MEASUREMENTS OF PETROPHYSICAL PROPERTIES OF COAL FOR CO_2 SEQUESTRATION

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ABSTRACT

The concept of storing carbon dioxide into coal seams is considered to be a safe and effective method for permanently storing it, with the potential added benefit of enhanced coalbed methane (ECBM) production. Recent field pilots have demonstrated the feasibility of CO_2 sequestration in coal seams. In this context it is important to improve the understanding of fluid flow and storage processes taking place. Unlike conventional reservoirs, coal primarily stores methane or CO_2 as adsorbed gas on the internal surface area of the matrix. Since the CO_2 sorption capacity of coal is much higher than that for methane, injection of CO_2 into coal beds can improve methane recovery.

The objective of this study was to evaluate the impact of CO_2 injection on a number of petrophysical properties of selected European coals. This paper presents experimental results on the matrix deformation and the simultaneous measurement of gas permeability and strain under varying stress-pore pressure conditions. The porous structure of the coals was characterised using NMR T_2 relaxation on water saturated core plugs. The coals studied presented a multimodal pore size distribution in the matrix, where the larger population corresponded to the micropores.

Coal matrix deformation (shrinkage or swelling) was measured at different sorption pressures under unconfined conditions. The matrix swelling was found to increase with CO_2 sorption pressure and displayed a positive correlation with coal rank. For a non-adsorbing gas, such as helium, an increase in net stress caused a reduction in permeability following a non-linear relationship. For the samples tested, matrix swelling due to CO_2 adsorption had a severe impact on permeability resulting in a permeability decrease. The permeability-stress-strain trend for CO_2 at constant pressure was similar to helium, indicating that it is the cleats that dominate gas flow. However, the permeability also decreased with rising CO_2 pressure, suggesting a complex interaction between coal and CO_2.

Further investigations are still necessary for different coal types and a wider range of conditions. However, the results presented in this paper provide basic characterisation of CO_2 injection in coalbeds and relevant experimental data that would enable ECBM simulators to better describe in-situ reservoir behaviour.

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INTRODUCTION

Coalbed methane is currently being produced almost exclusively by pressure depletion methods. Whilst this is simple and effective, it is not the most efficient technique as methane recovery factors are usually no more than 50% of the gas-in-place. One technology for addressing this issue is enhanced coalbed methane (ECBM) recovery. In addition to the potential for improved productivity, from an environmental perspective it serves as a possible means for permanently sequestering waste CO_2 in deep coal seams.

Coal is composed of solid matrix blocks bounded by a well-defined network of natural fractures known as cleats. In contrast to conventional sandstone and carbonate hydrocarbon reservoirs, adsorption is the principal mechanism by which gas is stored in coalbeds. Over 95% of the gas is stored at near liquid densities on the surface of the dense microporous structure constituting the coal matrix. The volume of gas contained within a coalbed will depend on factors such reservoir pressure, coal rank, pore size distribution, moisture content, wettability and inorganic ash content. Sorption isotherms are used to determine the level to which the reservoir pressure needs to be lowered in order for a certain fraction of the total gas-in-place to be recovered. The Langmuir isotherm (1916) is most commonly used to describe the sorption behaviour in coalbed methane reservoirs.

The definition of coal porosity can sometimes be an area of contention. If one were to refer to the storage capacity of coal then the porosity would be a measure of the amount of gas that can be adsorbed within the coal matrix. On the other hand, the more established understanding is that porosity is the volume fraction of the coal's interconnected cleat network that may be occupied by a particular fluid and will vary depending on the nature of the fluid (Levine, 1993). The porous structure of coal is highly heterogeneous with pore sizes varying from a few Angstroms to over a micrometer. According to the IUPAC Classification (1972), pores may be divided into macropores, mesopores and micropores, with pore diameters of $d_p > 50$ nm, $2 < d_p < 50$ nm, and $d_p < 2$ nm respectively.

