.957	0.8817	7.419	0.8880	5.270	0.9159	7.069	0.8922	15.76
8	9.945	0.8610	9.382	0.8664	7.069	0.8922	8.958	0.8708	16.42
9	11.966	0.8444	11.387	0.8487	8.958	0.8708	10.889	0.8527	16.91

Table 2. Results, Bulli Seam coal

INTRODUCTION

Coal has a capacity for storing large volumes of gas relative to conventional classes of carbonates. Petroleum coals along the eastern coast of Australia are no exception and contain up to 30 m³ methane and gas (oil) per tonne of coal. The amount of gas retained in a coal seam is controlled by many geological factors including coal rank, mineral and elemental composition, porosity, pressure, temperature, tectonic history and geological structure. The aim of this study was to determine the influence of coal rank, composition, porosity type of gas and pressure

on the capacity of gas retention in coal.

1. assess the effect of coal rank;
2. determine the effect of coal composition on gas retention;
3. determine the effect of coal porosity on gas retention.

The first of these studies, sorption, assessed the effect of coal rank on the gas held in coal.

It has been well established that the gas sorption capacity of coal increases with increasing pressure and coal rank, and decreases with increasing temperature (Kueh, 1976). The influence of the elemental composition of coal on its gas sorption capacity, however, is poorly understood. Previous work carried out in this area by various workers have indicated contradictory results. Esminger et al. (1950) concluded that at low and medium ranks, bituminous coals have a higher CH_4 sorption capacity than vitrinite-rich coals, whereas at higher ranks, both coal types show similar results. In contrast works of Paulitsky and Cooper (1961, cited in Esminger et al. 1965), and Greedy (1979) concluded that CH_4 sorption capacity of vitrinite is greater than bituminous. To further complicate matters, the