

Improvements in Measuring Sorption- Induced Strain and Permeability in Coal

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Abstract

Total worldwide CBM in-place reserves estimates are between 3500 Tcf and 9500 Tcf. Unminable coal beds have been recommended as good CO₂ sequestration sites as the world prepares to sequester large amounts of greenhouse gases. In the U.S., these coal seams have the capacity to adsorb and sequester roughly 50 years of CO₂ emissions from all the U.S. coal-fired power plants at today's output rates. The amount and type of gas adsorbed in coal has a strong impact on the permeability of the coal seam. An improved mixed gas adsorption isotherm model based on the extended-Langmuir theory is discussed and is applied to mixed gas sorption-induced strain based on pure gas strain data and a parameter accounting for gas-gas interactions that is independent of the coal substrate. Advantages and disadvantages of using freestanding versus constrained samples for sorption-induced strain measurements are also discussed. A permeability equation used to model laboratory was found to be very accurate when sorption-induced strain was small, but less accurate with higher strain gases.

Introduction

Coal bed reservoirs in the U.S. contain an estimated 141 Tcf of recoverable natural gas, which accounts for 10% of the total recoverable natural gas reserves in the U.S. (Nelson 1999). Coalbed methane (CBM) production also accounted for 10% of the total U.S. natural gas production in 2002 (Leach 2002). Total worldwide CBM in-place reserves estimates are between 3500 Tcf and 9500 Tcf (Olsen et al. 2003).

There is a strong relationship between CBM operations and CO₂ sequestration. Methane production from coal beds can be enhanced by injection of other gases to displace or strip methane from the coal and accelerate its

production at higher reservoir pressures in a process called enhanced coal bed methane (ECBM). The micro-porosity of coal can adsorb up to 10 times more CO₂ than methane on a molecular basis, while it can adsorb roughly half as much nitrogen as methane (Reeves 2003). Because coal is such a strong adsorber of CO₂, unminable coal beds have been recommended as good CO₂ sequestration sites as the world prepares to sequester large amounts of greenhouse gases (mainly CO₂) to limit their potential effects on climate change (Folger 2007). Coal seams in the U.S. that are either too deep or too thin to be economically mined have the capacity to adsorb and sequester roughly 50 years of CO₂ emissions from all the U.S. coal-fired power plants at today's output rates of 90 Gt of CO₂ per year (U.S. DOE 2007; Reeves 2003).

Unlike conventional gas reservoirs, methane in coal is not stored as free gas but rather as sorbed gas, at near-liquid densities on the internal surface area of the micro-porous coal (Puri and Yee 1990). As gas molecules are adsorbed onto adsorption sites within the coal matrix, the matrix blocks swell; and as gas is desorbed, the coal matrix shrinks. The more tightly the gas molecules are packed onto the adsorption sites, the larger the swelling. Chikatamarla et al. (2004) determined that at a given pressure, if a coal matrix block is saturated with a high-boiling-point gas, such as CO₂, the volume of the coal block will be larger than when saturated with a low-boiling-point gas, such as helium.

The amount and type of gas adsorbed in coal has a strong impact on the permeability of the coal seam. Permeability of a coal bed is a function of cleat spacing and width (Robertson and Christiansen 2006). The swelling and shrinkage that occurs within the coal matrix blocks as different gases are injected into coal beds to displace methane or as gas reservoir pressure changes can cause a significant change in cleat width and a corresponding change in permeability. Being able to accurately predict permeability changes in coal beds as gases are produced or injected is important for designing surface facilities, predicting production and injection rates, and anticipating economic profitability of operations.

Swelling and shrinking of coal induced by the sorption of gases is most commonly referred to as sorption-induced strain. Sorption-induced strain in coal is not a linear function of pressure; instead, strain rapidly increases at low pressure and asymptotically approaches a maximum value as pressure becomes high. Sorption-induced strain is most commonly modeled using an equa-

tion of the same form as the Langmuir isotherm used to model gas content in coal as a function of pressure. The Langmuir isotherm equation for a pure gas adsorbed onto the microporous coal matrix is:

$$V = V_L \frac{p_L}{p + p_L}, \dots\dots\dots (1)$$

where V is the volume of adsorbed gas in the coal matrix; V_L is known as the Langmuir volume, which corresponds to the maximum volume of adsorbed gas at infinite pressure; p_L is the Langmuir pressure, which corresponds to the pressure value at which V is equal to $V_L/2$; and p is the pressure of the gas surrounding the coal. The values for V_L and p_L are obtained by measuring the values of V as pressure p changes and then fitting Eq. 1 to the measured data.

