Carbon Dioxide Sorption Isotherms and Matrix Transport Rates for Non-Powdered Coal

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ABSTRACT

For enhanced coalbed methane/carbon dioxide sequestration field projects, carbon dioxide isotherms and the rate of diffusion of the carbon dioxide from the cleats into the matrix are important parameters for predicting how much carbon dioxide actually will be sequestered under various operating conditions. Manometric (or pressure swing) experiments on powdered coal provide a quick, simple, and relatively inexpensive method for measuring sorption isotherms. However, determination of the rate of transport from cleat into matrix from the rate of gas pressure drop is difficult, if not impossible. (The characteristic time constant for the transport depends on the cleat spacing as well as the rate of diffusion.) Manometric measurements often yield isotherms that are extremely problematic in the region of the carbon dioxide critical point; perhaps even worse, available data seem to indicate that the sorption isotherms measured for powders are much larger than the isotherms of coal cores. Measurements on centimeter-sized samples can take weeks or months to reach equilibrium; for such equilibration times gas leakage rates that would be of no significance in powdered-coal measurements can completely invalidate manometric measurements on coal cores.

We have tested and used a simple, inexpensive method for measuring isotherms and carbon dioxide transport rates in coal cores. One or more cores are placed in a simple pressure vessel, and a constant pressure is maintained in the vessel by connecting it to a gas supply (which contains a very large amount of gas compared to amount that could leak over the course of the experiment). From time to time the gas supply is shut off, the sample is removed, and its weight is recorded at ambient pressure at frequent time intervals for a period of about one hour. The sample is then returned to the pressure vessel, the carbon dioxide pressure restored to its previous value, and the equilibration resumed until the next sample weighing. For a point on the isotherm, the process is repeated until the sample weight reaches a constant value (i.e., typically equilibration
times of several weeks). The slope of a plot of sample weight vs. square root of elapsed desorption time gives a measurement for the rate of diffusion.

In order to advance all three experimental methods, results from this “ambient-pressure gravimetry” method were compared with data obtained by conventional manometry and by computer tomography.

The isotherm and “diffusion” rate measured for the core can be directly used in simulators for reservoir engineering studies of coalseam sequestration and enhanced coalbed methane production.

INTRODUCTION

The sorption isotherms establish a theoretical limit for how much carbon dioxide can be sequestered/will be needed in a sequestration/enhanced coalbed methane field project (Bromhal, et al., 2005). Because not all of the coal will be contacted by CO₂, in practice the theoretical limit is never reached. However, sorption isotherms are still needed as input for reservoir engineering computations of more realistic estimates. Furthermore, unless the CO₂ is injected very slowly and the cleats are closely spaced, the rate of diffusion of the carbon dioxide from the cleats into the matrix is also an important parameter for predicting how much carbon dioxide actually will be sequestered under various operating conditions (Sams et al., 2005; Smith et al., 2005). Thus the rates of diffusion and sorption isotherms are important parameters for both technical and economic purposes (Jikich et al., 2004; Gorucu et al., 2007).

Manometric (or pressure swing) experiments on powdered coal provide a quick, simple, and relatively inexpensive method for measuring sorption isotherms. However, mechanical confining pressures that mimic in situ lithostatic pressures cannot be applied to powdered samples. It is still unclear whether lithostatic pressures significantly decrease sorption amounts, or not. However, it can require several weeks to establish sorption equilibrium in a non-powdered sample sufficiently large for application of a confining pressure. Thus, for a coal core it is difficult to ascertain whether a decreased sorption (as compared to the powder isotherm) has been caused by the confining pressure, or by insufficient time for diffusion of the gas into the sample. An additional complication is that gas leakage rates which would be inconsequential in a manometric study on powdered coal become intolerable when each equilibration requires several weeks, or more.

For these (and other) reasons, we have “gone back” from the study of centimeter-sized coal samples under mechanical confining pressures to the study of similarly sized samples without confining pressure. Moreover, in place of the indirect, manometric technique, we have explored two techniques for direct measurement of sorption isotherms and rates of carbon dioxide transport in coal: (1) computer X-ray tomography (CT), (Karacan and Mitchell, 2003) and (2) a gravimetric technique, in which the sample is weighed at atmospheric pressure, using a conventional electronic balance. When the two techniques are applied to the same sample(s), results from the two methods can be directly compared (i.e., the gravimetric results can be used to “calibrate” the CT method. The gravimetric and CT methods are described in the following, Experimental, section. Our initial results from each method are presented and compared in the third, Results,
section; and some of the advantages, disadvantages, and limitations of each method are discussed in the Discussion that follows the Results.

