Final Report

Geologic Sequestration of CO_2 in Deep, Unmineable Coalbeds: An Integrated Research and Commercial-Scale Field Demonstration Project

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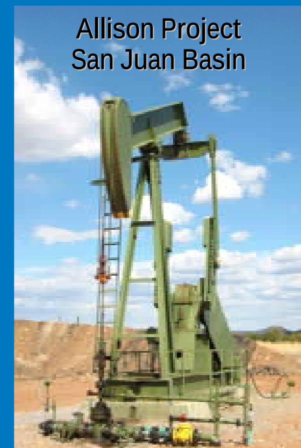

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Abstract

The Coal-Seq consortium is a government-industry collaborative consortium with the objective of advancing industry's understanding of complex coalbed methane and gas shale reservoir behavior in the presence of multi-component gases via laboratory experiments, theoretical model development and field validation studies. This will allow primary recovery, enhanced recovery and CO_2 sequestration operations to be commercially enhanced and/or economically deployed.

The project was initially launched in 2000 as a U.S. Department of Energy sponsored investigation into CO_2 sequestration in deep, unmineable coalseams. The initial project accomplished a number of important objectives, which mainly revolved around performing baseline experimental studies, documenting and analyzing existing field projects, and establishing a global network for technology exchange. The results from that Phase have been documented in a series of reports which are publicly available.

An important outcome of the initial phase was that serious limitations were uncovered in our knowledge of reservoir behavior when CO_2 is injected into coal. To address these limitations, the project was extended in 2005 as a government-industry collaborative consortium. Selected accomplishments from this phase have included the identification and/or development of new models for multi-component sorption and diffusion, laboratory studies of coal geomechanical and permeability behavior with CO_2 injection, additional field validation studies, and continued global technology exchange.

Further continuation of the consortium is currently being considered. Some of the topics that have been identified for investigation include further model development/refinement related to multi-component equations-of-state, sorption and diffusion behavior, geomechanical and permeability studies, technical and economic feasibility studies for major international coal basins, the extension of the work to gas shale reservoirs, and continued global technology exchange.

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1.0 Introduction

The Coal-Seq consortium is a government-industry collaborative consortium with the objective of advancing industry's understanding of complex coalbed methane (CBM) and gas shale reservoir behavior in the presence of multi-component gases via laboratory experiments, theoretical model development and field validation studies. This will allow primary recovery, enhanced recovery and CO_2 sequestration operations to be identified that can be commercially enhanced and/or economically deployed. The project was initially launched in 2000 as a U.S. Department of Energy (DOE) sponsored investigation into CO_2 sequestration in deep, unmineable coalseams. The initial project accomplished a number of important objectives, which are listed later, and for which complete reports are available for the interested reader.

An important outcome of the initial phase was that serious limitations were uncovered in our knowledge of reservoir behavior when CO_2 is injected into coal. To address these limitations, the project was extended into Phase 2 in 2005 as a government-industry collaborative consortium. In addition to U.S. DOE, the multi-national membership to the consortium include BP America, the CO_2-Cooperative Research Centre, ConocoPhillips, the Illinois Clean Coal Institute, Japan Coal Energy Center, Repsol YPF, Schlumberger, and Shell International Exploration & Production. Contractors performing R&D for the project include Advanced Resources International (program management, reservoir modeling, field studies technology transfer), Electrochemical Systems (equation-of-state and diffusion model development), Oklahoma State University (sorption model development), Southern Illinois University (core flood experiments), and Higgs-Palmer Technologies (geomechanical and permeability modeling). This paper describes the results and accomplishments achieved to date in the project, with particular focus on Phase 2, and describes some of the future activities being considered.

2.0 Phase 1 Results

Phase 1 (2000 – 2004) of the project accomplished a number of important objectives, which included:

- Performed detailed studies of two multi-well, multi-year enhanced coalbed methane recovery (ECBM) field pilots in the San Juan basin – the Allison Unit CO_2-ECBM pilot and the Tiffany Unit N_2-ECBM pilot[1,2].
- Created a field "best practices" manual based on the experience gained from those pilots[3].
- Performed study on geochemical reactions when injecting CO_2 into coal[4].
- Evaluated the applicability of commonly used isotherm models when applied to multi-component gaseous systems[5].
- Developed an improved model for predicting permeability changes in coal with CO_2 injection[6].
- Collected coal samples from most coal basins in the U.S. and created the first publicly-available database of CH_4, N_2 and CO_2 isotherms for these basins.
- Assessed the CO_2 sequestration and concomitant ECBM recovery potential of coal basins in the U.S[7].
- Developed a model for screening potential CO_2-ECBM/sequestration projects[8].
- Performed a technical and economic sensitivity study of ECBM[9].
- Participated in the design of the RECOPOL project in Poland[10].
- Facilitated global technology exchange and networking via the www.coal-seq.com website and annual Coal-Seq forums.

