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Project Title: **VIABILITY OF CO₂ SEQUESTRATION AND METHANE
PRODUCTION IN ILLINOIS COAL**

ICCI Project Number: 02-1/6.1A-4
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ABSTRACT

The overall objective of this research effort was to carry out a preliminary study to determine the feasibility of Illinois coal to produce coalbed methane, and further establish its suitability for CO₂ sequestration. Two properties of coal, namely the sorption characteristics, and impact of gas flow/substitution on the physical structure of coal and its effect on flow behavior, were studied. As a final step, a preliminary simulation exercise was carried out to determine the methane production potential of Illinois coal.

As expected, sorption results indicate that the affinity of coal for CO₂ is higher than that for methane. The results further indicate that injection of CO₂ enhances the recovery of methane, particularly after initial desorption has already occurred. The interesting finding is that the injection does not need to be at high pressures. Low pressure injection is equally effective in displacing the adsorbed methane. However, there seems to be an uncertainty about the exact mechanism responsible for the enhancement, whether it is a reduction in the partial pressure of methane or the preferential sorption property of coal. The results of the experimental work involving measurement of coal matrix volumetric strain clearly indicate that the coal matrix “swells” with injection of CO₂. The swelling is significantly greater than that for methane. Numerical analysis suggests that this matrix strain can result in a significant reduction in the fracture porosity of coal and its permeability. In fact, with continued injection, the fracture permeability is reduced so significantly that the flow starts taking place purely through the coal matrix, i.e., it becomes matrix flow as opposed to fracture flow.

Based on the findings of this study, it is concluded that injection of CO₂ should be practiced in combination with the primary depletion technique when the gas production becomes too low. This would enable recovery of methane that is otherwise left behind. CO₂ injection should be carried out at low pressures. This will not only have an economic advantage, but it will also significantly reduce any damage to the permeability of coal. Furthermore, the effectiveness of low pressure injection in displacing the methane is not hampered by low pressure injection. Finally, even at low pressures, coal is capable of holding a fairly large amount of CO₂.

EXECUTIVE SUMMARY

Carbon sequestration in geologic media, particularly deep and unmineable coal, is currently considered the only alternative that is feasible in the immediate future for storage of carbon dioxide. Coupled with the potential of enhancement of methane recovery from gas producing coalbeds, it offers an economic advantage over all other methods. Hence, the primary focus of this research effort was to carry out a preliminary study to determine the technical feasibility of Illinois coal to produce coalbed methane (CBM), and further establish its suitability for enhanced coalbed methane (ECBM) recovery by injecting CO₂ thus improving the recovery of CBM while simultaneously sequestering CO₂.

Two important properties of coal, both impacting the implementation of ECBM technology in Illinois, were investigated in this study. Since the basis of ECBM is the sorption characteristics of coal, primarily its higher affinity for CO₂ compared to methane, this property not only determines the amount of CO₂ that can be sequestered, but also the amount of additional methane that is displaced by CO₂. On the other hand, injection of CO₂, due to its higher sorptivity, results in a change in the microstructure of coal due to the “swelling” of the coal matrix and the accompanying reduction in the fracture aperture of coal thus resulting in a serious loss of permeability. The work described in this report is the result of a laboratory investigation currently underway to study these two properties. The report also describes the results of the numerical analysis carried out to determine the impact of the coal matrix volumetric strain on flow properties of coal, namely its porosity and permeability. Finally, the report concludes with the results of a preliminary simulation exercise carried out to determine the methane production potential of Illinois coal. The results of the experimental study, along with information obtained from the Illinois State Geological Survey, were used as input for the simulation. Unfortunately, extensive simulation could not be carried out as a part of the work reported here due to the complex nature of such a simulation, and lack of available data for Illinois coal. This effort will continue in the future.

The results of the sorption experiments clearly showed that the affinity of coal for CO₂ is significantly higher than that for methane. The results further indicated that injection of CO₂ enhances the recovery of methane, particularly after partial desorption has already occurred. The amount of methane released as a result of CO₂ injection is substantial even when the total system pressure is fairly high. Furthermore, the amount of methane released is the same when injection is carried out at low pressures. Hence, low pressure injection is equally effective in displacing the adsorbed methane. The experimental results of pure gas sorption fit the Langmuir model extremely well. The results of binary sorption experiments fit the Extended Langmuir model, commonly used to describe binary sorption, well although the model is more accurate for prediction of CO₂ adsorbed and not so for predicting the amount of desorbing methane. Finally, there seems to be an uncertainty about the exact mechanism responsible for the enhancement. It may be a reduction in the partial pressure of methane, preferential sorption property of coal, or a combination of these two effects.

The results of the experimental work involving measurement of coal matrix volumetric strain clearly indicate that the coal matrix “swells” with injection of CO₂. The swelling is significantly greater than that for methane. This is in line with the fact that CO₂ is substantially more sorptive than methane. In fact, the volumetric strain trend resembles that of the sorption isotherms established suggesting a linear relationship between the amount of gas adsorbed and the strain induced by it. Using the measured volumetric strain, a numerical analysis was carried out to determine the changes in fracture porosity of coal and its permeability. A bundle of matchsticks model was used to represent the physical structure of coal. The results, although very sensitive to the starting porosity of coal (initial porosity) which was only guessed, clearly show that the porosity of coal reduces very sharply with continued adsorption of CO₂. This change in fracture porosity translates to a drastic reduction in permeability. In fact, the analytical results suggest that the fracture permeability reduces to zero after injection beyond ~400, psi resulting in matrix flow alone. Matrix flow in porous media is usually lower than the fracture flow by orders of magnitude.

Based on the findings of this study, it is concluded that injection of CO₂ should be practiced jointly with the primary depletion technique when the gas production becomes low. This would enable recovery of methane that is otherwise left behind. Since the effectiveness of low pressure injection in displacing the methane is not hampered, consideration should be given to low pressure injection of CO₂. First, this would result in improved economics of the operation. Second, the damage to the permeability as a result of CO₂ injection would be reduced significantly. Furthermore, even at low pressures, coal is capable of holding a fairly large amount of CO₂. The sequestration potential is, therefore, not affected. However, this might not be possible because high pressure eases the entry and movement of gas in coal. This has been found to be the case in the deeper basins in the US. For potential sequestration site(s) in Illinois, this may be possible because of the relatively low depth of the coal seams, and due consideration should be given to this alternative.

Although ECBM recovery should be used in conjunction with the primary depletion method, for certain portions of Illinois coal, it might be necessary to practice ECBM for the entire life of producing reservoirs since the gas content is fairly low. In such cases, economics of sequestration should be evaluated carefully to determine if the amount of recoverable CBM is even viable on a commercial level.

Finally, since the release of methane appears to be primarily due to a reduction in partial pressure of methane, rather than the preferential sorption characteristics of coal, consideration should also be given to injecting a lower adsorbing gas, like nitrogen, which is known to enhance the recovery of methane, although the mechanism of “enhancement” is entirely different. In fact, injecting a mixture of nitrogen and CO₂ might be ideal since this would reduce/eliminate the cost of separation of CO₂ from power plant exhaust gases, which is a major cost of carbon sequestration. However, it is not certain that doing so would necessarily combine the advantages of both.

OBJECTIVES

Overall Objectives: The overall objective of this research effort was to carry out a preliminary study to determine the feasibility of Illinois coal to produce coalbed methane, and further establish the suitability of Illinois coal for CO₂ sequestration.

Specific Objective of the Study: In order to achieve the primary objective, the following specific objectives were selected for the first year of the effort:

- I. Study the sorption characteristics of Illinois coal in order to determine its ability to hold methane and CO₂, and establish the ability of CO₂ to enhance the recovery of methane.
- II. Measure the impact of gas flow/substitution on the physical structure of coal, and hence, the flow behavior of coal.
- III. Conduct a preliminary simulation exercise to determine the methane production potential of Illinois coal, coupled with CO₂ sequestration potential.

The tasks scheduled for the first year's effort are briefly described below:

Task I: Analysis of Coal Data from Illinois State Geological Survey: This task included obtaining the data developed by the ISGS as a result of their first drilling program. The data, taken from five wells drilled in the state, included depth intervals, gas content and results of gas analysis. The objective of this task was to identify the areas of Illinois coal that have adequate gas content to justify commercial CBM production in the future, and then review it further for suitability to serve as a CO₂ repository.

Task II: Procurement/Preparation of Coal Samples: This task included collecting blocks of coal samples from mines in Southern Illinois and preserving these in their native state to prevent weathering which results in formation of induced fractures. The objective was to test coal samples representative of *in situ* coal reservoir conditions. Preparation of whole samples included coring/cutting appropriate test specimens from blocks of coal, and for powdered samples for sorption experiments, chunks of coal were pulverized.

Task III: Sorption Experiments – CO₂ and Methane: This task included establishing sorption isotherms for the coal type to determine its ability to hold methane, release characteristics of methane, and the coal's ability to retain CO₂.

Task IV: CO₂/Methane Displacement Experiments: This task was aimed at determining if the injection of CO₂ into deep CBM reservoirs in Illinois had the ability to enhance the recovery of methane. The objective was to evaluate if the ultimate methane recovery is improved, and whether more methane is recovered during early stages of the reservoir's life, that is, determine the level of "enhancement" of methane recovery. The task involved injecting CO₂ during the desorption part of the sorption cycle and measuring the amount of methane released.