EXPERIMENTAL METHODS

Samples and Sample Preparation

Four samples were prepared from a large piece of Pittsburgh coal from the Emerald mine near Waynesburg (Greene Co), Pennsylvania. The samples included (a) a powdered sample, used in manometric sorption isotherm measurements; (b) a cylinder 4.6 cm long and 1.65 cm in diameter, also used in manometric sorption isotherm measurements; and (c) and (d) two cylinders, each 2.5 cm long and 2.5 cm in diameter. After drying, sample (c) was subsequently exposed to CO₂ at pressures 1.38 MPa (200 psi) and 3.10 MPa (450 psi); sample (d) was exposed to CO₂ at pressure 4.65 MPa (675 psi). The powdered coal was sieved to remove particles larger than 60 mesh, then resieved through a ~400 mesh sieve to remove particles less than 45 µm in size. The samples were dried in a vacuum oven for a week at 40°C; the weight losses after vacuum drying were 0.29%. The weights (after drying) of samples (c) and (d) were 21.3178 g and 22.0906 g, respectively. Standard petrographic techniques were used for the maceral analysis (ASTM, 2005a, b).

Computer X-ray Tomography (CT)

Computer X-ray tomography measurements were performed with a Universal 350E instrument operated at 100 mA and 80 kV or 130 kV, respectively. Contiguous “slices” of thickness 2 mm (i.e., “height” of the trans-axial cylinder sampled for each image, See Fig. 1) were scanned along the entire length of each cylindrical core studied. Each slice required 4s to complete. Scans were performed on “bare” cores, that is, without any core holder, both before exposure to CO₂, and after the selected CO₂ sorption time had elapsed. To minimize gas losses during the scanning procedure, instrument warm-up and sample alignment procedures were completed before depressurization of the core holder in which the CO₂ exposure occurred; hence, samples with sorbed CO₂ were scanned immediately after removal of the sample from the equilibration cell, or from the balance. The elapsed times from initiation of depressurization until each subsequent step were recorded, so that corrections for degassing by the bare core could be calculated. (See below.) Complete sets of contiguous scans took about one minute at each voltage. Calibration scans of aluminum, fused silica, and water (of known densities) were performed after each set of coal scans, for calculation of the sample densities and CO₂ contents of the cores. These measurements were made at 22 °C.

 Ambient-Pressure Gravimetry

The basic procedure of the ambient-pressure gravimetric method is very simple: The dried sample is weighed. The sample then is placed in a high-pressure cell, evacuated, and exposed to gas at the desired pressure and temperature. Once the desired exposure time has been reached, the cell is depressurized and the sample removed. The sample weight is automatically recorded periodically (e.g., every 30 seconds) for a suitable period of time (e.g., 60 minutes); the time required to depressurize the cell and
make the first weighing is recorded. These steps are repeated until the sample reaches the desired gas uptake—e.g., until a constant weight is achieved, if the sorption isotherm is being measured. The procedure can be repeated for as many gas pressures as are desired.

The sample weights \( w_s \) are plotted vs. \( t^{1/2} \) (where \( t \) is elapsed time, measured from initiation of depressurization until the time that each weight is taken). A linear regression to the plot is calculated. Because the gas pressure in the cell does not fall to atmospheric pressure immediately, the time at which the sample begins to degas is somewhat ill-defined. However, in practice this elapsed time can be about one minute. The first recorded weight provides an accurate lower limit to weight of the sample just before depressurization began; the intercept of the regression provides an upper estimate to the sample weight just before depressurization was started.

The slope of the regression provides a measure of \( D/\phi \), where \( D \) is the diffusivity and \( \phi \) the sample porosity (Castellan, 1964; Mavor et al., 1990).