The results from Phase 1 have been documented in a series of reports which are publicly available and can be downloaded from the project website. Numerous publications are also available summarizing the results[11-23].

3.0 Phase 2 Results

Phase 2 of the project, which covers the period 2005 – 2008, was jointly funded by the U.S. DOE and an international consortium of energy companies, service companies and research organizations. While the detailed results from the consortium are proprietary, selected accomplishments from this phase included:

- An improved multi-component isotherm model to estimate sorption capacity for coalbed gases based solely on readily accessible coal characterization parameters[24].
- Identification of a more appropriate multi-component counter-diffusion model[25].
- Laboratory setup, procedural development and experimental calibration for new equation-of-state (EOS) development[25].
- Laboratory setup for zero horizontal strain core-flood experiments[26].
- Measurement of excess stress in coal when CO_2 is injected and identification of significant coal mechanical weakening when exposed to CO_2[26].
- Comparative study of geo-mechanical and permeability models for CBM operations[27,28].
- Reservoir analysis of the RECOPOL (Poland) and Yubari (Japan) CO_2-sequestration pilots[29,30].
- Assessment of "best" reservoir environments and development strategies for CO_2-ECBM/sequestration projects[31].
- Development of an internet-accessible knowledge base.
- Continued the facilitation of global technology exchange and networking via the project website and annual Coal-Seq forums.

Further description of several of these accomplishments is provided below.

3.1 New Multi-Component Sorption Model[24]

In Phase 1 of the project, the commonly-applied extended Langmuir model was shown to produce unacceptably inaccurate predictions for multi-component adsorption of gases.

Moreover, the Langmuir model parameters required for multi-component adsorption predictions are determined from single-component isotherm measurements. This limitation of the Langmuir model necessitates the measurement of single-component isotherm on each coal for which multi-component adsorption predictions are desired. Attempts to generalize the Langmuir model parameters in terms of coal properties were also largely unsuccessful. This was a consequence of the semi-empirical nature of the extended Langmuir model which is known to be thermodynamically inconsistent[32].

Therefore, in Phase 2, a rigorous, multi-component adsorption model based on the local form of the density-functional theory was developed by coupling the simplified local-density adsorption model with the Peng-Robinson equation-of-state (SLD-PR). The model was generalized using a proprietary database of more than 100 unique and independent isotherms on 10 coals comprising around 1,000 data points. The generalized multi-component gas adsorption model was found to be capable of predicting, *a priori*, the adsorption of three gases (methane, nitrogen and CO_2) and their binary and ternary mixtures to within three times the experimental uncertainties, based solely on information provided by the coal characterization (i.e., the ultimate and proximate analyses).

The validation of the model was performed on a San Juan basin coal sample on which multi-component isotherms had previously been measured. The results are shown in Figure 1, and clearly indicate that the SLD-PR generalized model provides more accurate predictions of multi-component adsorption than the extended Langmuir model. Furthermore, the SLD-PR model predictions are *based solely on information provided by the coal characterization.* In comparison, the extended Langmuir model predictions shown in Figure 1 *require single-component isotherm measurements*.

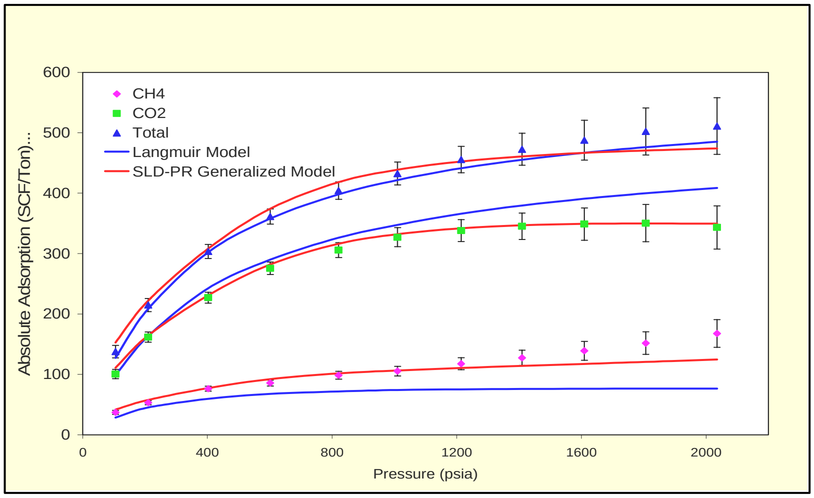
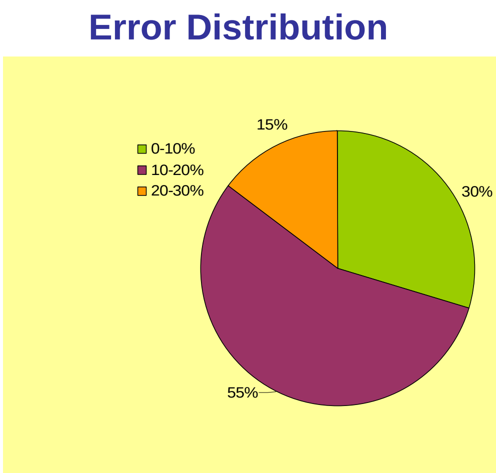


