

Hydrology [H]

H11J

MC:2002 Monday 0800h

Multiscale Science of Geologic CO₂ Sequestration I: Geochemistry

Presiding: A Navarre-Sitchler, University of Wyoming; J Kaszuba, University of Wyoming

H11J-01

Concept And Laboratory Verification Of In-situ Reaction Barrier For CO₂ Geological Storage

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In the Carbon dioxide Capture and Storage (CCS), the CO₂ is captured from emission source and stored into geological reservoirs at a depth below 800 m. The injected CO₂ is less dense than water, and as a result, it tends to migrate upward. For trapping to inhibit the upward migration of CO₂, the reservoirs should be covered with a sufficiently impermeable seal, i.e. caprock. However, the caprock may contain imperfections such as faults and fractures which will play a role of a high permeability path to arise leakage of the injected CO₂ from the reservoirs. We present here a concept to reduce the medium permeability along potential such leakage paths. An aqueous solution will be injected into the fractures and rocks through injection wells. The solution will have a low viscosity and will not impact formation permeability as long as the solution is left as it is, but when the solution encounters dissolved CO₂, precipitation will occur due to chemical reaction. As a result, the permeability will be reduced by filling the pores and fractures in the rocks with the precipitates. We have demonstrated this concept in the laboratory experiments by using the solution of Ca(OH)₂ in water. In this case, the chemical reaction between the Ca(OH)₂ solution and CO₂ should produce precipitation of calcium carbonate, CaCO₃. We prepared a cylindrical pressure vessel filled with glass beads and water. The temperature and pressure inside the vessel were kept at 35 degC and 10 MPa respectively simulating subsurface condition at 1000 m deep. The Ca(OH)₂ solution and CO₂ were injected into the vessel separately, and the permeability of the analogous rock of glass beads was measured before and after the treatment respectively. As was expected, the permeability decreased to about 10 percent within one day after the treatment and it was stabilized at least for a week until the end of the experiment. This concept may be also applicable to prevent the leakage through abandoned wells, to mitigate well cement degradation, and to enhance CO₂ mineral trapping.

H11J-02

Numerical Modeling of In-situ Reaction Barrier by Injection of Ca(OH)₂ Solution for CO₂ Geological Storage

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Containment of CO₂ in the storage reservoir is a very important issue. We present here an in-situ reaction barrier method to reduce the medium permeability along potential leakage paths of a deep CO₂ storage reservoir in a saline formation. An aqueous solution will be injected into the fractures and rocks through injection wells. The solution will have a low viscosity and will not impact formation permeability as long as the solution is left as it is, but when the solution encounters dissolved CO₂, precipitation will occur due to chemical reaction. As a result, the permeability will be reduced by filling the pores and fractures in the rocks with the precipitates. This concept has been successfully demonstrated previously through a laboratory experiment using Ca(OH)₂ solution. The reduction of permeability in the laboratory experiment has been reproduced by reactive transport modeling using TOUGHREACT. The concept of the in-situ reaction barrier has been applied to a 2-D caprock-aquifer system under field physical and chemical conditions using the modeling tool. Calcite precipitation, permeability reduction, and CO₂ leakage mitigation was achieved in the numerical experiment. This concept may be also applicable to prevent the leakage through abandoned wells, to mitigate well cement degradation, and to enhance CO₂ mineral trapping.

H11J-03

Magnesium Oxide Carbonation Rate Law in Saturated Brines

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Magnesium oxide (MgO) is the only engineered barrier certified by the EPA for emplacement in the Waste Isolation Pilot Plant (WIPP), a U.S. Department of Energy repository for transuranic waste in southeast New Mexico. MgO reduces actinide solubility by sequestering CO₂ generated by the biodegradation of cellulosic, plastic, and rubber materials. Demonstration of the effectiveness of MgO is essential for WIPP recertification. In order to be an effective barrier, the rate of CO₂ sequestration should be fast compared to the rate CO₂ production, over the entire 10,000 year regulatory period. While much research has been conducted on the kinetics of magnesium oxide carbonation in waters with salinity up to that of sea water, we are not aware of any work on

determining the carbonation rate law in saturated brines at low partial pressures of CO₂ (P_{CO_2} as low as $10^{-5.5}$ atm), which is important for performing safety assessments of bedded salt waste repositories. Using a Varian ion-trap gas- chromatograph/mass-spectrometer (GC/MS) we experimentally followed the CO₂ sequestration kinetics of magnesium oxide in salt-saturated brines down to a P_{CO_2} as low as $10^{-5.5}$ atm. This was performed in a closed reactor with a known initial P_{CO_2} . The results of this study show that carbonation is approximately first order in P_{CO_2} , in saturated brines. We believe that this method will benefit the study of the detailed kinetics of other similar processes.