**Ambient-Pressure Gravimetric and CT Methods Combined**

A major advantage of CT methods is their ability to measure spatial variations of the \( \text{CO}_2 \) concentration within the sample. These local values can be averaged over the entire sample to give the average concentration, e.g., a point on the conventional isotherm. When the gravimetric method and the CT method are applied to the sample under reasonably similar conditions, the gravimetric method can furnish a check on the accuracy of the CT measurement. In this procedure the sample is imaged as quickly as possible after it is removed from the depressurized sample cell. The sample is then weighed periodically and then rescanned. The times elapsed from initiation of depressurization until (successively) the first CT measurements, each periodic weighing, and the second CT measurements are each recorded. The sample weights \( w_s \) are plotted vs. \( t^{1/2} \) (where \( t \) is elapsed time, measured from initiation of depressurization until the time that each weight is taken), and the linear regression to the plot is calculated. The weight of the sample at the time of each of the CT measurements is calculated from the regression, and the amounts of \( \text{CO}_2 \) lost from the sample between the two sets of images is calculated from these two weights. The densities of the sample also are calculated from the measured CT numbers of the sample and of the calibration materials. The change in the amount of \( \text{CO}_2 \) in the sample between two sets of scans is calculated from the changes in sample density. The sorption need not have reached its equilibrium value for the calibration to be valid, e.g., if the transport rate is to be calculated from the CT-measured spatial distribution of sorbate in the sample. Both the CT and gravimetric measurements were made at 22 °C.

**Manometry**

To illustrate the effects of sample dimensions on measured isotherms of unconfined coals, a standard manometric technique (Goodman et al., 2004) was used on a sample of coal from the coal in non-powdered and powdered form. Helium manometry was used to obtain the volume of each of the samples. Measurements were made at 30 °C.
RESULTS

Maceral analysis of the Emerald Mine coal gave 68.0 vol% vitrinite, 19.6 vol% liptinite, 12.4 vol% inertinite on an mmf basis.

Figure 2 illustrates the sorption isotherms measured manometrically for Pittsburgh coal from the Emerald Mine in powdered and cylindrical (1.65 cm diameter) form, respectively. The dashed lines represent fits of the Langmuir equation (Langmuir, 1918) to the two sets of data. The values of the fitting parameters were \( P_L = 0.63 \text{ MPa} \) and \( V_L = 0.0327 \text{ g CO}_2/\text{g coal} \) (16.7 cm\(^3\)/g) for the powdered coal, \( P_L = 1.37 \text{ MPa} \) and \( V_L = 0.0207 \text{ g CO}_2/\text{g coal} \) (10.6 cm\(^3\)/g) for the non-powdered sample. (For the powdered coal \( R^2 = 0.9985 \) for the fit of \( P/V = P/V_L + P_L / V_L \) to the data; for the non-powdered coal, \( R^2 = 0.9962 \) for the fit of this equation.) Here \( P \) and \( V \) are the measured pressure and “volume” (converted to mass basis), \( P_L \) and \( V_L \) the Langmuir pressure and “volume.” The Langmuir volume for the non-powdered coal was only about two-thirds that of the powdered coal.

Figure 3 illustrates the decrease with time of the weight of a coal cylinder (length 2.5 cm, diameter 2.5 cm) as it degassed into the ambient air at atmospheric pressure. The Emerald Mine sample had been in contact with CO\(_2\) at 4.65 MPa (675 psi) for 17 days. As expected for Fickian diffusion from a sample that is approximately spherical, the curve in Fig. 3 is approximately parabolic for the 7 hr weighing period.

Figures 4 illustrate the degassing behavior of another cylinder of Emerald Mine coal (also length 2.5 cm, diameter 2.5 cm) as it degassed into the ambient air at atmospheric pressure. This sample had been in contact with CO\(_2\) at 3.10 MPa (450 psi) for 31 days. Figure 4 (a) is a plot of sample weight \( (w_s) \) vs. square root of time on the balance; Fig. 4 (b) is a plot of the same data, except that an approximate correction (42 s) has been applied for the time required to depressurize the equilibration cell and place the sample on the balance. The correction to the sample degassing time changed the regression intercept (i.e., calculated weight of the sample just before depressurization was started) from 21.973 g to 21.979 g; the correction changed the slope of the regression from -0.1610 g/h\(^{1/2}\) to -0.1673 g/h\(^{1/2}\); and the value of \( R^2 \) was slightly increased from 0.9977 to 0.9988. As illustrated by Fig. 4, the increases in linearity and in \( R^2 \) occurred mainly because of the effects of the time correction on the first few points. Obviously, the weight of the sample just before depressurization was greater than 21.961 g (the first weight recorded). Thus, we can take 21.973 \( \pm \) 0.006 g as our best value for that weight. The weight of the sample before exposure to was 21.3178 g; thus, we obtain 0.655 g \( \pm \) 0.006 g for the weight of the sorbed CO\(_2\) and (0.655 g \( \pm \) 0.006) g CO\(_2\)/(21.3178 \( \pm \) 0.001) g coal as the average concentration of CO\(_2\) sorbed by the sample after 31 days.