Task V: Changes in Physical Structure of Coal: This task was intended to determine if injecting CO₂ into a CBM reservoir resulted in a significant change in the physical structure of coal. The primary objective was to measure the volumetric strain induced as a result of CO₂ sorption, and its impact on cleat porosity and permeability. This task involved measuring changes in the coal matrix volume as a result of adsorption of methane/CO₂ under constant conditions of temperature, pressure and external conditions.

Task VI: Simulation Exercise: This task included using the experimental results and data from the ISGS to carry out a preliminary simulation exercise to evaluate the potential of long-term CBM production, CO₂ sequestration, and the difference in production with and without CO₂ injection for Illinois coal. A commercial CBM simulator, with the added capability of simulating the enhanced recovery scenario, was used for this. The primary objective of this task was to develop adequate expertise to use the simulator efficiently and effectively for future simulation needs.

Task VII: Communication: This task included dissemination of the results to as large a community as possible by communicating the findings to potential CBM developers and researchers at the Carbon Sequestration Forum/Conference.

INTRODUCTION AND BACKGROUND

Enhancement of oil recovery from oil-bearing rocks by injection of carbon dioxide (CO₂) is a standard practice [1]. Over the last decade, there has been an effort to use this technique to enhance the recovery of coalbed methane (CBM) from coal reservoirs. The effort has been augmented since the technique provides a means to sequester CO₂, a greenhouse gas, in deep and unmineable coals. In fact, this technique is currently considered the only practical and feasible alternative for CO₂ storage while other technologies are being studied in detail. Coalbed reservoirs are considered to have good potential to sequester large quantities of CO₂ because of the ability of coal to adsorb CO₂, the ease of availability of deep reservoirs throughout the world, and their proximity to power plants which are considered to be the main source of CO₂ emissions [2]. Also, a considerable amount of knowledge has been acquired, and technologies and models developed in the area of coalbed methane recovery, all of which can be easily adapted to CO₂ flow and storage. Thus, the concept of CO₂ sequestration, coupled with enhancement of coalbed methane recovery to serve as an incremental energy source, is considered to provide a good synergy with long-term benefits, one being environmental and the other economical.

Carbon sequestration by injection of CO₂ into deep coal is still in its primary stage and, as of now, the process is not fully understood, primarily due to a lack of available data. Field experience with CO₂ injection is limited to the Allison Unit (operated by Burlington Resources) in the San Juan Basin, Upper Silesian coal basin of Poland (RECOPOL project), and Alberta Sedimentary Basin of Canada (by Alberta Research Council) [3, 4, 5]. RECOPOL and Alberta projects are still in their development phase, although some preliminary assessment has been completed. The Allison unit CO₂ injection operation

started in 1995 and continued for over three years. Analysis of the injection data collected clearly showed that the potential to sequester CO₂ in CBM reservoirs is significant due to the higher affinity of coal for CO₂. The data further demonstrated that production of methane, both ultimate as well as rate of recovery, is enhanced significantly. Finally, the analysis of data showed a significant reduction in coal permeability, thus negatively affecting injectivity and, hence, the economics of the technique. Further testing and field demonstration are, therefore, needed to validate the results of the analysis. For successful implementation of this enhanced coalbed methane recovery (ECBM)/CO₂ sequestration technology, it is critical to understand several basic phenomena, two of which are as follows:

1. The mechanism of binary adsorption of methane and carbon dioxide on coal, the preferential sorption property of coal, and the ability of carbon dioxide to displace methane from coal. There have been a few laboratory scale experiments to study the enhanced methane recovery, along with the filed pilot-scale projects mentioned above, to study its application. Several researchers have studied the adsorption of methane and CO₂ mixture at different composition, temperature and pressure conditions to improve the understanding of the sorption mechanism [6, 7, 8, 9, 10]. Although these studies have all demonstrated the ability of CO₂ to improve methane recovery from coals, there are still several uncertainties regarding multi-component sorption behavior of coal.
2. It is well known that there is a significant change in the volume of coal matrix associated with desorption/adsorption of gas. This, in turn, results in a wide variation in the permeability of coal thus impacting continued production [11, 12, 13]. The understanding of the influence of this volumetric change, and its impact on the flow behavior of coal is a relatively new phenomenon in methane recovery from coalbeds. A number of studies, theoretical, numerical and experimental, have been conducted in the past to assess the changes in matrix volume due to desorption of methane. These studies have concluded that matrix shrinkage resulting from desorption of methane causes the coal to shrink, and results in opening up of the cleat system causing the permeability to increase [11, 12, 13, 14, 15, 16, 17]. However, there have been no studies to investigate the matrix “swelling” effect resulting from injection of CO₂ and its influence on coal permeability. There is a strong belief that there is such interdependence since CO₂ is significantly more sorptive than methane [8]. Field observations to date support this suspicion. A complete understanding of the interaction of CO₂ with coal, displacement of methane by CO₂ because of its ability to sorb preferentially, the impact of adsorption on the physical properties of coal and its permeability is, therefore, essential in order to predict the long-term effects of CO₂ injection.

Based on the brief background presented above, this experimental study was initiated to 1) better understand the sorption mechanism of coal in the presence of binary mixture of gases, and investigate whether methane recovery with CO₂ injection is, in fact, enhanced; and 2) estimate the changes in flow characteristics of coal as a result of volumetric strain associated with CO₂ injection.

EXPERIMENTAL PROCEDURES

Since two somewhat independent experiments were conducted as a part of this study, they are described in this section under two separate sub-headings.

Sorption Experiments

Sample Preparation: It is a standard practice to use powdered samples for sorption experiments in order to reduce the duration of the experiment by minimizing the distance that the gas molecules must diffuse prior to entering the “free” phase. Pieces of coal were first broken into lumps approximately one cm in size. These were ground in a ball mill and the resulting powdered coal was sieved to obtain the desired sample size of 40-100 mesh (0.425-0.149 mm). The pulverized samples were kept in air-sealed bags to prevent oxidation. Prior to sorption experiments, approximately 80 grams of the sample was taken and placed in the environmental chamber maintained at 73⁰F and 96% relative humidity for moisture equilibrium. It required approximately 24 hours for the sample to become fully saturated with moisture. After the sample attained equilibrium moisture, one gram was taken out for moisture and ash analysis, while the rest was used for the sorption experiment. The standard ASTM procedures D 3173 – 87 and D 3174 – 97 were followed for moisture and ash analysis respectively.

Setup and Procedure: The experimental setup design is based on the method of mass balance, utilizing the volumetric analysis method. It uses the gas expansion technique to measure the quantity of sorbed gas. Real gas equation is used to calculate the gas volumes for a single gas. The experimental setup consists of a stainless steel fixed volume cylinder (FV) with a standard volume, and another stainless steel sample container (SC), which holds the powdered coal sample. The two cylinders are separated by a valve. The schematic diagram of the setup is shown in the Figure 1. Since the process of sorption is very sensitive to temperature, the setup is placed in a constant temperature bath, which maintains temperature to within 0.2⁰C of the set value. A pressure transducer is attached to the fixed volume cylinder. A filter is placed between the two cylinders to prevent the fine coal sample from being blown away from the sample container into the fixed volume. Gas is allowed to expand from the fixed volume into the sample container, or from the sample container to the fixed volume. By monitoring the pressure before and after the expansion, volume of gas moving into, or out of, the SC is calculated. Prior to every sorption experiment, the calibration of the setup is carried out to determine the void volume (V_v) in the sample container by expanding helium gas from FV to SC, and measuring the equilibrium pressure.

To establish the adsorption isotherm in this study with a pure gas, the procedure was fairly standard as that used in the gas expansion volumetric technique. The constant temperature bath was set to a fixed value to simulate the reservoir temperature. Gas was then let into the FV and pressure, P_1 , was recorded. Next, the gas in FV was allowed to expand into the sample container. Once the gas pressure stabilized, pressure, P_2 , was recorded. For single component isotherms, minimum equilibrium time of 16 hours was allowed. Using P_1 , P_2 , volume of FV and the void volume (V_v) in SC, the adsorbed gas

volume was calculated using the real gas equation. The above step was repeated for increasing pressure steps up to a final pressure of ~1500 psi for methane and ~700 psi for CO₂. Once the highest pressure for the adsorption isotherm was reached, desorption isotherm was obtained using the process in reverse, the principle remaining the same. The FV was first bled out partially. After recording the pressure of gas in FV, the gas in SC was allowed to expand into the FV. The gas pressure was recorded after attaining equilibrium. This procedure was repeated for decreasing pressure steps until the pressure in the sample container was reduced to approximately 100 psi, at which time the experiment was terminated.

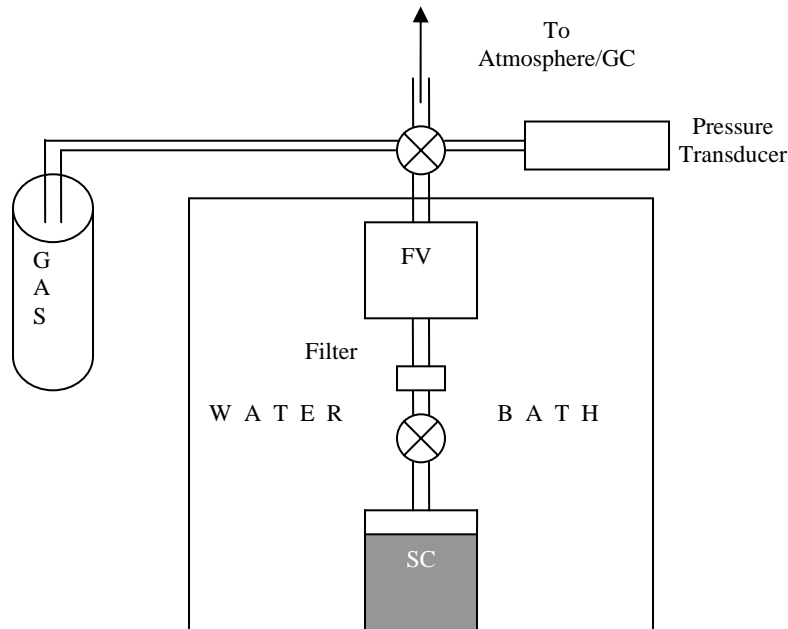


Figure 1: Schematic diagram of the experimental setup to establish adsorption isotherms.