Figure 1: Comparison of Langmuir and SLD-PR Adsorption Model Predictions for Mixed-Gas Adsorption of Methane/Nitrogen on Wet Fruitland Coal at 115°F

Further validation of the SLD-PR generalized model was performed on a dataset of 27 coal samples originating from Australia, New Zealand, USA and Poland[33]. The authors of that work attempted to correlate adsorption properties to coal quality data using regression methods applied to the experimental data obtained. They concluded:

"*None of the coal properties measured in this study correlated sufficiently with sorption capacity to be used as a general indicator of CO_2 capacity for individual coals. The implication of this finding is that there is no single measurement that can be made as a reliable surrogate for sorption capacity; the sorption capacity of each coal must be measured individually.*"

The SLD-PR model was applied to this dataset and was found to be capable of predicting, *a priori*, all adsorption measurements with an average absolute percent deviation of 14% (Figure 2), using only the information provided by the coal characterization of these diverse coal samples. The SLD-PR generalized model, thus, can provide accurate *a priori* multi-component adsorption predictions in the *absence of experimental isotherm data*. This capability of the SLD-PR model essentially eliminates the need for extensive multi-component isotherm measurements for such purposes. As such, our hypothesis of developing a rigorous, theory-based, thermodynamically consistent adsorption model has resulted in improving significantly our capability to predict multi-component adsorption behavior coalbed gases (methane, nitrogen and CO_2).



Coal	%AAD	Coal	%AAD
31	7.6	48	15.6
33	6.7	49	13.4
34	16.9	50	6.5
35	24.4	51	7.5
36	24.7	52	10.9
37	16.4	53	7.5
38	17.4	54	10.2
39	28.6	55	12.5
40	12.7	56	9.8
41	27.0	57	3.1
42	17.6	58	14.0
43	14.1	59	13.8
46	18.1	60	4.1
47	15.5	**ALL**	**13.9**

Figure 2: SLD-PR Generalized Adsorption Model Predictions for Adsorption of CO_2 on Twenty-Seven Diverse Coals

Additional model development is envisioned in the future to include water as a separate adsorbed component. This will enable better estimates of in-situ gas storage capacity and release rates to be made, and will have a profound impact on CBM gas-in-place calculations. Further, there is evidence that coal strength is affected by moisture content (sub-equilibrium moisture leading to a weaker coal), and since CO_2 can dehydrate a coal, accounting for moisture in coal will provide important insights into coal weakening, failure and permeability changes with CO_2 injection.

3.2 Identification of Multi-Component, Bi-Directional Diffusion Model[25]

In Phase 1 of the project, it was postulated that Ficks Law of diffusion, which is the broadly accepted method of modeling the diffusion of gases in coal reservoirs, was inappropriate for multi-component conditions since 1) it is not suitable for multi-component gas systems and 2) it is not suitable for bi-directional diffusion. Therefore a Phase 2 objective was to identify, and inasmuch as possible validate, an alternative diffusion model that addressed these limitations.

Based on a thorough literature review on the topic of diffusion of gases, the Maxwell-Stefan (M-S) equation was identified as the most suitable model for multi-component, bi-directional diffusion of gases.