The results illustrated by Figs. 4 were typical. For example, for data collected after 10 days, 31 days, 67 days, and 104 days, respectively, in every case the approximate degassing time correction used in Fig. 4 (b) increased the calculated sample weight at the initiation of depressurization by 0.005 \( \pm \) 0.001 g from the weight obtained without the time correction. As in Fig. 4, application of the time correction typically changed the slope of the regressions by about 4%. The weights of CO\(_2\) sorbed were typically about 0.6 g, i.e., much, much larger than the \( \pm 0.0001 \) g precision of the balance.

In Fig. 5, the y-axis is for the absolute values of slopes of linear regressions to plots of \( w_s \) vs. \( t^{1/2} \) [e.g., Fig 4 (b)] (where \( t \) is elapsed time measured from initiation of depressurization until the time of each weighing). The cylindrical sample of Emerald
Mine coal had been in contact with CO₂ at 3.10 MPa for the various elapsed times shown on the x-axis. Figure 5 illustrates how the diffusion rate apparently varied slightly with the length of time of exposure to CO₂. It is striking how the shape of the curve in Fig. 5 is similar to that of Fig. 6 (below) for the amount of sorption. Of course, the regression slopes and intercepts plotted in Figs. 5 and 6, respectively, are mathematically correlated. It is not yet clear whether there is any physical or chemical contribution to the maximum observed in Fig. 5.

Figure 6 illustrates the amounts of CO₂ sorbed after different times of exposure of the coal to CO₂. Results are shown for two different pressures; the gas pressures were 3.10 MPa (450 psi) and 4.65 MPa (675 psi), respectively. (The 3.10 MPa sample (c) was that of Fig. 4; the 4.65 MPa sample (d) was the sample of Fig. 3.) In Fig. 6 the sorption apparently reached an equilibrium value of 0.030 g CO₂/g coal after 31 days, then decreased slightly to about 0.026 g CO₂/g coal with longer equilibration times. It is unclear whether the unexpected decrease is correct, or an experimental artifact. However, the observed trend is both smooth and remarkably reproducible (between the two equilibration pressures). The agreement between the results for the two pressures suggests that the amount of CO₂ sorbed (0.030 g CO₂/g) were close to its maximum value.

In Fig. 2 the manometrically measured sorptions were much larger for powdered Emerald Mine coal than for the non-powdered sample. Comparison of the data in Fig. 6 with the data in Fig. 2 shows excellent agreement between non-powdered samples in Fig. 6 and the powder sample in Fig. 2. This indicates that the manometric technique may not be suitable for many measurements on non-powdered samples: although the pressures appeared to have reached constant values for the solid-sample points in Fig. 2, the technique evidently was simply too insensitive for the long times (~30 days) required to reach equilibrium.

Computer tomography images (at 130 kV) of a coal core (c) measured before and after degassing for about one hour are shown in Figs. 7 (a) and Fig. 7 (b), respectively. The sample had been exposed to CO₂ at 1.38 MPa (200 psi), but had not necessarily reached equilibrium (cf. Fig. 2). These images, which are more or less “typical,” highlight the heterogeneity of coals—including the heterogeneity of their sorption properties and diffusion rates—even over sub-centimeter distances.