For the second phase of the experimental work involving CO₂ injection, the adsorption procedure was completed following the steps described above up to a pressure of 1500 psi for pure methane. The usual desorption steps were performed until the SC pressure came down to ~500 psi. The FV cylinder was then evacuated and CO₂ was filled into it at a pressure of ~500 psi. After the pressure stabilized in FV, the valve between FV and SC was opened and the gas in the two containers was allowed to mix. After equilibrium, the final pressure was recorded and a part of the gas was passed through the GC to determine its molar composition. Using the measured molar composition, the partial pressures of the two gases were calculated. This allowed further calculation of the volume of each gas adsorbed/desorbed.

Changes in Physical Structure of Coal

Sample Preparation: Rectangular specimens (prisms) were cut from a block of coal using a handsaw. The samples with the least cleats were selected for experimental work. Subsequent to preparation, the test specimens were kept in an environmental chamber under controlled conditions of temperature and humidity.

Experimental Setup and Procedure: The experimental setup for the study was designed to enable measurement of volumetric strain due to changes in gas composition while keeping the total gas pressure constant. A schematic of the experimental setup is shown in Figure 2. The main components of the setup were pressure vessels capable of withstanding very high pressures, a data acquisition system to monitor strain, and a gas chromatograph (GC) to measure the composition of gas mixtures in the pressure vessels. As shown in the diagram, the setup consisted of three pressure vessels, capable of testing three coal samples simultaneously. Since sorption is very sensitive to temperature, the pressure vessels were placed in a constant temperature bath. Three strain gages were affixed to the surface of each sample in order to monitor strains in the three orthogonal directions. The gages were affixed using an epoxy recommended by the manufacturer. After attaching the gages, leads were attached to each strain gage. The entire assembly was then placed in the pressure vessel with the required outlets for gage connection to a data acquisition system. This procedure was repeated for all three test samples.

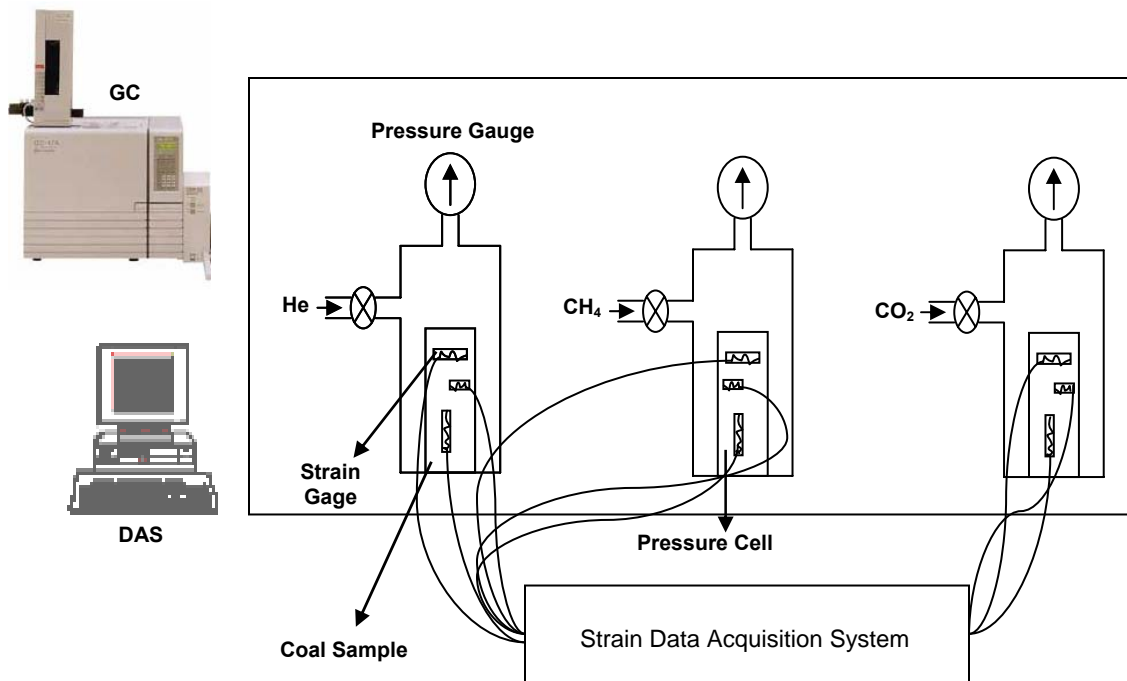


Figure 2: Schematic of the experimental setup to measure the sorption induced volumetric strain.

All three samples were first subjected to increasing helium pressure in steps of 400 psi and volumetric strain was measured at equilibrium for each step. Since helium is a non-adsorptive gas, the measured strain is purely due to the mechanical compression of the solid coal resulting from the change in pressure. After attaining equilibrium at 1500 psi, one of the samples was left alone to serve as the control test. The second sample was subjected to increasing concentration of methane, maintaining the total pressure constant at 1500 psi. This was achieved by injecting methane while bleeding helium/(helium + methane) mixture out every time methane was injected. This ensured that the solid and fractures in the samples were subjected to the same external conditions throughout the

experiment, the only difference being the composition of the gases within and around the sample. The procedure was continued until the gas in the vessel was pure methane. Hence, the sample was subjected to varying composition of methane, all the way from 0% to 100%. At each equilibrium step, a sample of gas was analyzed using the GC and concentration of methane was measured. For the third sample, the same procedure was followed using CO₂ instead of methane.

RESULTS AND DISCUSSION

Task I: Analysis of Coal Data from Illinois State Geological Survey

Excellent contact was maintained with the ISGS personnel throughout the project duration. Data was obtained from their first drilling program aimed at determining the potential of commercial CBM production in the state of Illinois. ISGS drilled five wells continuously cored to depths of 500 to 1200 feet. Fifty-nine coal samples were obtained and analyzed. The findings are summarized below:

- The gas content of recovered coal varied between 35 and 175 cu ft per ton of coal. As expected, gas content was found to increase with rank and depth. Coalbed methane exploration should, therefore, target deeper and more mature coals.
- A conservative estimate of CBM resource gives 21-25 trillion cu ft (TCF) for the Illinois Basin of which ~14 TCF are expected to occur in Illinois.
- Analysis of high gas content areas showed that 80-92% of the recovered gas is methane, with small mounts of CO₂, the rest being nitrogen. The PI believes that nitrogen is in free state and will drain out as soon as flow of gas starts, posing no problems with long-term production of methane.

For CO₂ sequestration, the ideal target coalbeds are ones that are deep, not mineable in the foreseeable future, and have an impermeable caprock. Based on the current level of information, this criteria is best met by the Herrin and Springfield seams. Hence, it was decided that sequestration work at this time should concentrate on the above two seams.

TASK: II: Procurement/Preparation of Coal Samples

Blocks of coal were collected from surface mining operations mining the Springfield and La Plata seams. Samples prepared from the latter were used initially to test the experimental setup. These blocks were kept under water to prevent oxidation during transportation and storage in the laboratory until it was time to prepare the test specimens. The blocks were taken out for a minimum period of time required for specimen preparation. After preparation, the samples were stored in an environmental chamber under controlled conditions of temperature and humidity. Brief details of the exact specimen preparation procedures are included in the **Experimental Procedures** section for the sake of clarity.

Towards the end of the project period, two cores from the Springfield and Herrin seams were obtained from the second drilling program of the ISGS. Hence, a majority of the

results presented in this study are based on tests carried out on coal samples not truly representative of coal where any CBM activity is likely to occur in the near future.

TASKS III and IV: Sorption and Displacement Experiments

Pure Gas Sorption: Langmuir theory [18], the one most commonly used model for the study of adsorption of gases on coal, was used for this study. The main assumptions of the Langmuir theory are:

1. Every adsorbed molecule is held at definite, localized site.
2. Each site can hold one and only one molecule.
3. The surface of the adsorbent is energetically homogeneous.

The adsorption isotherm derived by Langmuir is given as:

$$V = \frac{V_L P}{P + P_L} \quad (1)$$

where, V = Sorbed volume (ml/g) at pressure P (psi), and V_L (ml/g) and P_L (psi) are the Langmuir coefficients known as Langmuir Volume and the Langmuir Pressure respectively. Both Langmuir Volume and Langmuir Pressure have important physical significance from the coalbed methane recovery point of view. The constants, V_L and P_L , are determined by plotting experimental pressure (P) versus pressure/volume (P/V) results and calculating the slope and intercept of the best fitting lines.