The first step towards adapting this for coal reservoirs was to develop it for diffusion of CO_2 and methane in opposite directions, methane diffusing out of the matrix and CO_2 moving into the matrix. The second step was to include a pore structure using an appropriate model, unipore or bi-disperse, and modify the equation. The last step, a challenging one, was to validate the model using laboratory diffusion data, either developed for the purpose, or through the use of available data. Unfortunately, all of this could not be completed during Phase 2. The first step was completed - the M-S equation, and all the input parameters required to use it, was identified. A means to determine these parameters was also developed. Australian researchers associated with the consortium (although not directly funded by it) extended this by incorporating a pore structure for coal into the formulation[34]. The application of the new model was then demonstrated. However, rigorous laboratory validation of the model has not yet been performed.

The development and demonstration of the M-S multi-component, bi-directional diffusion model is an important advancement, and one that we believe will greatly enhance our ability to model and predict the performance of multi-component gas processes in coals. However model validation against laboratory and field data is still required, which would be the objective of future work.

3.3 Laboratory Setup for EOS Development[25]

It was discovered in Phase 1 of the project that the common equations-of-state (EOS) utilized today to predict the properties of gas mixtures (CH_4-CO_2-N_2 mixtures specifically) fail to do so to an acceptable level, particularly at conditions approaching the CO_2 critical point. Further, there is very little experimental data for such systems upon which to develop a new EOS. Gas mixture properties are necessary for a variety of analytic tasks relevant to ECBM/sequestration, including data reduction of sorption experiments, prediction of diffusion rates in the M-S model, and flow modeling in reservoir simulators.

To address this issue in Phase 2, an experimental setup was designed and fabricated to measure the density of two-component (to begin with) gas mixtures at different compositions and pressures, and thus begin to establish a database of gas mixture properties upon which a new EOS can be developed. The setup, consisting of three high pressure vessels, each fitted with a high precision pressure transducer, is pictorially shown in Figure 3. The entire experimental setup is placed in a constant temperature water bath to ensure that the temperature does not change over the duration of an experiment.

Figure 3: Experimental Setup for Measurement of Density of Gas Mixtures

The experimental procedure includes calibration prior to starting an experiment to calculate the exact volumes of all components of the setup. Methane and CO_2 (for example) are then injected in two of the vessels, while the third vessel is held under vacuum. A part of each of the two gases is then bled into the third vessel. By recording the pressure before and after bleeding, the exact quantity of each gas leaving the vessel is determined. This is the mass of each component entering the evacuated vessel. Using the pre-determined volume of the third vessel, the exact density is calculated. Hence, density of the gas mixture, along with composition and final pressure becomes known. The entire procedure is repeated for a step-wise change in the gas composition, thus providing a complete composition/density/pressure relationship for a binary gas system.

While the system was fabricated and calibrated in Phase 2, cost overruns prevented the acquisition of meaningful amounts of actual data. Thus the underlying data upon which a new EOS could be developed was not collected. Future work by the consortium would aim to collect a comprehensive set of gas mixture property data, extend the scope to three-component systems (CH_4-CO_2-N_2), with moisture, and develop a new EOS.

3.4 Measurement of Excess/Reduced Stress and Mechanical Weakening with CO_2[26]

Typically, permeability experiments in coal are conducted in the laboratory for stress controlled conditions, where external stresses, horizontal and vertical, are monitored and varied. However, based on recent field observations and development of permeability models, it has been proposed that to replicate *in-situ* conditions, such experiments should be carried out under strain controlled conditions, where a sample cannot physically swell or shrink with depletion, or injection, due to lateral confinement under *in situ* conditions (Figure 4). Instead, the horizontal stresses are altered to ensure zero strain. It was therefore decided that in Phase 2 of the project core flood experiments would be conducted in this manner. Hence, this element of the experimental work was unique. It was the first time ever to conduct laboratory flow experiments where the sample was held under uniaxial strain conditions, that is, the vertical stress representing the overburden was maintained constant throughout the experiment, while holding the sample at constant diameter, not letting it physically swell or shrink. The resultant change in horizontal stress was termed “excess” stress with CO_2 injection since the sample was prevented from swelling by increasing the stress, and “reduced” stress with N_2 injection, where the stress was decreased to ensure that the sample did not shrink, since nitrogen is less sorptive than methane and there is an overall shrinkage of the coal sample.