Because sorption-induced strain correlates well with gas adsorption, the equation used to model sorption-induced strain as a function of gas pressure is of the same form as the isotherm equation:

$$S = S_L \frac{p_{SL}}{p + p_{SL}}, \dots\dots\dots (2)$$

where S is the sorption-induced strain; S_L is the Langmuir strain, which corresponds to the maximum sorption-induced strain at infinite pressure; p_{SL} is the Langmuir pressure, which corresponds to the pressure value at which S is equal to $S_L/2$; and p is the pressure of the gas surrounding the coal. The values for S_L and p_{SL} are obtained by measuring the values of S as pressure p changes and then fitting Eq. 2 to the measured data.

The sorption-induced strain parameters (S_L and p_{SL}) can then be included into equations used to calculate permeability changes in coal as methane is produced from coal beds or as other gases, such as CO_2 , are injected and sequestered in unminable coal beds. Examples of coal permeability equations that incorporate the effect of sorption-induced strain include those presented by Palmer and Mansoori (1998), Shi and Durucan (2003), and Robertson and Christiansen (2006) among others.

If sorption-induced strain parameters (S_L and p_{SL}) are not measured or known, reservoir modelers can use them as history-matching variables or modelers can also ignore sorption-induced permeability equations altogether and assume a pressure vs. permeability relationship based on the performance of analog reservoirs. However, obtaining accurate sorption-induced strain parameters is preferable because it allows the use of equations that represent the physical processes involved with changes in permeability to be used, lessens inaccuracies in reservoir modeling, and helps in the understanding of how permeability changes within a coal seam.

The goals of the paper are to 1) summarize work to develop techniques to measure sorption-induced strain; 2) discuss the advantages of collecting sorption-induced strain data under stress-free or freestanding conditions; and 3) apply the permeability model proposed by Robertson and Christiansen (2006) for sorptive-elastic media such as coal to permeability data measured in the laboratory.

Sorption-induced strain measurements.

Until recently, sorption-induced strain data for use in permeability models have been difficult and tedious to collect resulting in very little published data on the sorption-induced strain properties of coal. However, recent advancements have allowed for the collection of strain data in a much more timely fashion.

Most of the sorption-induced strain data measured to date have been done by attaching strain gauges to coal samples. Resistance-type strain gauges have presented some problems associated with data collection. Harpalani and Schraufnagel (1990) used a coal sample 1.5 inches in diameter and 3 inches in length in their strain measurement experiments using strain gauges. They noted that the desorption process was extremely slow and that it took a long time for the readings on the strain indicator to stabilize while the adsorption process was somewhat faster. Seidle and Huitt (1995) noted that the strain gauges experienced some irreversible strain at the end of each pressure swing, which complicated their analysis. They found that with the large samples required for measurements using strain gauges, the sorption process was slow, requiring nearly three months for stabilization during adsorption, but desorption strain stabilized more rapidly—requiring only 10 days at each pressure to equilibrate, which was the reverse of the findings of Harpalani and Schraufnagel (1990).

Levine (1996) noted three main problems associated with strain gauges: 1) strain gauges may not adhere properly to the coal, 2) they may not deform homogeneously with the coal, and 3) the length of time required for equilibration caused by restrictions of the coal sample size could be very long. He found that some samples required exposure times as long as 200 hours (over eight days), with larger samples requiring even longer equilibration times; and any measurement errors due to the lack of equilibration would result in measurements lower than reality.

Zutshi and Harpalani (2004) presented coal volumetric strain data collected using strain gauges attached to coal samples of unspecified dimensions. The coal samples were placed in vessels and pressurized with various gases. Equilibration times for these tests were long; resulting in total time of about 220 days to collect five data points. Chikatamarla et al. (2004) also recently reported strain measurements using strain gauges and although they do not mention the amount of time needed for equilibration, they did report problems with gas reacting with the strain gauges forcing an early termination of some experiments.