H11J-04

Laboratory Studies on the Potential of Using Basalt for Large-Scale Mineral Sequestration of CO₂

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Geologic sequestration of CO₂ by means of injecting large quantities of supercritical carbon dioxide into deep geologic formations entails certain hazards, of which reservoir stability and risk of leakage are prominent examples. This study presents results from an international research project, where, alternatively, the CO₂ will be sequestered as carbonate minerals in basalt. CO₂ storage in basaltic rocks may provide a thermodynamically stable and therefore long-lasting solution to the problems of geologic sequestration. In nature, the carbonatization of basaltic rocks occurs in different settings, e.g. in deep ocean vent systems and through weathering of basaltic provinces. Yet, the rate of this carbonatization reaction is not well defined and hinges on crucial factors, such as the availability of the major divalent reactants (Ca, Mg, Fe) and the reactive surface area. Both parameters may change with the degree of carbonate coating. In order to investigate this interplay between desired carbonate precipitates and their potentially detrimental effect on the dissolution of the host rock by limiting the flux of Ca, Mg, and Fe into the fluid phase laboratory studies were carried out with NaHCO₃ saturated solutions in the presence of basaltic glass. Results from these dissolution experiments in mixed-flow reactors will be presented. Furthermore, release rates of the major carbonate cations are dependent on the degree of crystallinity of the host matrix and change from basaltic glass to crystalline rock. While such release rates are readily available for basaltic glasses (Wolff-Boenisch et al., 2006) they are missing for the crystalline counterpart. We aim at closing this gap and discuss implications for the injection of CO₂-charged waters into basaltic and largely crystalline terrain such as continental flood basalts. References: Wolff-Boenisch et al. (2006), *Geochim. Cosmochim. Acta* 70, 858.

<http://www.carbfix.com>

H11J-05

Mineral CO₂ Sequestration into Basalt: The Carbfix Project

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The reduction of industrial CO₂ emissions is considered one of the main challenges of this century. Among commonly proposed CO₂ storage techniques, the injection of anthropogenic CO₂ into deep geological formations is quite promising due their large potential storage capacity and geographic ubiquity. Finding a storage solution that is long lasting, thermodynamically stable and environmentally benign would be ideal. Storage of CO₂, as solid calcium magnesium iron carbonate, in basaltic rocks may provide such a long lasting, thermodynamically stable and environmentally benign solution. In nature, the carbonization of basaltic rocks occurs in a variety of well-documented settings, such as the hydrothermal alteration in geothermal systems and in deep ocean vent systems. The goal of this research project is to optimize industrial methods for storing CO₂ in basaltic rocks through a combined program consisting of, field scale injection of CO₂ charged waters into basaltic rocks, laboratory based experiments, study of natural CO₂ waters as natural analogue and state of the art geochemical modelling. A second and equally important goal of this research project is to generate the human capital and expertise to apply the advances made in this project in the future. Towards this goal the bulk of the research is to be performed by graduate student and post-doctoral trainees. At the Hellisheidi Iceland site, the hot gases released from geothermal energy production will be processed to separate the CO₂. It will then be dissolved in water at about 25 bar pressure and pumped into the porous basalt at 400 to 700 m depth, at the rate of 30 000 tonnes per year. Model simulations, natural analogues and experimental work suggest that the CO₂ charged waters will react with the basalt and form carbonate minerals such as FeCO₃ - MgCO₃ solid solutions and CaCO₃. By this method the fixed CO₂ will remain trapped as mineral for millions of years.