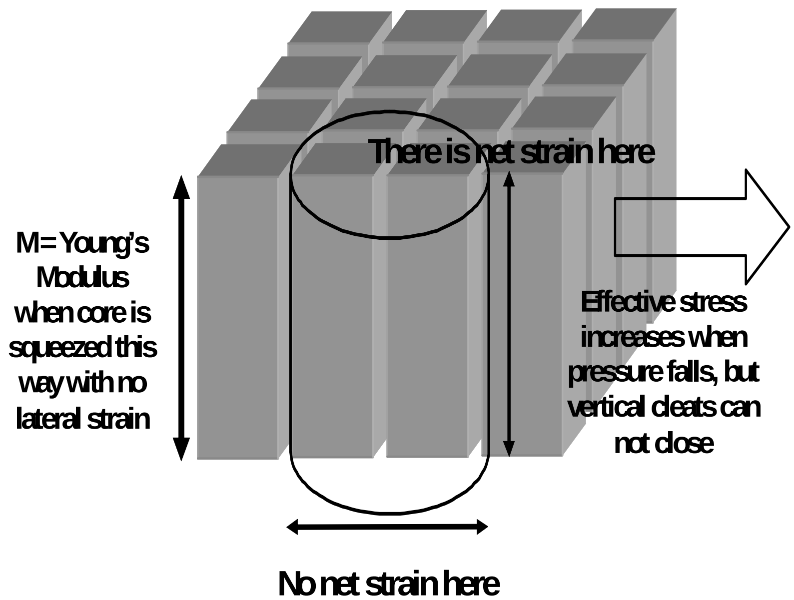


Figure 4: Concept Behind Zero-Strain Core Flood Experiments

It was found that excess stress required to ensure uniaxial strain conditions with CO_2 injection was so large that the sample could not endure it, resulting in failure. Similarly, the reduced stress with N_2 injection was also so significant that it required taking the stress off the sample completely, making it impossible to complete the experiments as originally envisioned. An example of experimental results is shown in Figure 5, showing the horizontal strain induced over one experiment. The first part of the plot shows the mechanical effect due to application of stresses. Without adjusting the stresses, nitrogen was injected and the resultant swelling due to sorption of nitrogen is shown as the second part of the plot. The third and fourth parts show the strains induced with continued swelling due to injection of methane, followed by CO_2. These results support the conclusion that excess and reduced stresses can be significant, making it difficult to carry out such experiments.

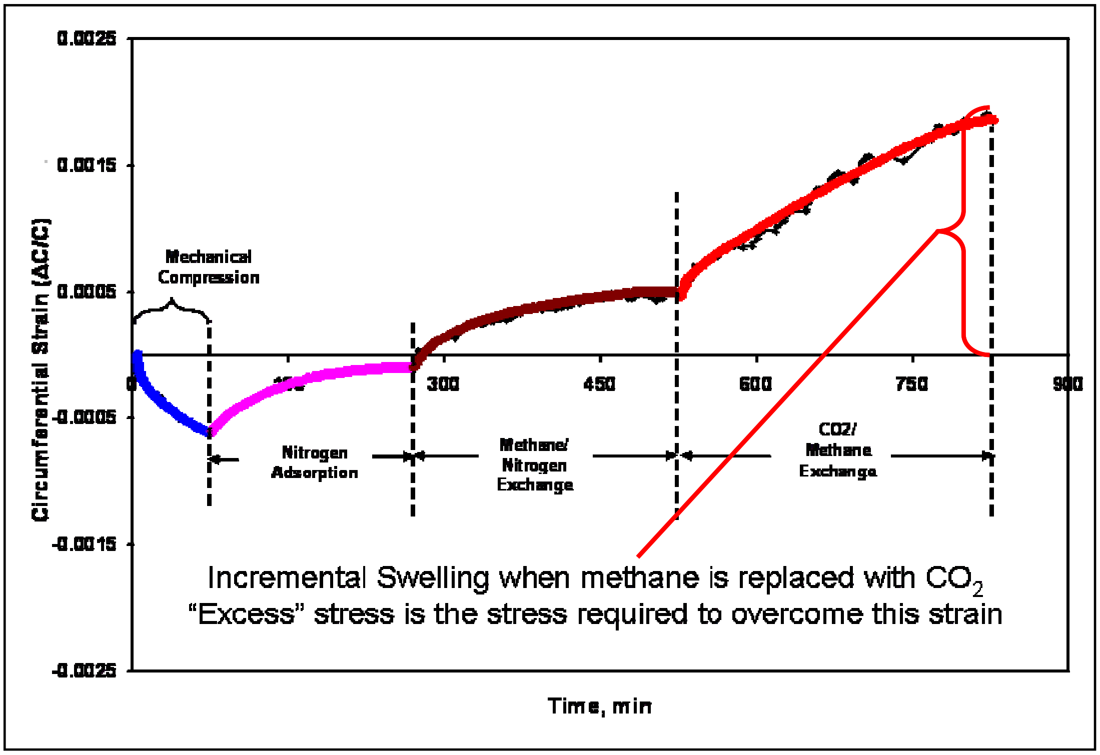


Figure 5: Horizontal Strain Induced by Nitrogen/Methane/CO_2 (stress-controlled)