Mazumder et al. (2006) attached strain gauges to sample surfaces to measure sorption-induced strain under differing confining stresses. Their experimental design was necessarily somewhat more complicated than others' using strain gauges to measure freestanding strain. The results of Mazumder et al. indicated a potentially quicker stabilization of sorption processes than either Harpalani and Schraufnagel or Seidle and Huitt, but the experiments were run under different stress conditions so direct comparisons are difficult.

Sorption-induced strain quantifies how much the coal matrix swells or shrinks as gas is adsorbed or desorbed. Another potential problem associated with strain gauges is finding a matrix block—a cleatless volume of coal—large enough to attach the gauges. Measured strain would not be entirely representative of matrix strain if a sample contained cleats, but would be clouded by the contribution of cleats to the total strain. Finding cleat-free matrix samples would be easier if smaller samples could be used.

Volumetric and one-dimensional strain.

Levine (1996) presented both longitudinal and volumetric strain for high-volatile bituminous Illinois coal using both carbon dioxide and methane showing that volumetric strain is roughly three times the longitudinal strain. Robertson and Christiansen (2006) point out that Levine's results demonstrate, at least on a matrix block scale, that coal is isotropic in nature with respect to sorption-induced strain. With an isotropic medium, sorption-induced strain in a single dimension can be directly converted to volumetric strain.

Measurement of sorption-induced strain in one dimension can be a much simpler process than measurement of volumetric strain. Robertson and Christiansen (2005) described a new apparatus from which one-dimensional sorption-induced strain in coal was obtained much more quickly than methods using resistance-type strain gauges. Because strain gauges were not used, the sample size used in their experiments could be smaller—ensuring that the measurements were done using cleatless samples. They compared their results to data collected using strain gauges and found that both data sets compared favorably.

Experimental strain and modeling for gas mixtures.

Gray (1987) reported that sorption-induced strain in coal varied linearly with gas pressure for both CO₂ and CH₄. Harpalani and Schraufnagel (1990) showed that sorption-induced coal strain was not necessarily a linear function of gas pressure, but might be non-linear with decreasing gas pressure. Seidle and Huitt (1995) noted that their strain tests yielded curves resembling sorption isotherm curves and opined that sorption-induced strain was linearly correlated with sorbed gas content.

Robertson and Christiansen (2005) showed conclusively that strain induced by the adsorption of pure gases could be modeled using the same type of Langmuir equation used to model gas adsorption in coal. Extended-Langmuir theory (Yang 1997) suggests that strain induced by the adsorption of gas mixtures can be predicted from the Langmuir constants defining the shape of the pure gas strain curves. The strain constants (S_L and p_{SL}) for pure gases should be able to be used to calculate the strain curve for a mixture of these gases by applying extended Langmuir theory.

Simple Langmuir theory assumes that there is an ideal localized monolayer and that there is no interaction between adsorbed molecules. The following equation is an extended version of the Langmuir equation modified for strain instead of adsorption that accounts for multiple components in the gas phase:

$$S_{mix} = \frac{p \sum_{i=1}^n \left(\frac{S_{L_i} y_i}{p_{S_{L_i}}} \right)}{1 + p \sum_{i=1}^n \left(\frac{y_i}{p_{S_{L_i}}} \right)}, \dots\dots\dots(3)$$

where S_{mix} is the coal strain predicted by the adsorption of a gas mixture, p is the free gas pressure, i represents each pure gas component of the mixture, n is the total number of pure gas components, S_L is the Langmuir strain constant for each pure gas component, p_{SL} is the Langmuir pressure constant for each pure gas component, and y is the mole fraction of each pure gas component. According to this equation, if the individual strain-pressure curves were known for each pure gas in the mixture, the strain-pressure curve for any mixture of these gases could be directly calculated.

The strains of coal samples were measured using a gas mixture of 51% N₂ and 49% CO₂ and plotted in Fig. 1 along with calculated strains using Eq. 3. The pure-gas parameters used in Eq. 3 were taken from Table 1.