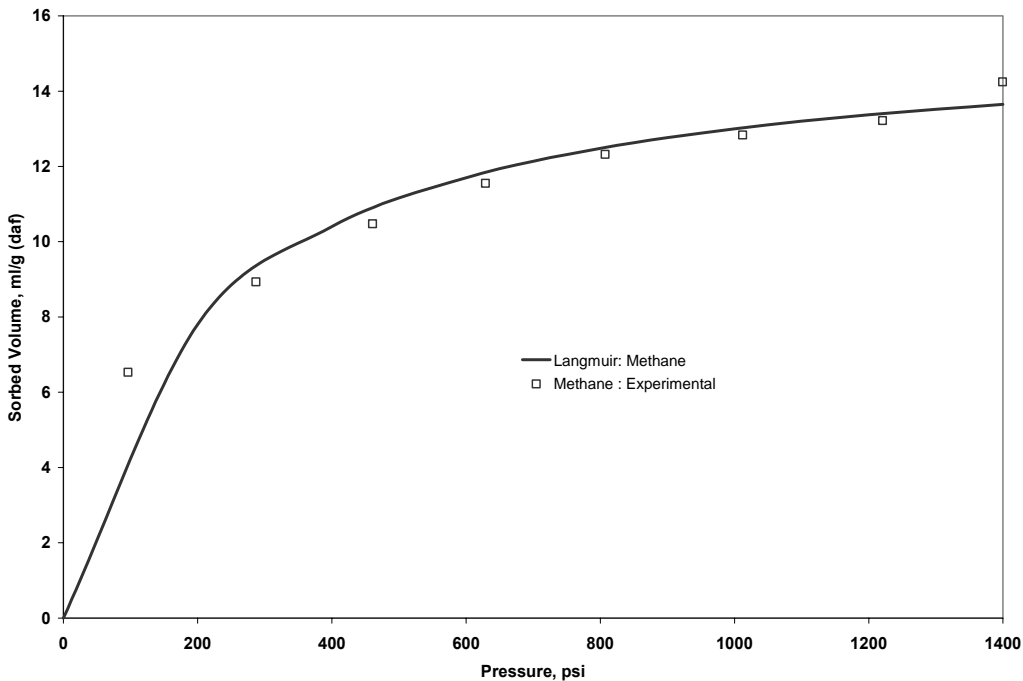


Figure 3: Adsorption isotherm for Herrin coal.

The experimental data for methane and CO₂ were analyzed using the Langmuir Equation and adsorption and desorption isotherms were established. It was observed that adsorption and desorption followed the same path, i.e., there was no hysteresis between the two isotherms. This reversibility of the adsorption process shows that adsorption of methane and CO₂ on coal are physical in nature. The methane isotherm for the sample taken from the Herrin core is shown in Figure 3. V_L and P_L constants were calculated to be 15.6 ml/g and 200 psi respectively. As a common practice, the sorption results presented are on a dry and ash free basis. Analytical results showed the moisture and ash content to be 7.7% and a 3% respectively. It should be borne in mind that these numbers are based on one test only.

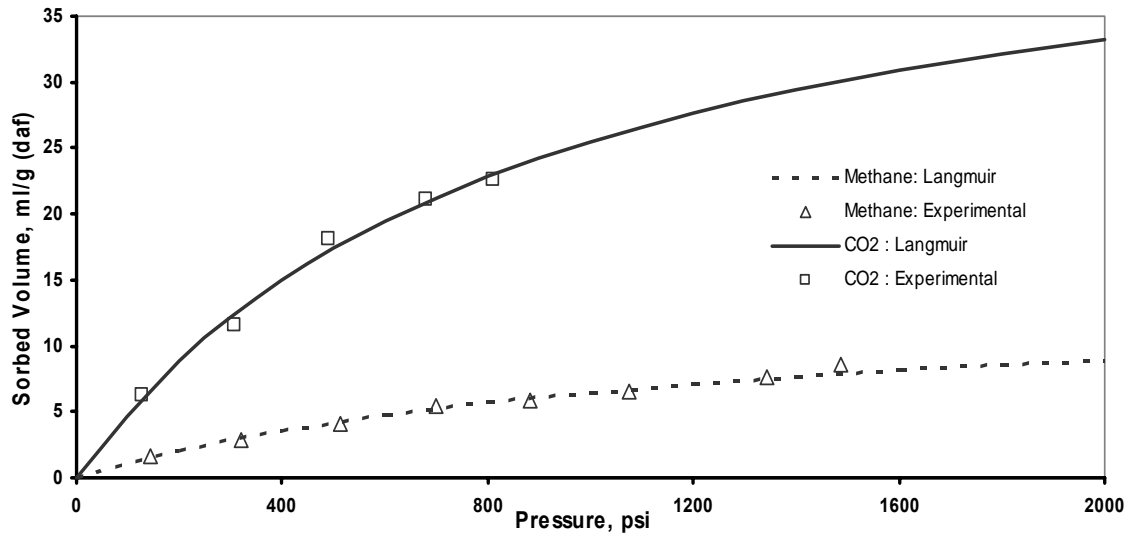


Figure 4: Langmuir isotherms for methane and carbon dioxide.

For samples taken from elsewhere, the equilibrium adsorption isotherm of methane and carbon dioxide, along with the experimental results, are shown in Figure 4. It is apparent that the sorption isotherms for methane and carbon dioxide exhibited Type I behavior according to the Brunauer classification of isotherms [19]. As expected with this type of isotherm, adsorption of methane and CO₂ on coal approaches a limiting value of gas as the free gas pressure increases and reaches the saturation pressure. This is logical since the adsorption on coal takes place in micropores and attains the limiting value as the micropores fill. The V_L and P_L values for methane based on the test completed were 14.4 ml/g, and 1235 psi respectively. The V_L and P_L values for CO₂ were calculated to be 43 ml/g and 590 psi respectively. Figure 4 also clearly shows that the Langmuir model provides a very good fit to the experimental data with the coefficient of correlation (R^2 value) for methane of 0.98, and for carbon dioxide, 0.96. As expected, the results clearly show that coal has a greater sorption affinity for CO₂ than methane. It can adsorb three times more CO₂ than methane. The high adsorption ratio has good implications for ECBM and carbon sequestration. It may be possible to sequester as much as three times more CO₂ than methane produced by ECBM. The P_L value for CO₂ is less than half of that for methane suggesting that coal adsorbs the bulk of CO₂ at pressures below 500 psi.

It may, therefore, be possible to sequester large amounts of CO₂ at relatively low pressure.

CO₂ Injection: CO₂ injection was performed at a pressure of approximately 500 psi. Adsorption of methane was initially completed to a pressure of 1487 psi. This was followed by desorption to a pressure of 500 psi in five steps. The amount of sorbed methane for this pressure change decreased from 8.6 to 5.0 ml/g. At this stage, CO₂ was injected into the sample container at a pressure of 504 psi. After attaining equilibrium, the total pressure decreased to 493 psi, with methane partial pressure at 205 psi and CO₂ partial pressure at 288 psi. The sorbed methane reduced to 2.1 ml/g. The amount of desorbed CH₄ as a result of injection was, therefore, 2.9 ml/g, while the amount of adsorbed CO₂ was 10.3 ml/g. In the subsequent step, CO₂ was injected at 489 psi. After injection, the total pressure remained at 486 psi, while the partial pressures of methane and CO₂ changed to 83 psi and 403 psi. The amount of additional desorbed methane at this stage was 1.1 ml/g, while additional CO₂ adsorbed was 3.6 ml/g. The methane sorption results for this test are shown in Figures 5 and 6. Figure 5 shows the adsorbed methane as a function of the partial pressure of methane while Figure 6 shows it as a function of the total system pressure. It was calculated that for 4 ml/g of methane desorbed, 14 ml/g of CO₂ was adsorbed by the coal. The ratio of CO₂ adsorbed to methane desorbed for this coal is, therefore, approximately 3.5: 1.

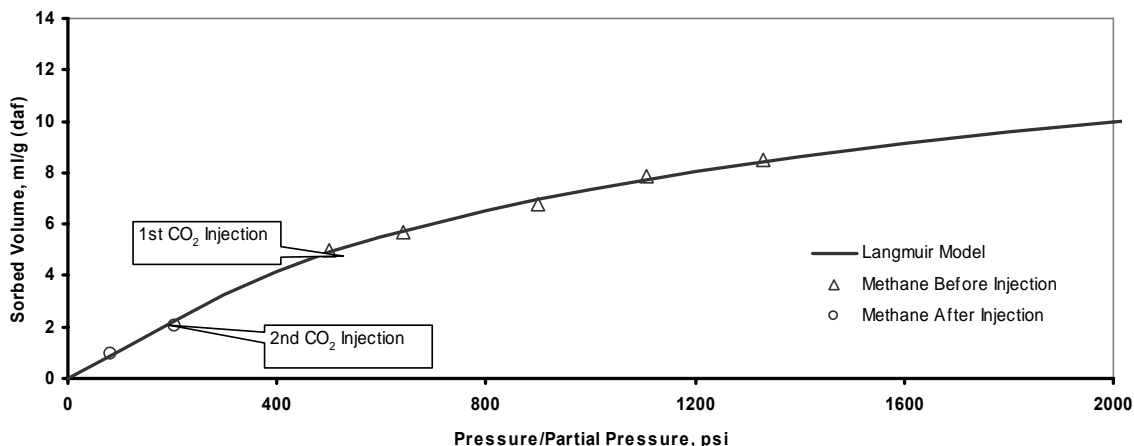


Figure 5: Displacement of methane as a result of CO₂ injection.

It is obvious from Figure 6 that with two injections of CO₂ at approximately 500 psi, almost all of the sorbed methane was released even when the total system pressure was still fairly high, 500 psi. However, Figure 5 suggests that the adsorption of methane still followed the Langmuir isotherm and depended only on the partial pressure of methane. Hence, desorption of methane was only due to reduction in the partial pressure of methane and not because of any preferential adsorption of CO₂ and desorption of methane in the presence of carbon dioxide, as has been suggested quite often [8]. However, the results of a second test completed in the laboratory indicate that the actual amount of methane desorbed is greater than the desorption resulting from the reduction of partial pressure of methane, as determined by the Langmuir isotherm, clearly suggesting that the incremental methane produced is because of methane getting preferentially desorbed due

to the presence of CO₂. Hence, it is not clear whether the observed enhanced methane recovery is due to partial pressure reduction, or due to preferential adsorption of CO₂. It is likely that the two mechanisms complement each other in enhancing the recovery of methane.