Measurement of coal petrophysical properties is essential for better characterising ECBM reservoirs and evaluating methane productivity and CO_2 storage behaviour. Indeed, laboratory tests performed on coal samples by Fulton *et al.* (1980) have demonstrated that it is possible to completely recover the adsorbed methane by CO_2 injection. Laxminarayana and Crosdale (1999) studied the effect of coal rank and composition on methane sorption characteristics of coals. However, little equivalent work has been done for CO_2 sorption on coal, and the effect of coal type and rank on CO_2 sorption is not well understood. Coal rank, which is a measure of the degree of maturity of a coal, is often regarded as the main parameter governing the gas sorption capacity. A direct relationship has often been assumed between rank and gas content. On the other hand, Reucroft and Patel (1986) found that lower carbon content correlated with a higher degree of swelling. Although some research has been carried out into the mechanism by which methane flows through coal and the effects of pore size distribution on porosity and permeability, comparatively little experimental work has been done on the permeability of coal subjected to reservoir stress conditions and under the influence of CO_2 injection. Moreover, the factors influencing the extent to which CO_2 induces matrix volume change in coal are still not clear.

This paper is concerned with the effects of coal matrix swelling induced by CO_2 adsorption on the permeability of different coals under simulated reservoir conditions in the laboratory. It is aimed at characterising the pore structure of these coals and identifying coal and reservoir properties affecting matrix volume change and gas flow.

EXPERIMENTAL METHODS

This section describes experimental techniques that were implemented to determine the effect of carbon dioxide injection on matrix behaviour in different European coals. This is one of the first experimental studies where coal permeability is measured simultaneously with matrix strain. Large coal blocks representative of ranks from high volatile bituminous to anthracite were collected from opencast and underground collieries situated in Germany, France and the UK. Basic coal characteristics are shown in Table 1 (Durucan *et al.*, 2004).

For the NMR experiments a range of plug sizes were used; from 20 to 38 mm in diameter and 25 to 80 mm in length. NMR T_2 measurements were performed in a Resonance Instruments MARAN 2 (2 MHz Spectrometer) at 34°C and ambient pressure. The CPMG sequence was used to generate the T_2 decay with an inter-echo time of 200 µs, 8000 echoes and 200 scans. The relaxation time (T_2) distribution was obtained with the DXP programme from Resonance Instruments.

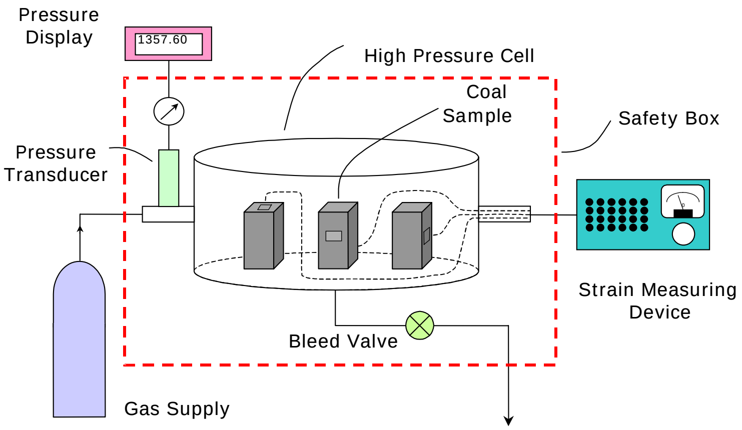


Figure 1. Schematic of experimental apparatus for matrix deformation tests.

The matrix deformation experiments were carried out using a high-pressure cell manufactured by Soilmoisture Equipment Corp, which was capable of withstanding pressures of up to 10 MPa. Cubic coal samples were placed inside the cell and volumetric strains generated in the samples were measured using sensitive strain gauges attached to each sample and a quarter-bridge monitoring strain meter as shown in Figure 1. In view of the length of time, i.e. nearly 4 months, required to complete a full adsorption/desorption cycle for solid coal using three different gases, the cell was designed to hold up to five samples simultaneously. Two strain gauges were attached to each sample, aligned in the direction of the face and butt cleats. Cubic samples were cut from coal blocks to lengths ranging between 30 and 40 mm. One-way strain gauges of 10 mm length were used during these experiments. All strain readings are reported as dimensionless microstrain values (µS). For instance, 2000 µS represents a strain of 0.002, which corresponds to an extension per unit length of 0.2%.