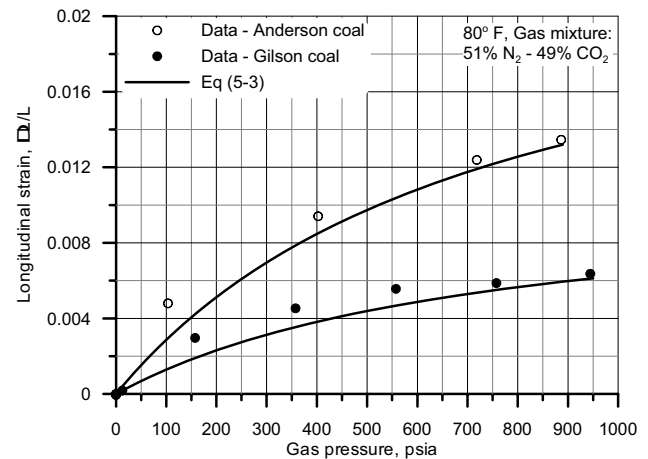


Fig. 1—Strain of Anderson and Gilson coal caused by the adsorption of a gas mixture of 51% N₂ and 49% CO₂. The data are modeled using Eq. 3 and the strain data for pure gases found in Table 1.

TABLE 1—LANGMUIR CONSTANTS FOR SORPTION-INDUCED STRAIN CURVES FOR ANDERSON AND GILSON COALS AT 80°F (AFTER ROBERTSON AND CHRISTIANSEN 2007).

Gas	Coal	Langmuir Constants for Strain Curves	
		S_L	p_{SL} , psia
CO ₂	Anderson	0.03527	555.25
	Gilson	0.01559	
CH ₄	Anderson	0.00931	886.03
	Gilson	0.00765	
N ₂	Anderson	0.00305	1119.93
	Gilson	0.00196	

Modeling the mixed gas strain data for both cores using Eq. (3) appears to be fairly accurate. However, both curves, however, under-estimated the amount of measured strain associated with adsorbing the gas mixture based on the strain curve of the pure gas components.

Yang (1997) presented a more sophisticated version of the extended Langmuir equation¹ than Eq. 3 that takes into account the lateral interactions among adsorbed molecules that could be used to more accurately model mixed gas strain:

$$S_{mix} = \frac{p \sum_{i=1}^n \left(\frac{S_{L_i} y_i}{\eta_i p_{S_{L_i}}} \right)}{1 + p \sum_{i=1}^n \left(\frac{y_i}{\eta_i p_{S_{L_i}}} \right)}, \dots\dots\dots (4)$$

where η describes the changes in interaction energies in the mixed adsorbates, which can either be greater or less than unity. If $\eta = 1$, Eq. 4 reduces to Eq. 3.

Because η accounts for the interaction energies between different species of gas molecules (not the adsorption energy between the substrate and the sorbing gas), it should be independent of the adsorbing substrate surface and should be a function only of the adsorbed gas. Once the gas interaction values are determined, they should be applicable to mixed-gas strain values for different coals. The strain data for the gas mixture are replotted in **Fig 2** along with the two extended Langmuir strain models—Eq. 3 and Eq. 4. Optimal values for η_{N_2} and η_{CO_2} of 0.25 and 0.58 respectively were determined for the Gilson coal data and then applied to the Anderson coal data.

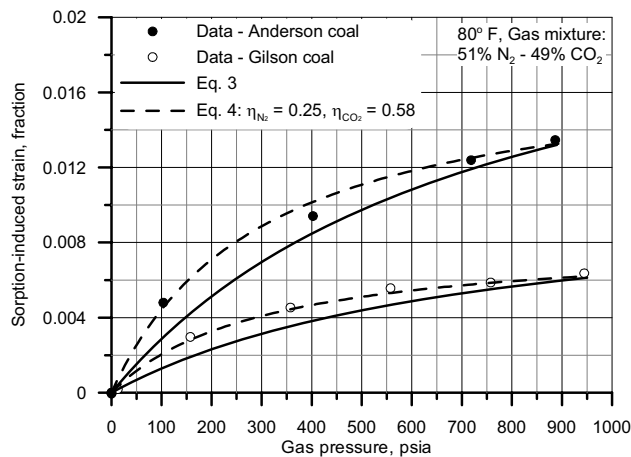


Fig 2—Strain data for Anderson and Gilson coals induced by the sorption of a mixed gas modeled by two different versions of the extended Langmuir strain equation: one neglecting interaction energies between molecules and the other accounting for molecule interaction.

The mixed gas isotherm model (Eq. 4) matched the both datasets much better than Eq. 3.

Permeability model.