The unexpected failure of coal with injection of CO_2 was analyzed using principles of geo-mechanics. The results of the analysis did not support the finding of coal failure with increased horizontal stress (Figure 6), since the stresses present at the time of failure were uncharacteristically below the expected failure conditions for a coal of this rank. A conclusion of the experimental finding was that the mechanical properties of coal change with injection of CO_2, resulting in either microfracturing, weakening and/or plasticization. There is (some) field evidence that injectivity of CO_2 improves with continued injection, which has never been explained. Further work is needed to understand the levels of excess/reduced stress created when CO_2 or N_2 is injected into coal, the impact of CO_2 on coal mechanical strength, and the mechanisms leading to the changes. This would be an important objective for future work by the consortium.

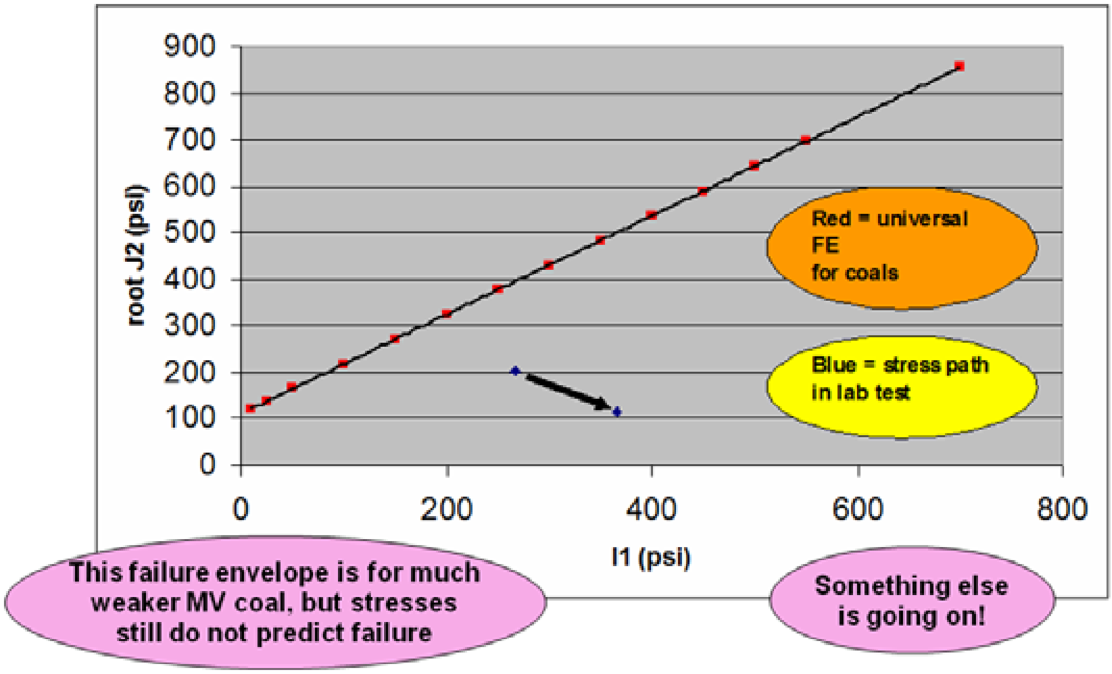
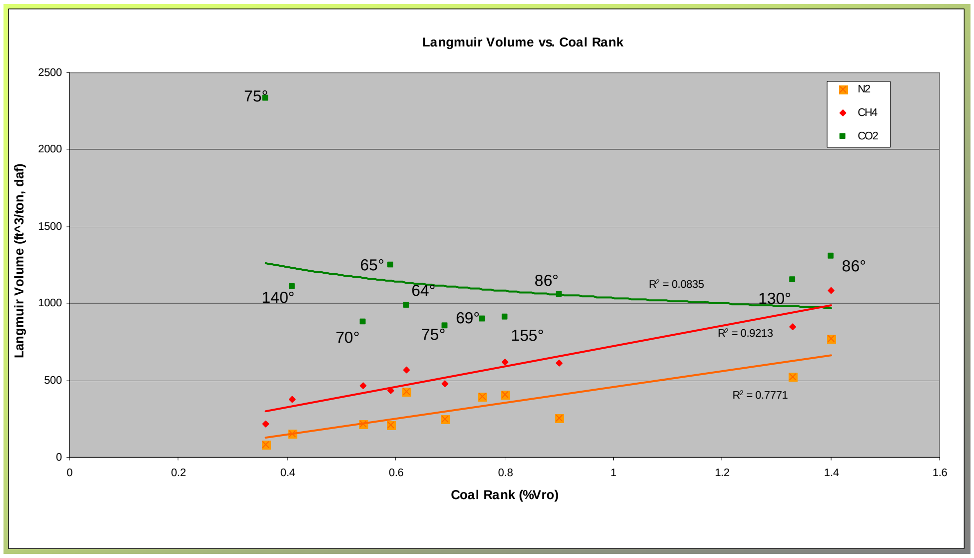