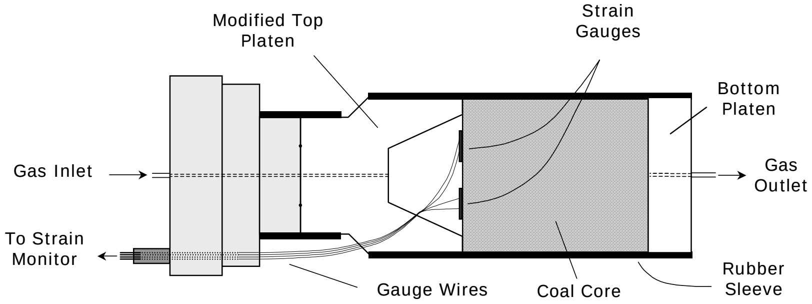


Figure 2. Schematic representation of the simultaneous strain-permeability setup.

Having tested the vessel for leakages, the cell was pressurised with helium in stages up to 7 MPa to evaluate the mechanical response of the matrix to a non-adsorptive gas. At each pore pressure, the strains were allowed to equilibrate before increasing the pressure. Once the maximum pressure had been reached, the system was depressurised in stages by bleeding controlled quantities of gas. The process was repeated using 99.7% purity methane gas or pure carbon dioxide. In the case of methane, the sample cell was pressurised up to 8 MPa, while for carbon dioxide a maximum pressure of only 5 MPa.

Having studied the effect of gas sorption on matrix swelling behaviour under static reservoir pressure conditions, dynamic laboratory tests were then performed. More specifically, simultaneous strain-permeability experiments were carried out on coal samples subjected to CO_2 injection. Cores plugs of 50 mm were cut from coal blocks belonging to each coal type using water as the coring fluid. The freshly cut cores were placed in a desiccator to remove any residual gas from the samples. To avoid oxidation the cores were then vacuum dried at 60°C. The same procedure had been applied to the cubic samples used for matrix deformation experiments. In order to achieve a simultaneous measurement of coal matrix swelling and permeability under CO_2 injection, a hydrostatic core holder capable of withstanding up to 15 MPa was used to house a single coal sample. The crucial difference between this set up and a conventional flow cell was in the distribution platen that had been specially designed to enable strain measurements to be made. The concept of the modified platen was based on creating a headspace cavity between the platen and the coal surface that could accommodate a gauge and the strain wires, which were connected to the strain indicator. This platen and two single strain gauges were placed perpendicular to one another on a non-cleated area of the upstream end of the coal sample. The modified platen and associated parts are illustrated in Figure 2. Two-way rosette gauges were employed when the available surface area was judged to be limited.

The coal core plugs were wrapped around with PTFE tape before being inserted into a Viton sleeve and loaded into the cell. The strain and permeability measurements were carried out in steps by increasing the upstream gas pressure. Once the system had fully equilibrated at a given gas pressure (showing constant strain), gas flow was initiated by controlling the downstream backpressure and steady-state permeability was measured. Further details on the experimental procedure can be found in Ahsan (2006).

COAL PORE STRUCTURE

It is commonly accepted that coals can be characterised by two distinctive porosity systems: a well defined network of natural fractures (cleats), and matrix blocks containing a highly heterogeneous porous structure. The matrix pore structure has a wide range of pore sizes ranging from a few Angstroms to more than a few microns. Some previous laboratory measurements, using mercury injection and nitrogen adsorption, indicate that the matrix pore structure is bidisperse (bimodal) or multimodal. However, for simplicity, pore transport in the matrix is usually modelled as a unimodal pore distribution. In a recent study by Shi and Durucan (2005a) a bidisperse model for gas storage was presented which successfully predicts the field gas production rates for a coalbed reservoir. Yet the assumptions made in the model need to be validated by a study of the pore size distribution and pore structure of different coals.