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H11J-06

Potential for in situ carbonation of peridotite for geological CO₂ storage

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The rate of natural carbonation of tectonically exposed mantle peridotite during weathering and low temperature alteration can probably be enhanced to develop a significant sink for atmospheric CO₂. Formation of solid carbonate minerals in situ constitutes an important alternative that should be explored. It may be less costly than ex situ mineral carbonation involving transport of solid reactants, grinding, heat treatment, and reaction in pressurized vessels. It is certainly safer and much easier to monitor than storage of super-critical CO₂ fluid in pore space at depth. Natural carbonation of peridotite in the Samail ophiolite, an uplifted slice of oceanic crust and upper mantle in the Sultanate of Oman, is surprisingly rapid. Carbonate veins in mantle peridotite in Oman have an average 14C age of approx 26,000 years, and are not 30 to 95 million years old as previously believed. These data and reconnaissance mapping show that 10,000 to 100,000 tons per year of atmospheric CO₂ are converted to solid carbonate minerals via peridotite weathering in Oman [1]. Peridotite carbonation can be accelerated via drilling, hydraulic fracture, input of purified CO₂ at elevated pressure, and particularly increased temperature at depth. Our simple 1D thermal models suggest that, after an initial heating step, CO₂ injected at 25 or 30°C can be heated by exothermic carbonation reactions that sustain high temperature and rapid reaction rates at depth with little expenditure of energy. In situ carbonation of peridotite could consume more than 1 billion tons of CO₂ per year in Oman alone, affording a low-cost, safe and permanent method to store atmospheric CO₂ [1]. In the appropriate PTX regime, solid volume changes associated with peridotite carbonation may induce reaction driven cracking as well as exothermic heating. If cracks expose fresh, new surface area to sustain continued reaction, carbonation rates could accelerate over time. Alternatively, if cracking is too slow, then armoring of unreacted peridotite minerals with carbonate reaction products will lead to decreasing rates. In Oman, there is a marked difference between (a) mildly carbonated peridotite, in which partially serpentinized, olivine-rich rocks host carbonate veins, and reaction products lack talc and quartz, and (b) rarer, completely carbonated peridotite composed of carbonate + talc and/or quartz with no remaining olivine. This suggests that the natural system has accessed both self-limiting and self-catalyzing conditions over time [2]. It is vital to use observations and models to predict conditions that cause reaction driven cracking and exothermic heating, and test predictions in field studies. We are just beginning this work. [1] Kelemen & Matter, Proc Nat Acad Sci 2008, in press [2] Kelemen, Matter & Streit, Proc Conf Accelerated Carbonation for Environmental & Materials Eng., 2008, in press

H11J-07

Modeling Study on Injection of Supercritical CO₂ Into a Deep Saline Carbonate Formation

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A modeling study on injection of supercritical CO₂ into a deep saline carbonate formation was performed using TOUGHREACT Pitzer ion-interaction model. The carbonate formation consists of calcite (72.5%), dolomite (21.5%) and anhydrite (<6%). The brine of the formation is known as NaCl-dominant with salinity at about 250,000 ppm (NaCl equivalent), temperature at 102° C and pressure at 225 bars. The detailed chemical composition of the brine was unknown. It was reconstructed according to the salinity and the known detailed composition of a brine from a similar formation with slightly lower salinity (about 190,000 ppm). The reconstructed formation brine has an ionic strength ~5 molal and pH 5.4 with considerable concentrations of Ca⁺², Mg⁺², HCO₃⁻ and SO₄⁻². CO₂ injection was considered at a constant rate and for a period of 1 year, through a vertical well in a 2D radial model domain, and a horizontal well in a 3D model domain, respectively. The preliminary simulations found that: (1) at the end of the injection, a dryout zone is developed within a few meters from the injection well due to displacement by the injected supercritical CO₂ and the evaporation of water from brine into CO₂; (2) at the front of the dryout zone, brine is further concentrated (ionic strength up 20 molal) due to water evaporation, pH is lowered to 3.1, halite (NaCl) and anhydrite (CaSO₄) precipitate, and the brine is converted into CaCl₂-dominant; (3) precipitation of halite in the dryout zone reduces the formation porosity by about 5%-10%; (4) HCl gas is generated from the dryout front; (5) calcite dissolves close to the injection well and precipitates at areas far from the well, however, the overall mineral trapping is not significant in hundreds of years for this carbonate formation. These findings are valuable for the assessment of the potentials of this carbonate formation for CO₂ sequestration, injectivity changes, and well degradation by potential corrosion.