Figure 8 is a graph of CT number plotted vs. density, for three calibration materials (water, silica, aluminum). The CT numbers of the calibration materials were measured with the same instrumental settings (e.g., 130 kV) as used for the images of Fig. 7. The slope of the regression in Fig. 8 is 0.0005293 g cm⁻³ (CT number)⁻¹ and the intercept is 0.8891 g cm⁻³ (R² = 0.9959). From this regression and the CT numbers measured for the coal samples, the densities of the latter were calculated. From the data illustrated by Figs. 7 and 8, the change of density for each pair of “before and after” images in Fig. 7 (a) and Fig. 7 (b) was calculated; these changes then were averaged over all of the image pairs. Thus, by CT we obtained a loss of 1.17 cm³ CO₂ (g coal)⁻¹ [0.00230 g CO₂ (g coal)⁻¹] from the sample between the two sets of images.

The sample whose images are shown in Figs. 7 was weighed at fixed time intervals during the period of degasification between the collections of the two sets of CT data. These sample weights are plotted in Fig. 9 as weight (wₛ) vs. t¹/₂, where t is elapsed time measured from initiation of depressurization until the time of each weighing. The
regression to the data gave \( w_s = -0.1307 \text{ g hr}^{-1} + 21.845 \text{ g} \) (\( R^2 = 0.9992 \)). From the regression equation and times elapsed from depressurization, the loss of CO\(_2\) by gravimetry was calculated for comparison with the loss obtained from the imaging. The values obtained were, respectively, 1.25 cm\(^3\) CO\(_2\)/g coal (2.45 mg CO\(_2\)/g coal) and 1.17 cm\(^3\) CO\(_2\)/g coal (2.30 mg CO\(_2\)/g coal). Although this is (to our knowledge) the first direct calibration of a CT-measured sorption by means of another technique, the agreement is encouraging for the future of CT measurements.

The ambient-pressure gravimetry and CT imaging methods each illustrate that, for centimeter-sized samples, gas-loss rates may be sufficiently small that the sample can be accurately studied at ambient pressure (or quickly transferred from a high-pressure, long-term-equilibration, sample-preparation vessel, to a high-pressure instrument, without major gas loss).

**DISCUSSION**

Figure 2 illustrates one of the dilemmas that the ambient-pressure gravimetric method seeks to address: for the manometric measurements the Langmuir volume of the non-powdered sample was only two-thirds the Langmuir volume of the powdered sample, although pressure vs. time measurements seemed to indicate that the non-powdered sample had reached equilibrium. The fit of the Langmuir equation to the powdered-coal data one calculates 0.27 g CO\(_2\)/g coal at 3.10 MPa and 0.29 g CO\(_2\)/g coal at 4.65 MPa for the powdered-coal sorptions. By the ambient-pressure gravimetric method we obtained 0.25 g CO\(_2\)/g coal to 0.30 g CO\(_2\)/g coal in this pressure range, depending on which part of the curve in Fig. 6 we choose. From this we conclude that although the non-powdered, manometric sample appeared to have reached equilibrium, it fact it had not. We also conclude that the manometric-powder and gravimetric-cylinder data were in good agreement for two totally independent experimental methods. Similarly, using Pocahontas coal, Kelemen and coworkers (2006) obtained sorption data for both powdered and a solid piece measuring approximately 7x7x5 mm\(^3\) in size; they found that the sorption capacity of the powder and that of the solid piece were comparable.

The other comparison performed was between ambient-pressure gravimetry and CT imaging. The major advantage conferred by CT imaging is its ability to measure density heterogeneities in the coal (before sorption), as well as inhomogeneities in the amounts of gas sorbed by various portions of the sample. However, data obtained by an independent method were needed to check the accuracy of CO\(_2\) sorption measured by the CT method. For the loss of CO\(_2\) between two sets of images we obtained 2.30 mg CO\(_2\)/g coal by the CT method, 2.45 mg CO\(_2\)/g coal by gravimetry; that is, the CT result was about 6% smaller. To our knowledge, this is the first calibration of CT methods for CO\(_2\) sorption with another method; further calibrations of this type are needed. However, this agreement between two totally independent experimental methods is very satisfactory.

For measurements on non-powdered samples, the advantages of the ambient-pressure gravimetric method include directness, simplicity, cost, and productivity. Unlike manometric methods, the measurements are made directly on the coal. Unlike high-pressure gravimetric methods, the instrumentation is inexpensive and commonly available, and no buoyancy correction for the density of the gas is needed.
Regardless of the measurement technique, equilibration times can be shortened only by shortening diffusion distances; no instrumental method can eliminate this requirement when non-powdered samples are required. However, ambient-pressure gravimetry allows multiple coal samples to be simultaneously “pre-equilibrated” within a single, inexpensive high-pressure cell; and the time required to actually make simultaneous measurements of a gas content and rate of gas loss is little more than an hour. If the sample were at equilibrium, these two simultaneous measurements provide a point on the sorption isotherm, and a measure of the coal diffusivity, both needed for the design and interpretation of carbon dioxide sequestration/enhanced coalbed methane field projects.