Figure 6: Coal Failure Conditions

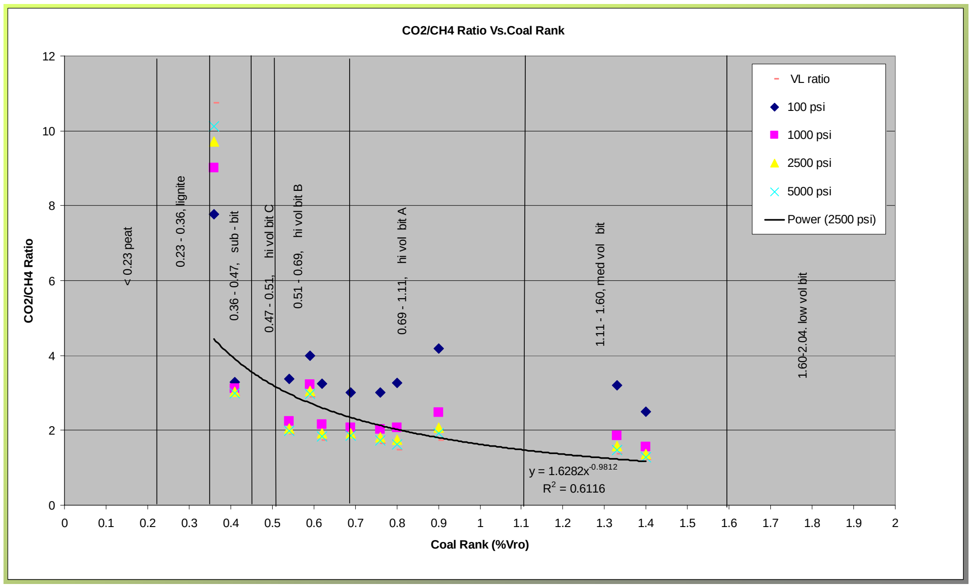
3.5 Assessment of "Best" Reservoir Environments and Development Strategies[31]

In Phase 1 of the project, coal samples from most major CBM basins in the U.S. were collected and CH_4, CO_2 and N_2 isotherms measured. Analysis of these data suggested that while sorption capacity to methane increases with coal rank (an already well established fact), sorption capacity for CO_2 seems much less dependent on coal rank (Figure 7 (a)). The implication is that lower rank coals will have a much greater CO_2 to CH_4 replacement ratio than higher rank coals (Figure 7 (b)). Given the effect of coal swelling and permeability reduction on CO_2 injectivity and project feasibility (notwithstanding the findings from the previous section), in Phase 2 a reservoir simulation study was performed to investigate what levels of initial permeability would be required to successfully implement a CO_2 injection project for different levels of coal rank, and whether advanced development strategies could extend the window of applicability to lower permeability coal reservoir environments. To perform the study, a matrix of simulation cases that consisted of three coal ranks (low, medium and high), three levels of permeability (1, 10 and

100 md), three types of injection wells (vertical damaged, vertical stimulated, and multi-branch horizontal), and two types of production wells (vertical and pinnate) were established. Some of the flood patterns are illustrated in Figure 8.



(a) Gas Storage Capacity as a Function of Coal Rank



(b) CO_2/CH_4 Replacement Ratio as a Function of Coal Rank

Figure 7: Gas Storage Capacity and CO_2/CH_4 Replacement Ratio as a Function of Coal Rank