H11J-08

Reaction of CO₂ and brine at the interface between Portland cement and casing steel: Application to CO₂ sequestration

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Prediction of CO₂ leakage through wellbore systems is a multiscale problem in geologic sequestration. In order for wellbore leakage to occur, km-scale processes must deliver CO₂ from the point of injection to the wellbore. But, in order for the wellbore to actually leak, µm-scale processes must operate to

allow CO₂ to flow up the wellbore. In this study, we describe experiments and modeling of microscale processes accompanying CO₂ leakage along the cement-casing interface. This work fits within a broader predictive study of CO₂ sequestration performance (Viswanathan et al. 2008, *Env Sci and Tech*, in press) that includes calculation of CO₂-migration times to wellbores. Experiments carried out in this report consisted of synthetic wellbore systems constructed of Portland cement and casing-grade steel in which a mixture of CO₂ and brine were forced along the cement-casing interface at in situ sequestration conditions (40 °C and 14 MPa). The CO₂-brine mixture was pre-equilibrated by flow through limestone before encountering the cement-casing composite. (The limestone-equilibrated fluid was calculated to be strongly out of equilibrium with both cement and the casing.) We used a high CO₂-brine flux (10-20 ml/hour along the interface) and hypothesized that the interface would widen with time due to dissolution of either or both cement and steel. In addition to experiments, we conducted reactive transport modeling of cement reactivity using FLOTRAN, which was modified to allow representation of solid solution in the dominant cement phase, calcium-silicate-hydrate. We also developed a corrosion model for the steel. The experimental results showed that the steel was more reactive than the Portland cement. Extensive deposits or oxidation products of FeCO₃-rich material developed at the interface and in some places led to an apparent closure of the interface despite the large flux through the system. In contrast, alteration of the cement appeared to be limited by diffusion of CO₂ into the cement matrix and carbonation of the cement to CaCO₃. The cement interface did not appear to have been significantly eroded. The experiment was used to calibrate numerical models for corrosion rates and for cement carbonation. These results were applied to interpret samples recovered from a CO₂-enhanced oil recovery field (SACROC in West Texas; Carey et al. 2007, *Int J. Greenhouse Gas Control*, 1: 75-85). The results suggest that CO₂-brine flux must have been limited along the cement-casing interface because the casing showed very little corrosion. They also suggest that CO₂ penetration along the cement-formation interface was limited in volume because the depth of carbonation at SACROC was limited. These microscale models suggest that cement-casing flow has the potential to be self-limiting due to precipitation of CO₂ and that standard logging measurements of casing integrity can be used to assess whether significant flow of CO₂-brine has occurred at the casing interface.

H11J-09 INVITED

On the Role of Multi-Scale Processes in CO₂ Storage Security and Integrity

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Consideration of multiple scales in subsurface processes is usually referred to the spatial domain, where we may attempt to relate process descriptions and parameters from pore and bench (Darcy) scale to much larger field and regional scales. However, multiple scales occur also in the time domain, and processes extending over a broad range of time scales may be very relevant to CO₂ storage and containment. In some cases, such as in the convective instability induced by CO₂ dissolution in saline waters, space and time scales are coupled in the sense that perturbations induced by CO₂ injection will grow concurrently over many orders of magnitude in both space and time. In other cases, CO₂ injection may induce processes that occur on short time scales, yet may affect large regions. Possible examples include seismicity that may be triggered by CO₂ injection, or hypothetical release events such as "pneumatic eruptions" that may discharge substantial amounts of CO₂ over a short time period. This paper will present recent advances in our experimental and modeling studies of multi-scale processes. Specific examples that will be discussed include (1) the process of CO₂ dissolution-diffusion-convection (DDC), that can greatly accelerate the rate at which free-phase CO₂ is stored as aqueous solute; (2) self-enhancing and self-limiting processes during CO₂ leakage through faults, fractures, or improperly abandoned wells; and (3) porosity and permeability reduction from salt precipitation near CO₂ injection wells, and mitigation of corresponding injectivity loss. This work was supported by the Office of Basic Energy Sciences and by the Zero Emission Research and Technology project (ZERT) under Contract No. DE-AC02-05CH11231 with the U.S. Department of Energy.