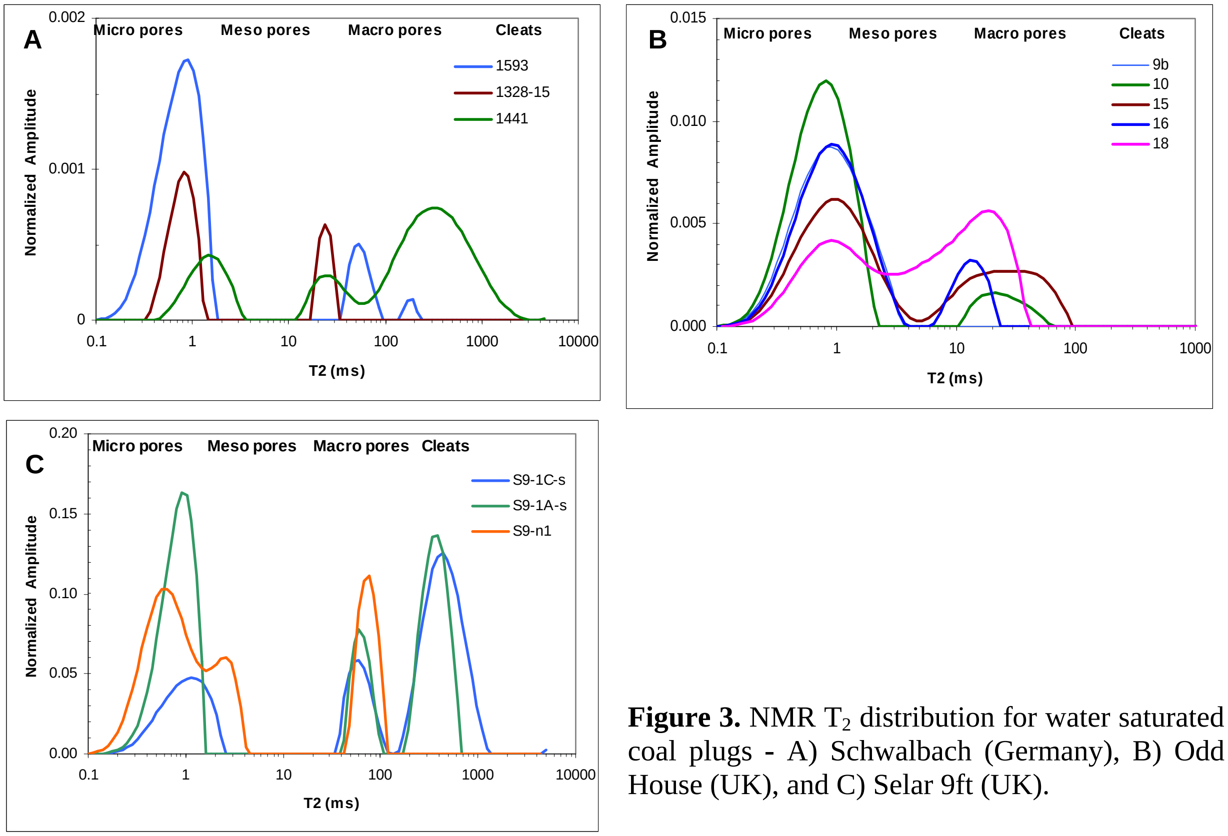


Figure 3. NMR T_2 distribution for water saturated coal plugs - A) Schwalbach (Germany), B) Odd House (UK), and C) Selar 9ft (UK).

NMR has been widely used within the oil industry to characterise oil and gas reservoirs rocks and the fluids contained within them. NMR has several advantages over conventional pore structure analysis techniques, such as mercury porosimetry and nitrogen adsorption/condensation. These include the ability to study wet materials, i.e. the pore structure of coals may change significantly during drying, and the fact that it is fast and non-destructive, so cores can be used for further studies. In our work, NMR T_2 relaxation has been successfully applied to study the coal matrix pore structure, fluid mobility and pore occupancy. However, only the pore structure of water saturated cores will be discussed here.

An example of some coal T_2 distributions of water saturated coals are shown in Figure 3. It can be clearly seen that each coal has its unique distribution and they present a bimodal or multimodal pore structure. Most of the coals tested have up to three populations of pores, plus some of them have cleats. The estimated pore classification, assuming a surface relaxivity of 1 nm/ms, is shown at the top of each plot. The micropores have similar distributions for different ranked coals. However, large differences were found in the meso and macropore distribution for different ranks. The difference in pore structure could be used to help understand the diffusion, adsorption, matrix deformation and permeability behaviour.

The total porosity of the coal samples determined by NMR was compared with the porosity obtained by weighing the plugs fully saturated with water and after drying. The agreement of both methods is very good as shown in Figure 4. A good agreement was also found for partially saturated samples. Thus, NMR T_2 relaxation can be employed as a quick method to evaluate the moisture content of coals. It should be noted that after short or mild drying most of the coals exhibit a small peak in the T_2 distribution at shorter times (~1 ms), indicating the presence of some form of hydrogen (methane or structural water) in the micropores. This peak disappeared when the coal was dried for long periods under vacuum.