Robertson and Christiansen (2006) describe a new permeability equation derived for sorption-elastic media such as coal specifically for confining condition found commonly in the laboratory, but not in the field. This model can be especially useful when dealing with laboratory experiments where many of the other factors that cloud field measurements are eliminated. Their permeability model can be re-written in the following form:

$$\frac{k}{k_0} = \exp \left[\underbrace{3c_0 \frac{1 - \exp(\alpha \Delta p_p)}{-\alpha}}_{\text{fracture compressibility}} + \underbrace{\frac{9(1-2\nu)}{\phi_0 E} \Delta p_p}_{\text{matrix compressibility}} - \underbrace{\frac{9}{\phi_0 (p_{SL} + p_{p_0})} \ln \left(\frac{p_{SL} + p_p}{p_{SL} + p_{p_0}} \right)}_{\text{sorption of gases}} \right] \dots\dots\dots (5)$$

Robertson and Christiansen defined the terms of Eq. 5 as: k/k_0 is the permeability change ratio, c_0 is the initial cleat compressibility, α is the cleat compressibility change rate, Δp_p is the change in cleat pressure, ϕ_0 is the initial porosity of the cleat system, ν is Poisson's ratio (matrix block), E is Young's modulus (matrix block), S_L and p_{SL} are the sorption-induced Langmuir strain parameters for a matrix block defined above, p_p is the cleat pressure, and p_{p_0} is the initial cleat pressure.

The first term describes the fracture compressibility complement to the permeability ratio, the second term describes the complement of the matrix compressibility, and the third term describes the complement of sorption-induced strain to the permeability ratio.

Presentation of Data and Results

The Robertson-Christiansen permeability model for sorption-elastic media such as coal shown in Eq. 5 was applied to coal permeability data measured in the laboratory under hydrostatic confinement pressures. The permeability data was taken from Robertson and Christiansen (2007).

Parameters for each coal that are required for the model are listed in **Table 2** (with sorption-induced strain parameters taken from Table 1).

TABLE 2—PARAMETERS USED TO MODEL THE PERMEABILITY CHANGES IN THE COAL CORES.		
	Anderson	Gilson
Rank	Subbituminous	High-volatile bituminous
Initial fracture compressibility, C_0 (psi^{-1})	1.16E-04	8.34E-05
Fracture compressibility change rate, α (psi^{-1})	2.17E-03	3.90E-03
Initial porosity, ϕ_0 (fraction)		0.015
Poisson's ratio, ν (fraction)		0.339
Young's modulus, E (psi)		393,500

Using the parameters from Table 1 and Table 2, measured permeability data with nitrogen being injected into the cores was modeled and results are shown in **Fig. 3**. The confinement pressure was held constant at 1000 psia

¹ Eq. (3.8) on page 51 of Yang (1997).

throughout the measurements. The model matched the measured data for nitrogen very well for both coal ranks.

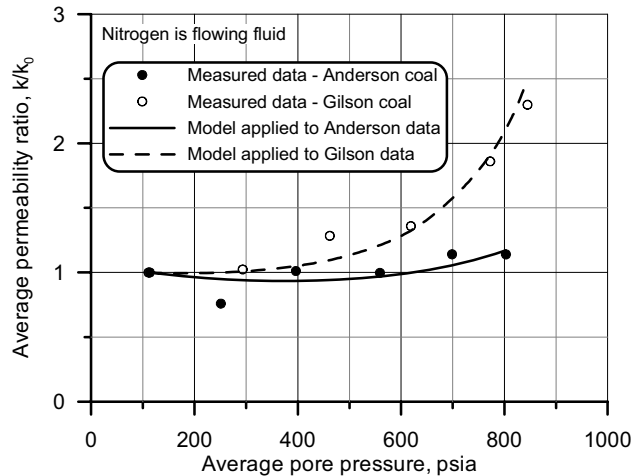


Fig. 3—The Robertson-Christiansen permeability model applied to measured coal permeability data with N_2 as the flowing fluid.

Fig. 4 shows permeability data and the model results for the same coal cores using methane as the flowing fluid. The only difference between the two sets of experiments (Fig. 3 and Fig. 4) was the composition of the flowing fluid. Methane had a higher Langmuir strain value (around 0.8%) for both coals than did nitrogen (around 0.25%). This figure shows that the permeability model underestimated the permeability as the pore pressure increased from 100 psia to 800 psia.