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Hydrology [H]

H12C

MC:2002 Monday 1020h

Multiscale Science of Geologic CO₂ Sequestration II: Monitoring and Leakage

Presiding: A Navarre-Sitchler, University of Wyoming; R Pawar, Los Alamos National Laboratory

H12C-01

Basin-Scale Hydrologic Impacts of CO₂ Sequestration within the Mt Simon Formation, Illinois Basin; Scaling Calculations using Sharp-Interface Theory

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The Illinois Basin hosts dozens of coal fired power plants generating more than 80 million metric tons of CO₂ annually. Here we present a suite of basin-scale, hydrologic models of the Mt Simon formation, Illinois Basin using sharp interface theory. The goal of these models is to determine what are the basin-scale hydrologic consequences of CO₂ injection and whether some regions of the Illinois Basin would represent a better venue for carbon sequestration than others. While this approach makes some restrictive simplifying assumptions, it allows us to assess the problem at the sedimentary basin scale. Our solution domain spans the northern two thirds of the Illinois Basin (about 230,000 km²). We allowed porosity and permeability to decrease with depth from 0.2 to 0.05 and 400 to 2 mD, respectively. We injected CO₂ using 727, 10 inch diameter injection wells delivering about 210 kg/minute/well. The wells were positioned about 2 km apart in a radial pattern around known power plant locations. We ran the injection wells for 100 years. The wells were then shut in for an additional 900 years. Results indicate that after 100 years of continuous injection, deviatoric fluid pressures varied between 9.2 to 0.5 MPa between the deepest and shallowest injection wells. For the deepest portion of the basin (~ 3.1 km), deviatoric pressures reach about 22 percent of lithostatic levels. Owing to the rather subtle regional hydraulic gradient (200m/500km), long-range separate-phase CO₂-migration is driven by buoyancy at a rate of only 2 m/year. If CO₂ remained as a separate phase on time scales of 100,000 years, the injected CO₂ would migrate about 200 km to the north before charging gentle structural traps. Owing to the radial, bowl-shaped geometry of the Illinois Basin, net brine displacement to the north would be small, probably less than 100 m. Our analysis suggest that the Mt. Simon formation represents a good venue for CO₂ sequestration although shallower regions (~ 2 km depth) would pose less risk of catastrophic breaching due to high deviatoric fluid pressures. Fluid pressures do not return to hydrostatic conditions after 1000 years due to buoyant forces resulting from the presence of a separate CO₂ phase.

H12C-02

A Hypothetical Scenario for Full-Scale Deployment of Geological Carbon Sequestration: Investigating the Interaction Between Multiple CO₂ Storage Sites in a Sedimentary Basin

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Most ongoing projects of geological carbon sequestration (GCS) are relatively small in size, with annual injection rates from a few thousand to less than a million tonnes. These projects help build the GCS technology with respect to modeling, monitoring, risk assessment, and mitigation, and have been successful so far in terms of CO₂ containment and caprock geomechanical integrity. In the future, GCS will be implemented at full-scale, multiple industrial-size CO₂ storage sites in large sedimentary basins to make full use of the potential storage capacity. Simultaneous injection into multiple not-too-distant storage sites will lead to interference between the individual regions of pressure build-up and possible interference between the individual CO₂ plumes. The Illinois Basin is used to model the future impact of multiple injection sites in the thick, extensive Mount Simon Formation. The basin-scale model domain of 241,000 km² covers a core injection area of 24,000 km², a larger near-field area where significant pressure buildup is expected, and an even larger far-field area for investigating environmental impacts on groundwater resources. The model assumes that there are twenty sequestration sites (spaced 30 km apart) within the core injection area. Three injection scenarios are considered, featuring annual injection rates of 5, 10, and 15 million tonnes of CO₂ at each site, respectively. These scenarios correspond to 33%, 67% and 100% of the current single-point large CO₂ sources in the relevant states (Illinois, Indiana and Kentucky). The model adequately captures the characteristics of the Mount Simon Formation in the core injection area, which include (1) an overall thickness of 300 to 680 m, (2) an upper unit of sandstone and shale tidally influenced and deposited, (3) a thick middle unit of clean sandstone of relatively high permeability, and (4) a lower arkosic unit of higher permeability (one Darcy) with an average thickness of 90 m. At each site, CO₂ is injected into the lower arkosic unit to ensure sufficient permeability to accommodate high injection rates. A three-dimensional unstructured mesh is used for the model, with