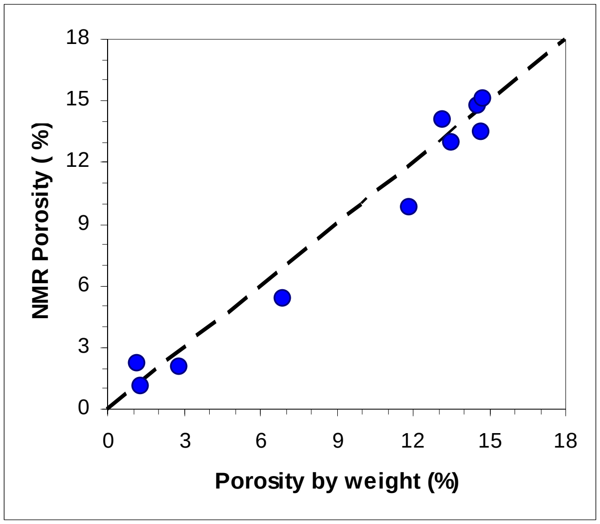


Figure 4. Comparison between porosities obtained by weight and NMR porosity

COAL MATRIX DEFORMATION

The matrix deformation behaviour, swelling and shrinking, can be clearly observed by measuring the strain versus pressure for different gases under unconfined conditions. The objective of these experiments is to measure volumetric changes in the coal matrix associated with changes in gas pressure. A typical set of strain- pressure responses are shown in Figure 5. The behaviour of the coal matrix affects the cleat aperture and therefore, the coal permeability, as will be shown later.

Coal matrix shrinkage becomes evident when helium is used, as it has a small molecular size and presents non-adsorption characteristics. Although, the coal blocks are subject to isotropic pressure conditions, the increase in helium pressure produces a linear shrinkage of the matrix due to grain compressibility. This effect is reversible with very little or no hysteresis, demonstrating the elastic response of the coal. This effect has been observed for the different of coals tested and appears to be correlated with the internal pore structure. A different response is obtained when an adsorbing gas is used, such as methane or carbon dioxide (Figure 5). The matrix swells according to a non-linear relationship due to the different amounts of gas adsorbed at each pressure. The strain-pressure relationship therefore resembles the Langmuir sorption isotherm, to which it could be related. It was found that the matrix swelling due to CO_2 sorption correlated with the rank of coal as shown in Figure 6. With regards to the strain-hysteresis of CO_2 and methane, the results were inconclusive since a range of responses were found and more studies would be needed to elucidate this behaviour. It should be noted that these results only represent the equilibrium conditions and the kinetics of the process are much slower for adsorbing gases. The transient strain response could be related to the diffusion and sorption rates which are also dependent on the internal pore structure. Coals with a larger proportion of macropores and cleats tend to have shorter equilibration times.

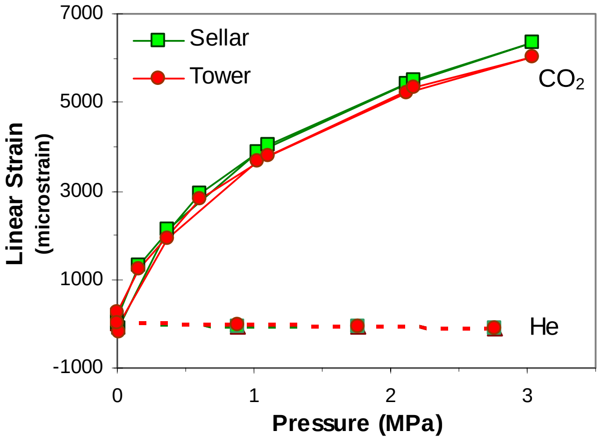


Figure 5. Coal matrix deformation (in microstrain) induced by helium and CO_2 for two anthracitic coals (Selar 9ft and Tower 7ft).