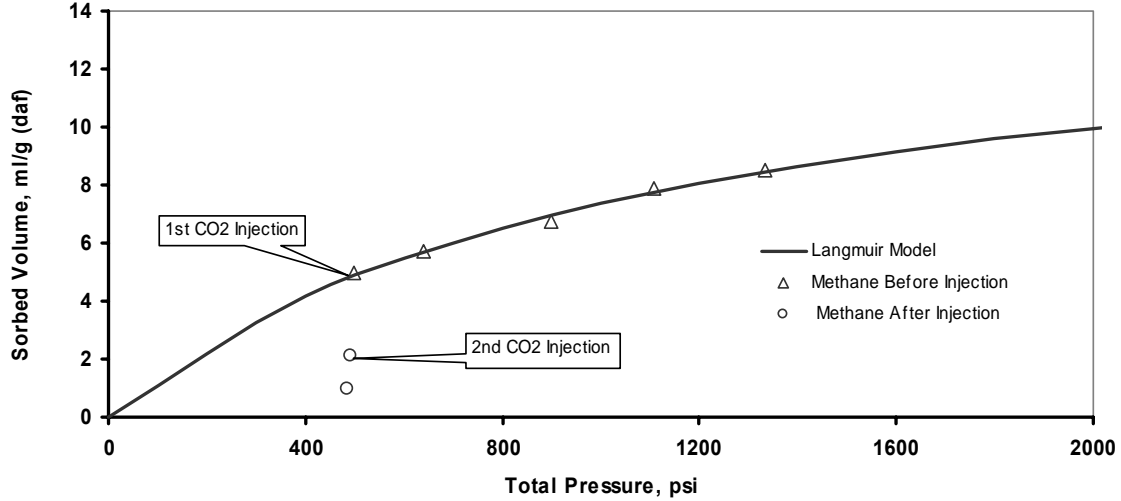


Figure 6: Displacement of methane as a result of CO₂ injection.

Extended Langmuir Model: The Extended Langmuir (EL) equation is the simplest and most commonly used model for the prediction of mixed gas adsorption on coal [20]. In order to predict the binary adsorption equilibria, it requires pure component isotherm data. The EL equation is given as:

$$V_i = \frac{(V_L)_i b_i P_i}{1 + \sum_j b_j P_j} \quad (2)$$

where, V_L and $b(=1/P_L)$ are the pure gas isotherm Langmuir constants, and $P_{i,j}$ are the partial pressures of individual gas in free-gas phase. The partial pressure is related to the total pressure by the relation:

$$P_i = P y_i \quad (3)$$

where, y_i is the gas phase mole fraction of the component i . The relative adsorption of the two components is calculated by estimating the separation factor or the selectivity ratio. The selectivity ratio of a binary gas adsorption system is defined as:

$$\alpha_{ij} = \frac{(x/y)_i}{(x/y)_j} \quad (4)$$

where, x and y are the molar composition of a component gas in the adsorbed and free phase respectively. For the EL isotherm, the separation factor is simply the ratio of the adsorption equilibrium constants [8 and 21]:

$$\alpha_{ij} = \frac{(V_L b)_i}{(V_L b)_j} \quad (5)$$

The EL equation was used in order to predict adsorption equilibria of binary mixture of methane and carbon dioxide resulting from the injection of carbon dioxide into the coal-methane system. The predicted values were compared with the actual experimental value. Figure 7 shows the sorption isotherm of methane along with the predicted adsorbed phase equilibria of methane and CO₂ mixture, and compares it with the actual experimental results of the first test. The adsorbed CO₂ is within 3 percent error band of the EL predicted value. The measured adsorbed CH₄ is within 10 percent error band of the predicted value. Also, it can be observed that EL model is under-predicting the methane sorbed value, and over-predicting the sorbed CO₂ value. The selectivity of carbon dioxide over methane is 4.3:1. Thus, the EL model is very accurate in predicting sorbed CO₂, while it is only moderately accurate in predicting sorbed methane. This is consistent with the observation of Reeves [22] that the EL model predicts more accurately the more strongly adsorbed component. Also, the selectivity ratio for the tests is not very accurate as compared to the experimental CO₂/methane sorbed ratio.

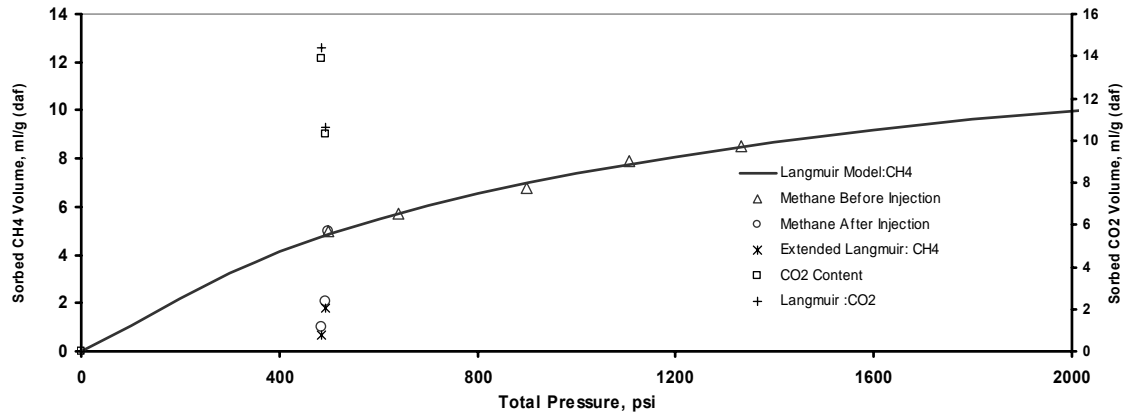


Figure 7: Binary equilibrium adsorption.

TASK V: Changes in Physical Structure of Coal

Experimental Results: The first part of this experimental phase involved dosing all samples with increasing amount of helium in pressure steps up to a maximum of 1500 psi while monitoring the strain continuously. A sample was considered to be in equilibrium if the strain remained constant for eight hours or more. This required three to four days for each pressure step. Using the measured strain for the helium cycle, the volumetric strain was calculated for each sample. Figure 8 shows the volumetric strain with increasing helium pressure. As expected, the volume of coal matrix decreased with increasing gas pressure due to compression of the coal grains. The matrix, or grain,

compressibility (C_m), defined as the change in the volume of solid grains as a result of changes in external pressure, is given mathematically as:

$$C_m = \frac{1}{V_m} \left(\frac{dV_m}{dP} \right) \quad (6)$$

where, V_m is the volume of solid coal and dP is the change in pressure. Using the measured strain for helium, this was calculated to be approximately $-2.07E-6 \text{ psi}^{-1}$. The results were similar for all three samples.

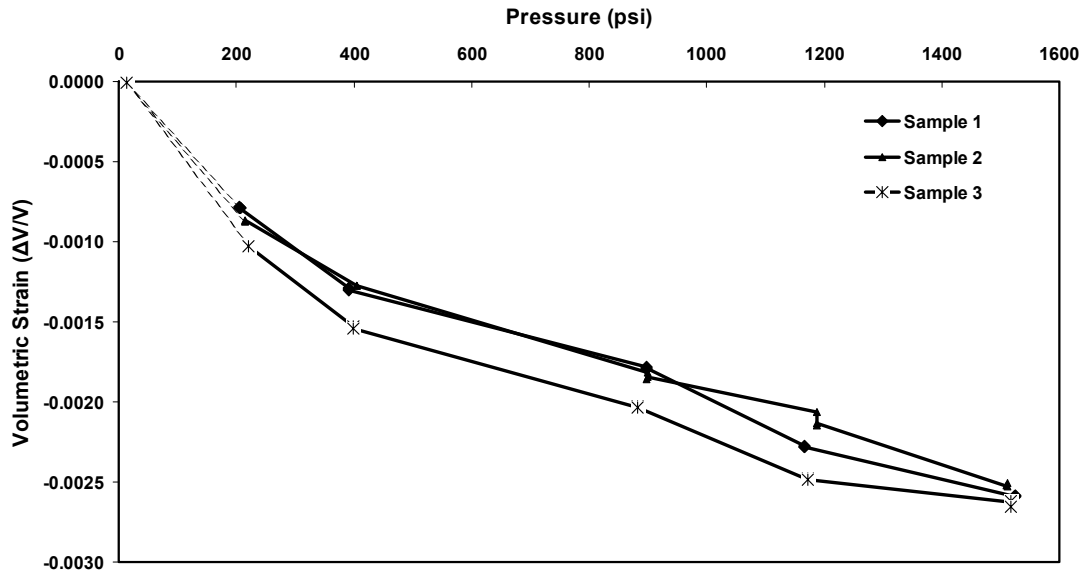


Figure 8: Volumetric strain of coal samples with increasing helium pressure.