CONCLUSIONS

We have developed and tested an ambient-pressure, gravimetric technique for measuring sorption isotherms and diffusivities for gases in laboratory-sized, mechanically unconfined coal cores. The results support the following conclusions:

1. Ambient-pressure gravimetry is a direct, accurate, simple, inexpensive, and rapid (exclusive of equilibration times) method for measuring isotherms and diffusivities of mechanically unconfined coal cores.
2. Manometry (while suitable for powdered coals) may give misleadingly low sorptions for coal cores, even when pressure measurements over extended equilibration times seem to indicate that equilibrium had been attained.
3. Computer tomography (CT) measurements, can provide not just qualitative measures of spatial variations of carbon dioxide concentrations within coal cores, but quantitatively accurate measures of CO₂ sorptions.
4. Often losses of CO₂ from coal cores will be sufficiently slow during sample transfers at atmospheric pressure, that it may be useful to maintain a supply of samples pre-equilibrated (for approximately one month each) at various gas pressures, which can be quickly transferred to the laboratory’s various measurement instruments.

ACKNOWLEDGEMENTS

We thank Gino Irdi for performing the maceral analysis, Bryan Tennant for other laboratory help, Tom Mroz for the coal sample, and Kathy Bruner for obtaining information on Emerald Mine coal. This work was funded by the U.S. Department of Energy.

REFERENCES


Fig. 1. Illustration of geometry of computer tomography (CT) scanning of a coal core.

Fig. 2. Sorption isotherms measured manometrically for Pittsburgh coal from the Emerald Mine in powdered and cylindrical form, respectively. The dashed lines represent fits of the Langmuir equation to the two sets of data.
Fig. 3. Decrease with time of weight of a degassing, cylindrical coal sample that had been in contact with CO₂ at 4.65 MPa (675 psi) for 17 days.
Fig. 4. (a) Sample weight ($w_s$) vs. square root of time on the balance and linear regression to the data; (b) $w_s$ vs. $t^{1/2}$ and linear regression, where $t$ is elapsed time measured from initiation of depressurization until the time that each weight. The cylindrical sample of Emerald Mine coal had been in contact with CO$_2$ at 3.10 MPa (450 psi) for 31 days.

Fig. 5. Y-axis: absolute values of slopes of linear regressions to plots [e.g., Fig 4 (b)] of $w_s$ vs. $t^{1/2}$, where $t$ is elapsed time measured from initiation of depressurization until the time of each weighing. The cylindrical sample of Emerald Mine coal had been in contact with CO$_2$ at 3.10 MPa for the various elapsed times shown on the x-axis.
Fig. 6 Carbon dioxide sorptions of Emerald Mine coal as a function of length of exposure to CO₂, as measured by ambient-pressure gravimetry: diamonds--coal core (c), equilibrated at 3.10 MPa (450 psi); circles--coal core (d), equilibrated at 4.65 MPa (675 psi).
Fig. 7. Computer tomography images of a coal core (c), measured at 130 kV and atmospheric pressure: (a) before degassing, “immediately” after depressurization from exposure to CO$_2$ at 1.38 Ma (200psi); (b) after gravimetric degassing for about one hour. The order of the contiguous images in Fig. 7 (a) and (b) is from 1 to 12.

Fig. 8. Density vs. CT number at 130 kV for calibration materials (H$_2$O, SiO$_2$, Al), and linear regression to the data; the slope of the regression is 0.0005293 g cm$^{-3}$ (CT number)$^{-1}$ and the intercept is 0.889.1 g cm$^{-3}$, ($R^2 = 0.9959$).
Fig. 9. Sample weight ($w_s$) vs. $t^{1/2}$, where $t$ is elapsed time measured from initiation of depressurization until the time of each weighing; the weightings were performed within the time interval between acquisitions of the CT images of Fig. 6.