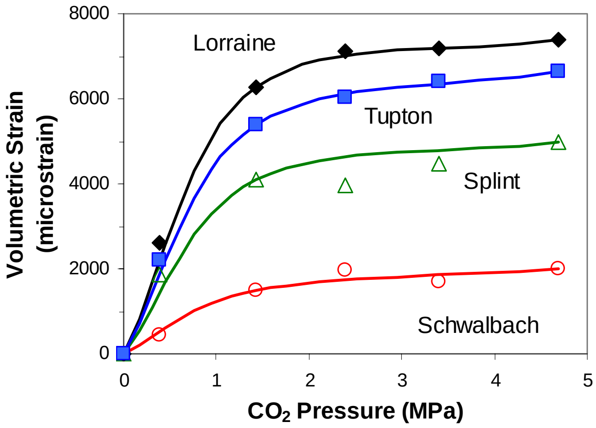


Figure 6. Matrix strain as a function of CO_2 pressure for coals of different rank.

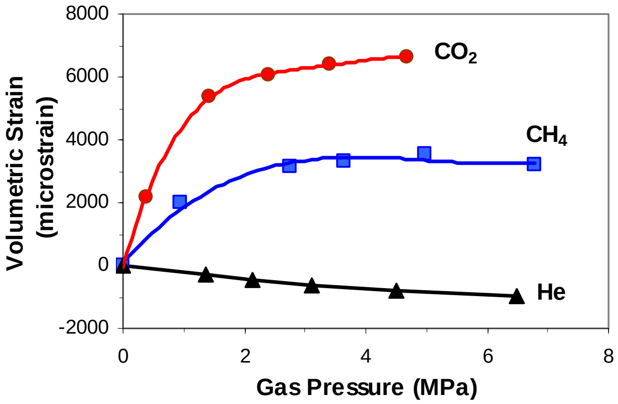


Figure 7. Matrix deformation induced by different gases for Tupton coal (high volatile bituminous A).

Matrix swelling due to CO_2 was found to be between 2-4 times larger than for methane (Figure 7). Matrix swelling has the effect of reducing the cleat size and would therefore have a large impact on gas flow.

GAS PERMEABILITY AND MATRIX DEFORMATION

In the previous section the equilibrium matrix deformation behaviour for different gases was presented. However, at reservoir conditions the coal is under a different stress regime and the flow behaviour under these conditions is very important, since it will affect the CO_2 injection-storage capacity and methane production. A novel experiment was thus designed to simultaneously determine the effects of stress and matrix shrinking/swelling on gas permeability.

In general, rocks rearrange their pore space when the stress applied to them changes, inducing changes in porosity and permeability. It is expected that coal may have similar behaviour for a non-adsorbing gas, though the response may be more complex due to matrix elasticity and the presence of cleats. As the matrix shrinks, the strain decreases linearly with increasing stress, following the same behaviour as in the matrix deformation experiments. All the coals studied presented a large dependence of permeability with net stress (confining pressure minus pore pressure) for helium. The permeability to helium showed the same trend for both increasing confining pressure and decreasing pore pressure. Figure 8 displays an example of this, where a decrease in permeability by a factor of 7 is observed when the net stress is increased from 1 to 5.5 MPa.

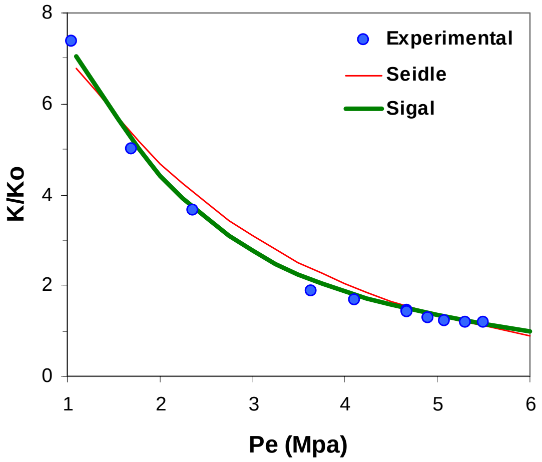


Figure 8. Experimental variation in permeability to helium as a function of net stress for Schwalbach coal and the predictions using two permeability models.

There are several coalbed permeability models in the literature, some of which have been adapted for CO_2 injection and storage (Shi and Durucan, 2005b). Two of these models that were tested here are as follows:

Seidle *et al.* (1992):
$$K = K_0 \, exp^{(-3 \, C_f \, Pe)} \tag{1}$$

Sigal (2002) based on Walsh's model:
$$K^{1/3} = K_0^{\ 1/3} + B \ln(P_e) \tag{2}$$

where K is the permeability, K_0 is the permeability at a reference stress, C_f is the cleat compressibility, P_e is the net stress, and B is proportional to the mean cleat size.