For the second part of this experimental work, methane was injected into the second pressure vessel, maintaining the total pressure constant at 1500 psi. In the third vessel, helium was bled out to reduce the pressure to approximately 850 psi since the maximum CO_2 pressure in a gas cylinder is approximately 850 psi. Once the sample reached equilibrium at 850 psi, CO_2 was injected, once again, maintaining the total pressure constant. With subsequent injections, volumetric strain was calculated for all samples for each step. Figure 9 shows the volumetric strain for the three samples for different pressure steps. As expected, there is no further strain in the sample left alone for the duration of the experiment. It is also evident from the graph that the swelling due to carbon dioxide adsorption is significantly greater than that for methane. This is consistent with the fact that the sorption capacity of coal for CO_2 is significantly higher than that for methane [8]. Using the same approach as that used for estimating the matrix *shrinkage* coefficient, *swelling* coefficients were calculated for each sample. For methane injection, swelling coefficient was calculated to be $6.53E-6 \text{ psi}^{-1}$ for pressure up to 1075 psi. For CO_2 injection, swelling coefficient was calculated as $16.5E-6 \text{ psi}^{-1}$ for pressure up to 760 psi. Interestingly, the ratio of the two coefficients is 2.5, once again, suggesting a direct dependence of the quantity sorbed and the resulting strain.

On attaining equilibrium after each injection step, the concentration of methane and CO₂ in the samples was determined using a gas chromatograph. This was used to calculate the partial pressure of the two gases. The results are shown in Figure 10. The plot shows the change in volumetric strain due to increasing methane and CO₂ pressure in the samples. For pressures up to 725 psi, the volume of coal matrix increased by ~0.6% due to methane adsorption, and ~1.25% due to CO₂ adsorption.

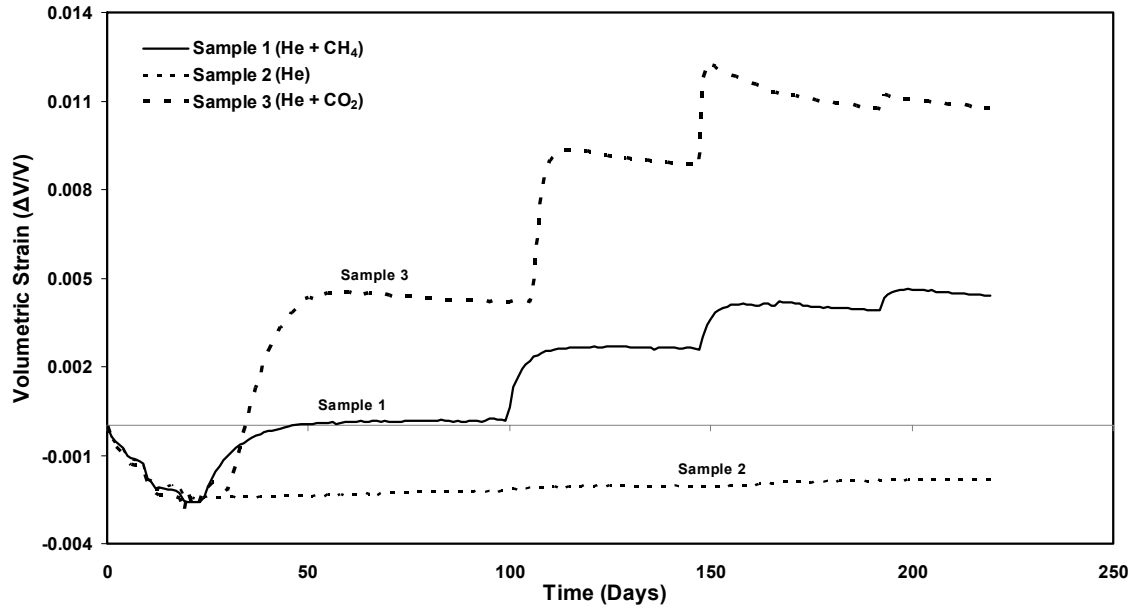


Figure 9: Volumetric strain of coal samples with time for various pressure steps.

Impact of Matrix Swelling on Cleat Porosity and Permeability: The swelling of the coal matrix due to CO₂ injection results in a change in cleat porosity, and hence, the cleat permeability. Following the procedure described by Harpalani and Chen [14], and briefly described below, changes in cleat porosity and permeability as a result of changes in the coal matrix volume were calculated. The increase in the coal matrix volume due to swelling was considered equal to the decrease in cleat aperture (b) as shown in Figure 11. This is a reasonable assumption since the total volume of coal *in situ* (matrix and cleats) remains constant with degasification, or injection of a second gas.

Once again, assuming a matchstick geometry [14] and, $a_1 = a_2$, the initial coal porosity ($\phi_{Initial}$) is given by $2b/a$. The change in matrix dimension, Δa , as a result of swelling depends on the change in pressure and swelling behavior of the coal, and is given by $\Delta a = a l_m \Delta P$, where a is the initial cleat spacing, l_m is the change in dimension of the coal matrix in the horizontal direction per unit pressure, and ΔP is the change in pressure of the gas being injected. Since the flow of the gas is considered along the cleat systems perpendicular to the bedding plane, change in dimensions along the vertical cleats only

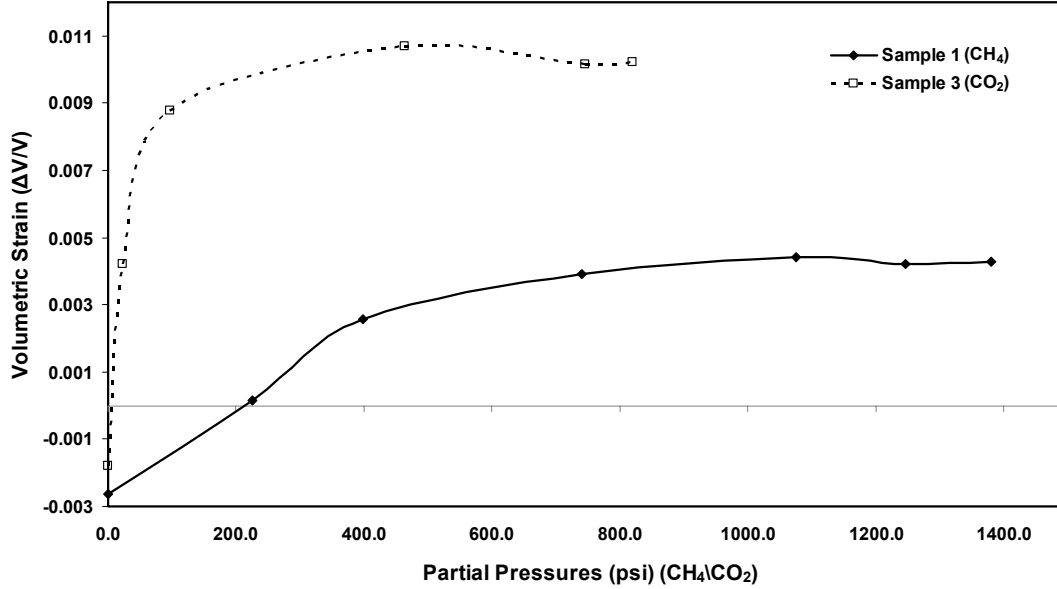


Figure 10: Volumetric strain of samples with respect to partial pressure of methane and CO₂.

(Figure 11) has been taken into account necessitating the introduction of the term l_m . After a pressure change of ΔP , the new cleat porosity (ϕ_{New}) can, therefore, be written as

$$\phi_{New} = \frac{2(b - \Delta a)}{(a + \Delta a)} \quad (7)$$

The ratio of the modified cleat porosity to the initial cleat porosity is then given as:

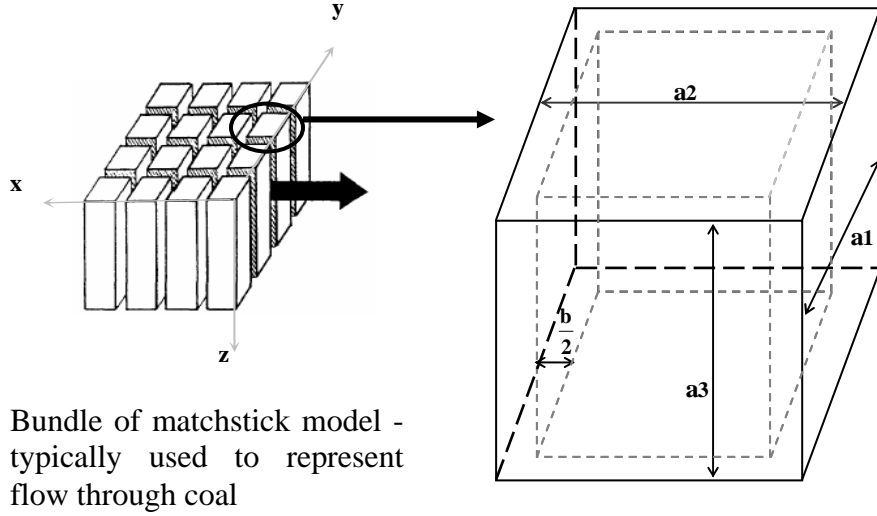
$$\frac{\phi_{New}}{\phi_{Initial}} = \frac{2(b - \Delta a)}{(a + \Delta a)} \bigg/ \frac{2b}{a} \quad (8)$$

Substituting for Δa and simplifying gives the following:

$$\frac{\phi_{New}}{\phi_{Initial}} = \frac{1 - (2l_m \Delta P / \phi_{Initial})}{(1 + l_m \Delta P)} \quad (9)$$

Hence, using estimates for initial porosity and the measured change in matrix dimensions of coal, change in cleat porosity with changes in pressure can be estimated. Considering the initial cleat porosity to be 1%, the change in porosity with change in pressure was estimated as shown in Figure 12. At CO₂ pressure of approximately 400 psi, for coal with an initial cleat porosity of 1%, the cleat porosity is reduced to zero. Negative values are obtained for higher pressures since the swelling is greater than the starting porosity of coal. However, since this is a physical impossibility, the results beyond this pressure can

be interpreted as suggesting that the flow is taking place through the coal matrix and can no longer be considered fracture or cleat flow. The results are being re-analyzed to ensure that this is, in fact, the case. Furthermore, the results clearly indicate a linear relationship between porosity ratio and the pressure of gas, i.e., with increase in CO₂ concentration in the coal, the porosity decreases linearly.