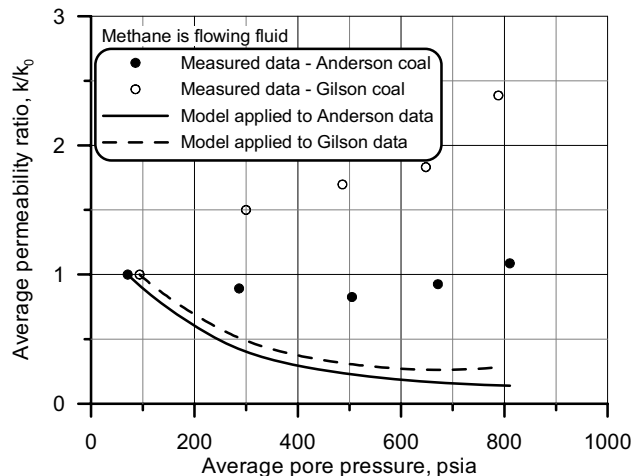


Fig. 4—The Robertson-Christiansen permeability model applied to measured coal permeability data with CH_4 as the flowing fluid.

The adsorption of CO_2 has a much larger impact on matrix strain than either nitrogen or methane in freestanding cores. Table 1 shows that the Langmuir strain value for CO_2 with the high-volatile bituminous coal from the Gilson seam is about 1.5% and the CO_2 Langmuir strain for the subbituminous coal from the Anderson seam is about 3.5%. Fig. 5 plots the change in permeability for the

same two coal cores but with carbon dioxide as the flowing fluid. The model vastly underestimated the changes in measured permeability ratio when CO_2 was injected through both ranks of coal as the flowing pore pressure was increased from 100 psia to 800 psia.

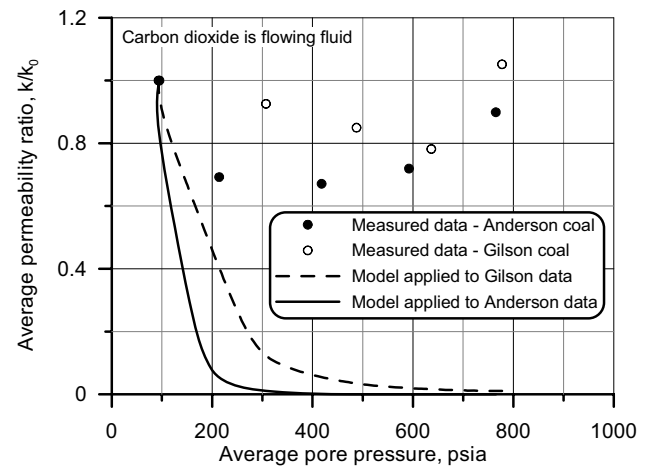


Fig. 5—The Robertson-Christiansen permeability model applied to measured coal permeability data with CO_2 as the flowing fluid.

Discussion

Just as with gas content isotherms, sorption-induced strain is a strong function of the adsorbed gas components as well as the coal rank and probably other factors such as temperature, etc. If accurate permeability models are to be developed, more sorption-induced strain data need to be available in the open literature. Strain gauge measurements can be very time consuming and difficult to perform, which most surely relate to why so few data are available. The method for obtaining sorption-induced strain data proposed by Robertson and Christiansen (2005), may provide a more rapid alternative to strain gauges.

Advantages of stress-free sorption-induced strain data.

As mentioned above, two types of experimental designs have been used when gathering sorption-induced strain data: one using freestanding (stress-free) samples and the other placing the samples under some confining force (stressed). This section discusses some of the advantages and disadvantages of each method.

Gray (1987) specifically states that unconstrained (stress-free) strain data is appropriate for use in his coal permeability model: "the strain derived from testing samples of coal that are free to move in all directions . . . can be directly used in calculations where strain is related to varying equivalent sorption pressure." Other permeability models (Sawyer et al. 1990; Palmer and Mansoori 1998; Pekot and Reeves 2003; Shi and Durucan 2003) do not specifically state that unconstrained strain data is appropriate for use in their models, but they cite stress-free strain data (Harpalani and Zhao 1989; Harpalani and Schraufnagel 1990; Levine 1996) for use in their models, from which one could deduce that they, along with Gray,

believed that the use of stress-free strain data was appropriate for use in their permeability models.