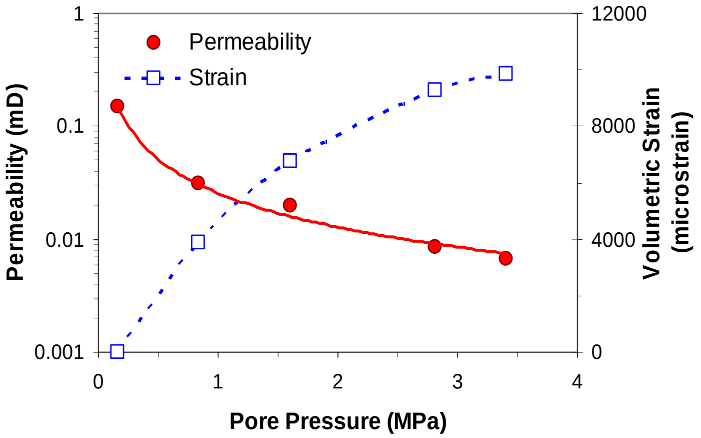
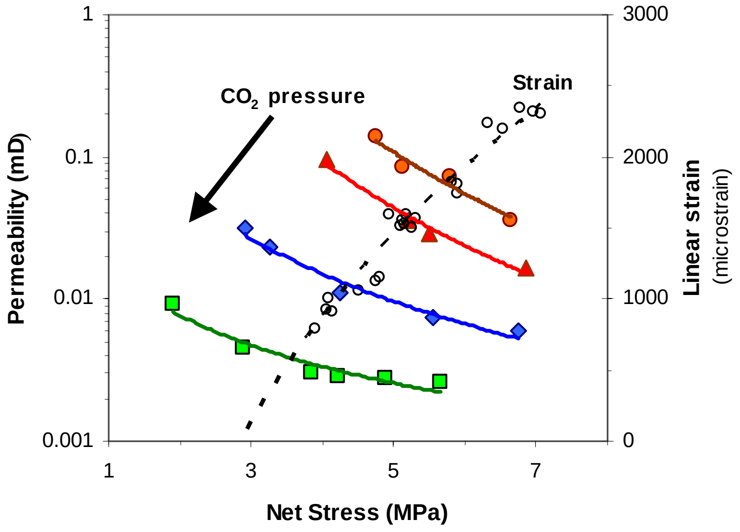


Figure 9. Permeability and volumetric strain at constant confining pressure (7 MPa) and equilibrated conditions for Lorraine coal (semi-anthracite) as a function of CO_2 pore pressure.



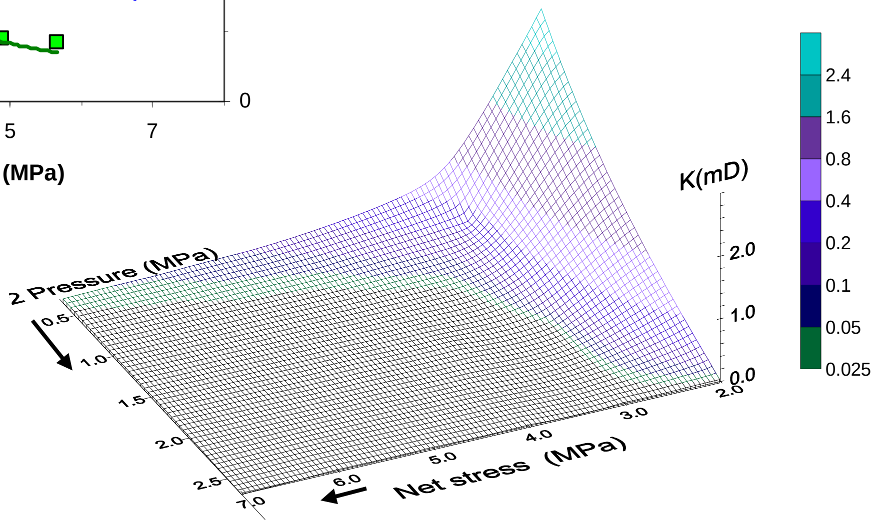


Figure 10. Simultaneous permeability and matrix deformation tests using CO_2 on Selar 9ft coal: A) Permeability and strain versus net stress, B) 3D plot of the effect of net stress and gas pressure on permeability

Both models have a good agreement with the experimental data. They support the idea that the main flow occurs through the cleats and that the decrease in permeability is controlled by cleat closure. However, the model proposed by Sigal (2002), based on a generalised Walsh's model, fits the data for this coal sample more closely. As the matrix shrinks, the strain decreases linearly with increasing net stress, following the same behaviour as in the matrix deformation experiments.