..... Initial Matrix Volume

— Swollen Matrix Volume after CO₂ Injection

For the matchstick geometry, the initial cleat porosity ($\phi_{Initial}$) of the coal if ($a_1 = a_2 = a$) is given by $\phi_{Initial} = \frac{2b}{a}$ where, b is the cleat aperture

Figure 11: Change in cleat geometry with change in matrix volume.

The change in cleat porosity as a consequence of change in matrix dimension results in a change in cleat permeability. For the same matchstick geometry, and considering the same assumptions, the initial cleat permeability ($k_{Initial}$) is given by $k_{Initial} = \frac{b^3}{12a}$. After a pressure change of ΔP , the new cleat permeability (k_{New}) can be written as:

$$k_{New} = \frac{(b - \Delta a)^3}{12(a + \Delta a)} \quad (10)$$

The ratio of the modified cleat permeability to the initial cleat permeability can then be written as:

$$\frac{k_{New}}{k_{Initial}} = \frac{(b - \Delta a)^3}{12(a + \Delta a)} \bigg/ \frac{b^3}{12a} \quad (11)$$

Substituting for Δa and simplifying gives the following:

$$\frac{k_{New}}{k_{Initial}} = \frac{\left(1 - \left(\frac{2l_m \Delta P}{\phi_{Initial}}\right)\right)^3}{(1 + l_m \Delta P)} \quad (12)$$

The variation in permeability as a function of the initial permeability is shown in Figure 13. As expected, at CO₂ pressure of 465 psi, the permeability decreases to zero. The results do suggest a continuous decrease in permeability ratio with increasing gas pressure, i.e., with increase in CO₂ concentration in coal, the permeability decreases continuously. This correlates well with the field observation at Burlington Resources' Allison Unit site [3] where a reduction of *in situ* coal permeability from 100 – 130 md to >1 md, a two orders of magnitude reduction, is suspected. However, these results indicating a decrease in permeability are due to changes in volume of coal matrix alone. Since the total pressure was kept constant throughout the experiment, the effective stress throughout the experiment was zero, and the impact of this effect has been neglected.

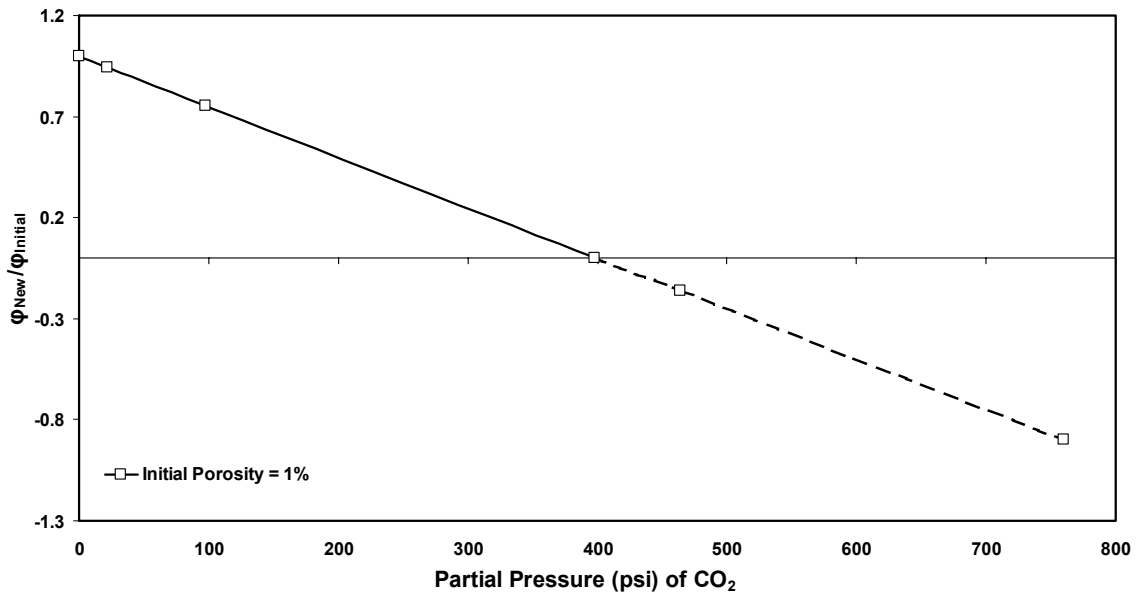


Figure 12: Estimated variation in cleat porosity due to change in matrix volume.

TASK VI: Simulation Exercise

This was one of the most challenging tasks of this study. Hence, the results do not show the advanced stages of simulation that the PI had anticipated completing at the beginning of this project. The CBM simulator, COMET, was procured in March 2003. It is an extremely complex software package and requires extensive training prior to use. It is basically a black oil simulation package, modified to work well for CBM operations, and further modified to simulate ECBM (injection of a second gas) scenarios. Finally, as expected, it requires too many input parameters. On the plus side, the simulator has

Initial Reservoir Conditions: Temperature: 75⁰ F
 Pressure: 500 psi
 Water Saturation: 75%

Figure 14 shows a typical production plot obtained from simulation. The curve can easily be divided into four segments. The first one shows an increase in gas production. This is the result of dewatering and reduction in water saturation in the cleats, and a corresponding increase in gas saturation. The second segment shows a slight decrease in production due to a decrease in permeability resulting from increased effective stress associated with depletion and reduction in pore pressure. The third segment shows increased production due to increased permeability resulting from matrix shrinkage. The last segment shows a slow but steady reduction in production rate as a result of depletion. Figure 15 shows the impact of increasing well spacing from 27 acres to 75 acres. For this well spacing, the third segment stretches significantly, and at the end of three years, the production rate is still increasing. Hence, with larger well spacing, higher production rates are achievable for longer periods of time. However, the cumulative recovery is reduced since only one well, instead of three, is producing.

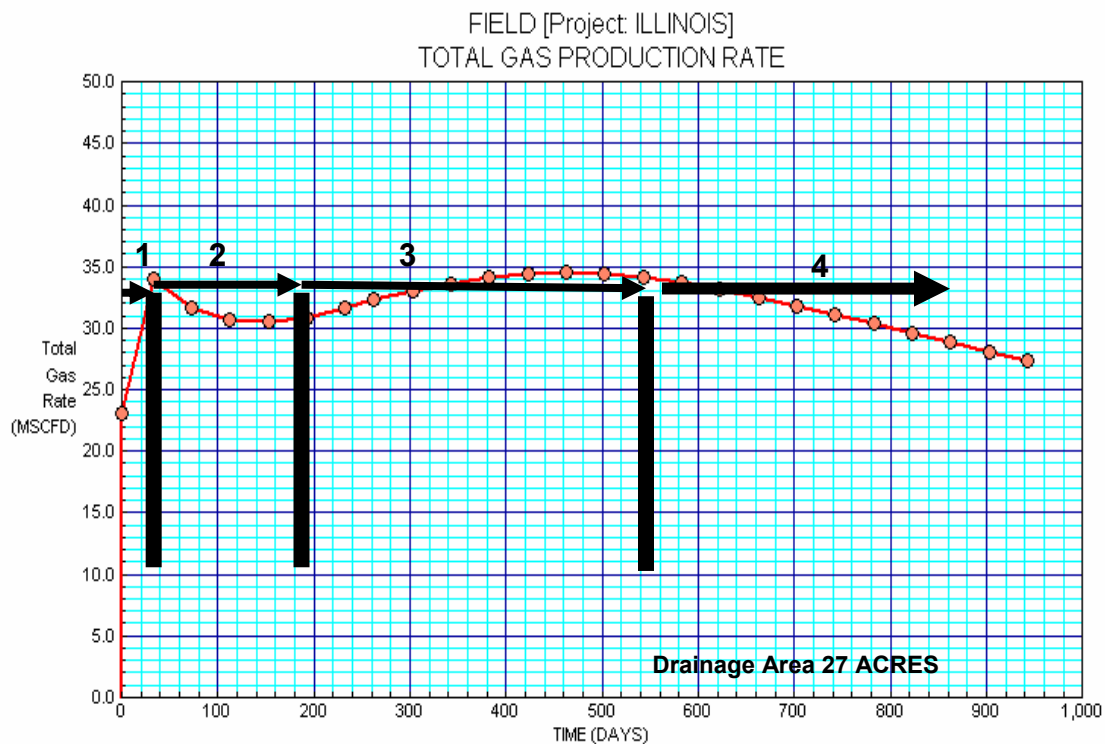


Figure 14: A generalized production profile for 3 years – 27 acres spacing.