Robertson and Christiansen (2007) argue that because of permeability models' poor fit of permeability data, stress-free sorption-induced strain data could be used, but should be modified before being inputted into permeability models to account for the depression of sorption-induced strain caused by partially confined matrix blocks.

As discussed earlier in this paper, stressed sorption-induced strain data requires a more complicated experimental setup and stress-free data. However, stress-free methods using strain gauges can still be cumbersome measurements requiring inordinate amount of time to complete. The optical method of measuring stress-free sorption-induced strain data proposed by Robertson and Christiansen (2005) appears to be a viable alternative to strain gauge methods and can be used to collect large amounts of sorption-induced strain data much more rapidly.

Constrained sorption-induced strain data may appear initially to represent field conditions better than stress-free data. However, the stress conditions that exist in the field may not be exactly reproducible in the laboratory. Additionally, the collected constrained strain data would not be easily translated other stress conditions and its wider use may be limited. On the other hand, freestanding sorption-induced strain data could be manipulated to represent any number of perceived stress conditions and would be much more widely applicable.

Freestanding sorption-induced strain is probably not representative of what occurs in the field or in the laboratory when the sorption of gases causes changes in permeability. However, freestanding strain can be modified to represent a wide range of real-world physics. Stressed or confined strain tests may or may not truly represent conditions during field or laboratory permeability tests and modifying the resulting data so it applies to different stress conditions might not be as straightforward as modifying freestanding strain data.

Constrained sorption-induced strain measurements are usually done in connection with permeability tests (Mazumder 2007) so that strain and permeability can be made simultaneously on the same sample. The presence of cleats in the coal sample is necessary for permeability measurements, but cleats should be avoided when measuring matrix swelling/shrinkage.

Permeability model discussion

The data and model results plotted in Figures 3 through 5 show that as freestanding sorption-induced strain increases, model accuracy decreases. The presented model matches the lightly adsorbing nitrogen permeability data very well, but the more adsorbing the gas (greater the strain), the more inaccurate the permeability model became. The permeability experiments were necessarily done under a confining stress applied to the outside area of the core. The model deviated from the measured permeability data probably because the actual sorption-induced strain was less than the measured freestanding strain.

Conclusions

In an isotropic medium such as coal (on a small scale), one-dimensional sorption-induced strain can be used to calculate volumetric strain.

An optical method for measuring one-dimensional strain appears to be a more rapid and simple alternative for obtaining freestanding sorption-induced strain compared to the traditional method of using strain gauges.

An improved mixed gas adsorption isotherm model based on the extended-Langmuir theory has been shown to correctly model mixed gas sorption-induced strain based on pure gas strain data and a parameter accounting for gas-gas interactions that is independent of the coal substrate.

Freestanding sorption-induced strain data may be more desirable than constrained strain data because it can be easily modified to represent a wide range of physical conditions; whereas constrained data appears to be applicable to only one set of conditions, which may or may not truly represent the desired conditions.

The permeability equation used to model measured permeability data was less accurate as Langmuir-strain values increased. This was probably because actual sorption-induced strain was less under the constrained conditions of the test than the freestanding strain used in the model.

More sorption-induced strain data, as well as permeability data is necessary to develop a more accurate understanding of how stress affects sorption-induced strain in coal and to ultimately develop a more accurate permeability model.

Acknowledgments

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Nomenclature

- C_0 = Initial compressibility of the coal cleat system, psi^{-1}
- E = Young's modulus, psi
- i = a counter representing each pure gas species
- k = permeability, md
- k_0 = initial permeability, md
- n = total number of pure gas species
- p = pressure, psi
- p_L = Langmuir pressure, psi
- p_p = pore pressure, psi
- p_{SL} = Langmuir strain pressure, psi
- S = Sorption-induced strain, dimensionless
- S_L = Langmuir strain, dimensionless
- S_{mix} = Sorption-induced strain caused by adsorption of a gas mixture, dimensionless
- V = volume of adsorbed gas, scf/ton of coal
- V_L = Langmuir volume, scf/ton of coal
- y = mole fraction of each pure gas component, dimensionless
- η = parameter describing interactions molecules of different gas species, dimensionless

- α = cleat compressibility change rate, psi^{-1}
 ϕ_0 = initial porosity of coal cleat system, fraction
 ν = Poisson's ratio, dimensionless

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