When CO_2 was used the behaviour became more complex due to the increased adsorption with pore pressure, at constant confining pressure, and this has a considerable impact on permeability as shown in Figure 9.

In this example the permeability decreases by a factor of 22 when the pore pressure is increased from 0.15 to 3.5 MPa. In other rocks an increase in permeability with pore pressure would be expected due to a reduction in net stress. However, for coal the increase in pore pressure produces more adsorption, greater swelling of the matrix and therefore a reduction in the cleat size. The matrix swelling is clearly shown by the volumetric strain increase in Figure 9. Note that the strain magnitude and behaviour is similar to that observed during the matrix deformation tests (Figures 4 and 7). Similar behaviour was observed for methane but the permeability reduction and strain increase were of lower magnitude. This is in agreement with the matrix deformation tests.

Under reservoir conditions the swelling produces an increase on the net stress. This also occurred in the experiments during the equilibration period and is reflected by an increase in confining pressure when the confining volume is kept constant. Further tests were therefore performed to study this effect on Selar 9ft, an anthracitic coal, as indicated in Figure 10. It is clear that the permeability to CO_2 is not only a function of the net stress but is also dependent on the pore pressure, and decreases significantly with an increase in either of them.

CONCLUSIONS

The basic characterisation of petrophysical properties of coals for storing carbon dioxide has been determined for a range of European coals. It has been shown that the response of coals is complex, mainly due to their internal pore structure, mechanical and physicochemical interactions. The main points to highlight from this work are as follows:

- The pore structure is specific for each coal type and can be identified using traditional NMR T_2 relaxation on water saturated plugs. This information can be used to support the behaviour of other coal properties and help with the interpretation and selection of diffusion models for simulation studies.
- NMR T_2 relaxation can be used to quickly determine moisture content at any condition without altering the coal structure through drying.

- The coal matrix is elastic and deformable, and changes in its volume can affect the dimensions of natural fractures. Hence, the strain-stress-permeability in the presence of a non-adsorbing gas, such as helium, does not differ greatly from other soft rocks.
- The adsorption or desorption of gases, such as CO_2 and methane, induces a swelling or shrinkage of the matrix consisting of micro to macropores. The swelling reduces the cleat dimensions and significantly reduces permeability of the coal.
- Matrix deformation, measured as strain variation, exhibits non-linear behaviour with CO_2 pressure in a trend similar to adsorption isotherms. The responses under confined or unconfined conditions are comparable. The matrix deformation appears to correlate with coal rank.
- The permeability of coal to CO_2 is a function of both pore pressure and net stress.

The results presented in this paper are only for ‘equilibrium conditions’ and single pure gases. Further research is required for different coals, gas mixtures and a wider range of operating conditions. However, this work can provide some of the basic characterisation needed for EBCM simulators to better describe the process of CO_2 injection.

ACKNOWLEDGMENTS

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Table 1. Summary of proximate analysis and petrology data for various coals (after Durucan *et al.*, 2004)

Coal Seam	Rank	Carbon Content (%)	Volatile Matter (%)	Ash (%)	Vitrinite	Liptinite	Inertinite
Odd House	High Vol. Bitum. A	n/a	33.2	7.4	-	-	-
Schwalbach	High Vol. Bitum. B	56.4	41.2	5.6	75.4	17.4	5.0
Warndt No.1	High Vol. Bitum. A	58.4	40.5	2.8	74.4	15.6	9.0
Splint	High Vol. Bitum. B	59.8	30.9	9.8	-	-	-
Tupton	High Vol. Bitum. A	63.2	29.2	6.2	59.4	14	25.8
Lorraine	Semi-Anthracite	83.5	10.7	36.7	31.4	0.0	0.0
Selar 9ft	Anthracite	90.4	9.5	1.5	85.6	0.0	14.2
Tower 7ft	Anthracite	90.9	8.8	3.2	84.6	0.0	15.2