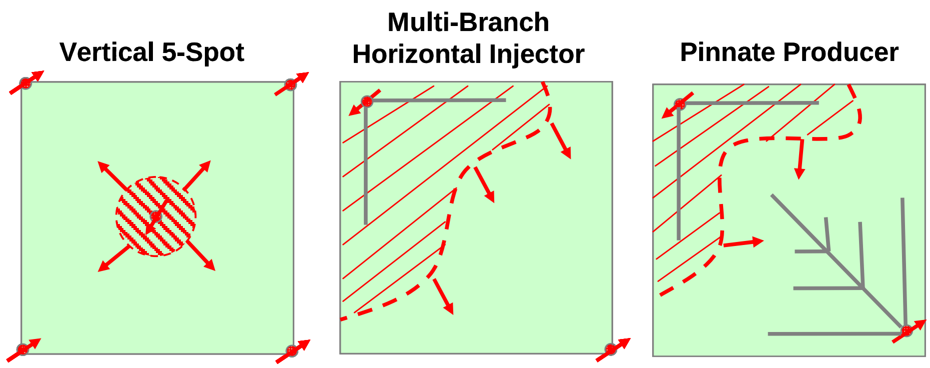


Figure 8: Illustration of Flood Patterns Studied

Results from the study indicated that due to the high CO_2/CH_4 replacement ratio for low rank coals, and the associated coal swelling and permeability reduction, even in a high coal permeability environment (i.e., 100 md), advanced pattern strategies consisting of multi-branch horizontal injectors and pinnate producers would be required to successfully sweep the pattern with CO_2 for the conditions assumed. For medium rank coals, successful sweep can be achieved with vertical wells in a high coal permeability environment, but advanced pattern strategies would be required if the permeability is in the 10 md range. In high rank coals, coal swelling with CO_2 injection is minimized, so successful sweep can be achieved in a 10 md coal permeability environment with vertical wells, but advanced pattern strategies would be required in a 1 md coal permeability environment.

This work suggests that the permeability required for a successful project is lower for high rank coals due to less coal swelling and permeability reduction, and that advanced pattern strategies can indeed lower the permeability required for successful sweep, without consideration to development costs. These findings can be used as an initial screening criteria for identifying reservoir environments and development strategies amenable to CO_2-ECBM/sequestration projects. To be of practical benefit to that end, this analysis should be extended to the actual geological and reservoir conditions that exist in the world's most significant coal basins with CBM activity.

4.0 Phase 3 Objectives

Further continuation of the consortium is currently being considered. Building upon the findings from Phase 2, some of the topics that have been identified for investigation in Phase 3 include:

- Further development of a robust sorption model to account rigorously for water as a separate adsorbed component.
- Laboratory validation of the multi-component, bi-directional diffusion model.
- Completion of the EOS work undertaken in the Phase 2 of the project, and extension to full CH_4-CO_2-N_2 ternary gaseous systems with moisture.
- Laboratory experiments to understand the conditions and nature of coal weakening and/or mechanical failure with CO_2 injection.
- Laboratory experiments to understand how coal compressibility factors, as utilized in the various permeability models, vary with pressure and/or gas concentration.
- Technical and economic feasibility assessments of CO_2/N_2 – ECBM/sequestration in major worldwide coal basins (e.g., San Juan, Powder River, Western Canadian, Surat, Ordos, Kuznets, etc.).
- Begin to examine the potential of organic shales to sequester CO_2 by collecting core and measuring CH_4, CO_2 and N_2 isotherms in most gas shale basins across the U.S.
- Provide a regular tele-forum for members and various project performers to exchange findings and ideas; also create a web-based discussion board.
- Continue the facilitation of global technology exchange and networking via the project website and annual Coal-Seq forums.

5.0 Final Remarks

The Coal-Seq consortium has been and continues to be the leading, internationally recognized project for advancing industry's understanding of complex coalbed methane and gas shale reservoir behavior in the presence of multi-component gases. This is accomplished via laboratory experiments, theoretical model development and field validation studies such that primary recovery, enhanced recovery and CO_2 sequestration operations can be commercially enhanced and/or economically deployed. The accomplishments from the first two project phases have yielded considerable advancements and insights towards these objectives, but as with any emerging technology, and particularly in a complex reservoir environment, many technical unknowns and challenges remain. Future work planned by the consortium would seek to continue to address these unknowns and challenges, advance the overall project objectives, and ultimately lead to commercial application of the technology.

6.0 Acknowledgements

The Coal-Seq consortium is sponsored by the U.S. Department of Energy, BP America, the CO_2-Cooperative Research Centre, ConocoPhillips, the Illinois Clean Coal Institute, Japan Coal Energy Center, Repsol YPF, Schlumberger, and Shell International Exploration & Production. The valuable financial and technical contributions of each sponsor is hereby acknowledged and appreciated. Contractors performing R&D for the project have included Advanced Resources International, Electrochemical Systems, Oklahoma State University, Southern Illinois University, and Higgs-Palmer Technologies. Their technical contributions are also acknowledged and appreciated.

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