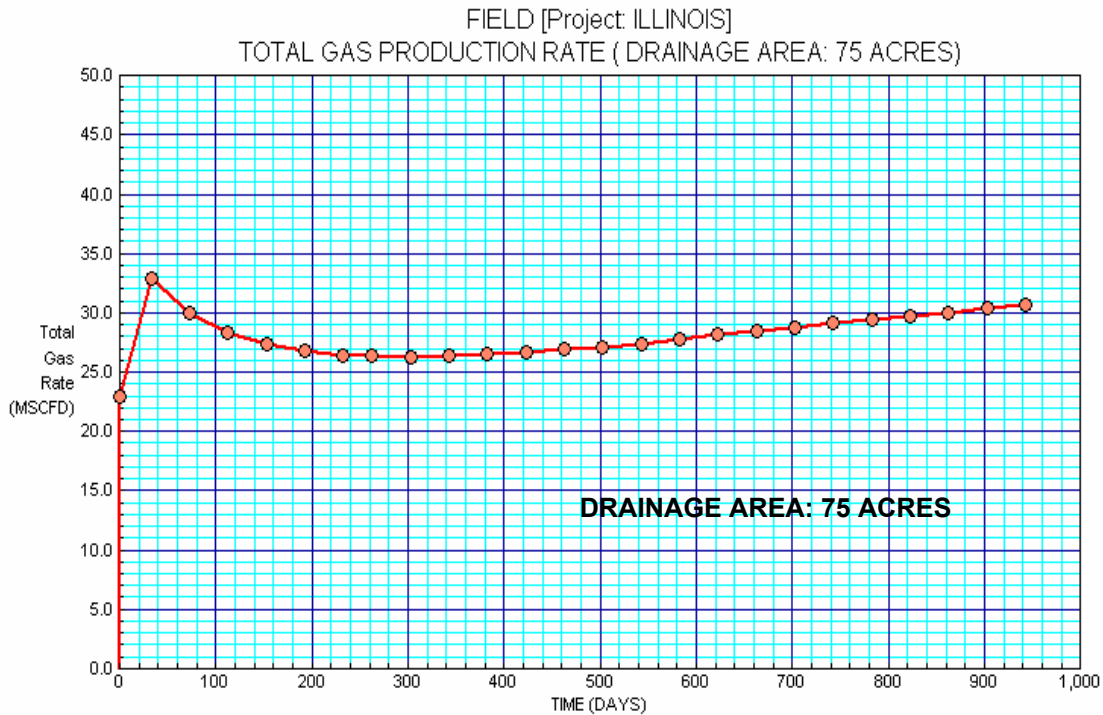


Figure 15: Production profile for 3 years – 75 acres spacing.

Figure 16 shows the impact of treating permeability as a variable over the life of the producing reservoir. This was input as a table giving changes in permeability as a percentage of the initial permeability for changes in pore pressure, i.e., with continued production. The increase in permeability is greater and it tends to remain higher in the long run.

Once again, this is simply an attempt to carryout a simulation for a completely hypothetical reservoir in Illinois. The ECBM scenario, where CO₂ is injected, could not be simulated during this project duration. However, since effort in this area continues, this will be done in the future.

TASK VII: Communication

The work carried out as a part of this project was discussed with other researchers and potential producers at every opportunity that arose in the last eighteen months. For the research community, two presentations will be made in May 2004 at the International Coalbed Methane Symposium to be held in Tuscaloosa, Alabama, and the Carbon Sequestration Conference to be held in Washington, D.C.

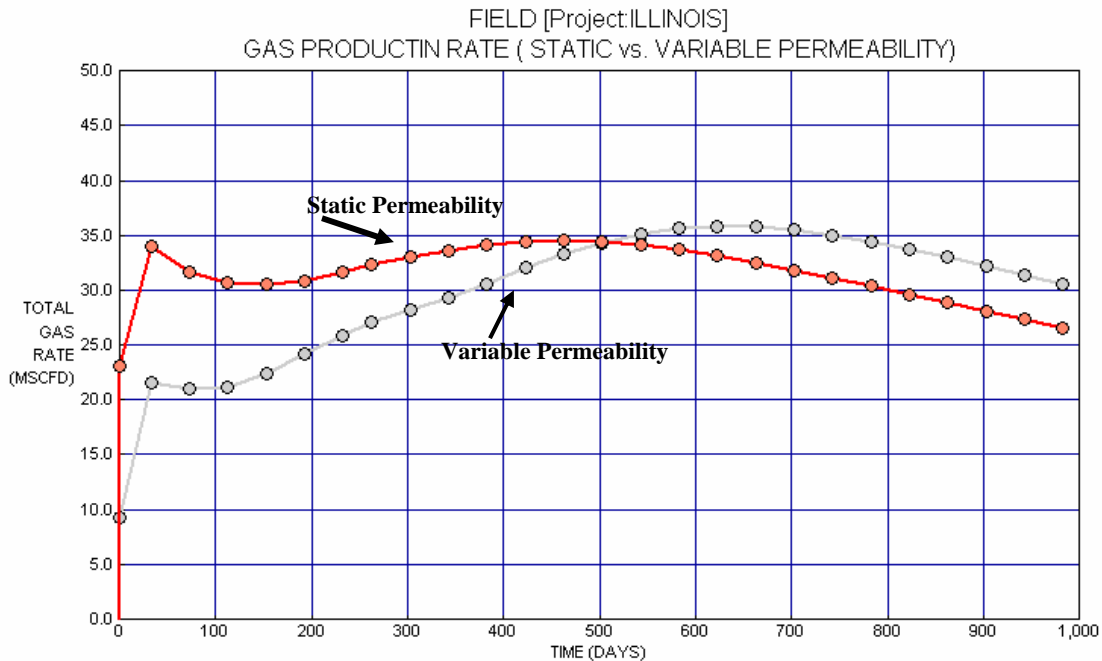


Figure 16: Production profile for 3 years with static and dynamic permeability.

CONCLUSIONS AND RECOMMENDATIONS

CBM is likely to play a large role in meeting the increasing gas demand in the US over the next few decades. Although Illinois has been overlooked in the past, it has potential to contribute to this demand. It is not known at this time if ECBM recovery has a potential in Illinois or not. The PI believes that this is because the gas content of Illinois coal is not as high as other basins, like the San Juan and Black Warrior. Illinois Basin, therefore, can not afford to leave a large fraction of gas-in-place behind as non-recoverable. Hence, ECBM has good potential although its application would depend on the availability of cheap CO₂ close to the injection sites, i.e., economic separation of CO₂ from power plant exhausts.

The specific conclusions based on the work completed to date are as follows:

1. The recovery of methane can be enhanced by injecting CO₂ into partially saturated coal. Relatively low pressure CO₂ injection was found to be fairly effective in the laboratory. This translates to injection at low pressures after partial depletion of CBM to improve the recovery of methane that is otherwise left behind and wasted. This might be problematic under field conditions since high pressure is needed to drive the gas into coal although this would depend on the depth of the injection site and permeability of coal at the point of injection.
2. The exact mechanism of methane recovery is not very clear. The enhancement is probably due to a combination of the effects of partial pressure reduction and

preferential sorption. Although based on limited observation, this is contrary to the general belief that it is the preferential sorption affinity of coal for CO₂ that is responsible for the enhancing mechanism. Hence, when a highly adsorbing gas is injected, it has all the advantages of inert gas stripping (nitrogen), and the added benefit of non-mixing since the injected gas does not mix with the recovered methane.

3. The EL model is very accurate in predicting sorbed carbon dioxide, but it is not so accurate in predicting sorbed methane. This is in agreement with the findings of previous researchers where EL was found to be more accurate for the higher adsorbing gas.
4. Injection of CO₂ results in a swelling of the coal matrix, which can have a profound effect on coal permeability. The study further facilitated estimation of the matrix *swelling* coefficient due to changes in gas composition alone.
5. The swelling induced due to CO₂ adsorption is more than twice that due to methane adsorption. This correlates well with the fact that CO₂ is significantly more sorptive than methane.
6. The major impact of this swelling is a decrease in cleat porosity of coal. Depending on the initial cleat porosity and adsorption pressure, the cleat porosity decrease to almost zero with continued injection. This decrease in coal porosity results in a decrease of the permeability to almost zero, once again, the time taken to reach this stage depending on the initial porosity. Hence, to the extent possible, injection of CO₂ should be carried out at low pressures only. This is contrary to the practice adapted at the Allison Unit where high pressure injection was carried out for three years even though the permeability of coal reduced by two orders of magnitude.

Based on the experience gathered during the project duration, both experimental and knowledge gained about Illinois coal, it is recommended that the following be pursued further:

1. Rigorous experimental studies should be conducted to better understand the phenomena involved in CO₂ injection as a means to enhance the recovery of CBM and its impact on flow behavior.
2. As a continuation of this effort, injection of CO₂ into the sample saturated with methane will be carried out to estimate the volumetric changes due to methane/CO₂ exchange.
3. This will be followed by an experimental investigation to study the coupled effect of stress and CO₂ induced volumetric changes in the coal matrix. The results will assist in estimating the effect of CO₂ injection, and the resulting changes in effective stress, on the volume compressibility and permeability of coal.

4. In the area of simulation, experimental studies should be carried out to obtain reasonable values of some of the input parameters. At this time, there is very little information available for Illinois coal. A few parameters that have a significant impact are the compressibility of coal (pore volume and fracture), relative permeability, and stress-dependence permeability.
5. The concept of low pressure injection should be pursued. This option not only has an economic advantage, but also results in lower reduction in permeability.
6. The main reason for not including the results of the injection experiments in this report extensively, although a significant amount of time was spent on this work, is that PVT (pressure-volume-temperature) behavior for the methane-CO₂ system is not well quantified at this time. Injection experiments are difficult to conduct because analysis of the results poses a serious computational complexity. Scientists at Oak Ridge National Laboratory (ORNL) are conducting studies at this time, and it is anticipated that the equation of state (EOS) for CH₄-CO₂-N₂ mixtures that would work will be developed.

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