progressive refinement horizontally from the far-field area to the core injection area and radial refinement toward each injection center within the injection area, as well as progressive vertical refinement from four model layers in the far-field area to 50 model layers for each CO₂ plume. The overlying Eau Claire seal and the underlying Pre-Cambrian granite unit are also included in the model. Both the two-phase CO₂-brine flow within the twenty CO₂ plumes and the single-phase brine flow away from the plumes are simulated using the parallel TOUGH2/ECO2N simulator. Preliminary simulation results are discussed with respect to (1) the dynamic evolution and migration of individual CO₂ plumes, (2) the possible interference between different plumes, (3) the interference between individual regions of pressure buildup, (4) the possible impacts of GCS on the groundwater resources at the basin's boundary, and (5) the possibility of caprock damage.

H12C-03

Analytical solution for pressure buildup and plume evolution during injection of CO₂ into saline aquifers

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If geo-sequestration of CO₂ is to be employed as a key greenhouse gas reduction method in the global effort to mitigate climate change, simple yet robust methods must be available to help design and monitor injection into saline aquifers. There has been significant development of simple analytical and semi-analytical techniques to support screening analysis and performance assessment for potential carbon sequestration sites. These techniques have generally been used to estimate the size of CO₂ plumes for the purpose of leakage rate estimation. A common assumption of previous work has been that both the fluids and the geological formation are incompressible. Consequently, calculation of pressure distribution requires the specification of an arbitrary radius of influence. In the present work, we relax this restriction by incorporating fluid and formation compressibility into our governing equations. These equations are transformed into ordinary differential equations using a similarity transformation, and are then solved using the method of matched asymptotic expansions. By allowing for compressibility in the fluids and formation, the solutions improve on previous work by not requiring the specification of an arbitrary radius of influence. Our solution is also capable of accounting for non-Darcy inertial effects modeled by the Forchheimer equation. These analytical solutions are validated by comparison with finite difference solutions. Our analysis leads to a simple yet highly accurate algebraic equation for estimating the evolution of a CO₂ plume, and the associated pressure buildup, as a function of time.

H12C-04

Analysis of Heterogeneity Length-scales on Residual Gas trapping and Buoyancy-driven CO₂ Migration

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The primary purpose of this study is to elucidate how variations in the length-scale of permeability (k) heterogeneity affect CO₂ sequestration processes, especially buoyancy-driven flow and residual gas trapping. We systematically implemented 24 scenarios corresponding to 141 simulations representing a systematic variation in k: homogeneous and isotropic (four scenarios), homogeneous and anisotropic (five scenarios), random (five scenarios), homogeneous with a low k inclusion (two scenarios), correlated k field with k isotropy (four scenarios), and k anisotropy (four scenarios). Sequential Gaussian simulation was used to generate an equally probable 10 realizations of each scenario except for the homogeneous cases. Effective permeabilities were calculated to evaluate the ambient velocity field in each realization. In addition, the first and second moments of CO₂ plume distribution were examined to explore both the lateral and vertical CO₂ displacement behavior according to geologic complexity. With quantified velocity information and plume distribution, we investigated the systematic variation of CO₂ trapping mechanisms corresponding to different heterogeneity length-scales. Simulation results reveal that residual CO₂ trapping increases with effective vertical k in both homogeneous and uncorrelated random fields. Increasing the effective vertical k increases CO₂ velocities, resulting in farther CO₂ migration. Consequently, the injected CO₂ plume sweeps a larger area thus the amount of residual-trapped CO₂ increases. Interestingly, horizontal connectivity also increases residual trapping by increasing the lateral CO₂ plume extension. For example, a longer correlation length enhances the lateral extension of a CO₂ plume, also increasing the amount of residual gas trapping. In sum, residual trapping is enhanced when CO₂ plume migrates farther vertically (vertical k control) or laterally (horizontal k control or correlation lengths control). In practice, the goal is to store more CO₂ as residually-trapped gas, and to minimize buoyancy-driven CO₂ migration. These simulations suggest that heterogeneous k fields with longer correlation lengths are more promising target formation for geologic CO₂ sequestration. Finally, these findings imply that horizontal injection wells may enhance residual CO₂ trapping significantly, because such horizontal wells will likely increase the early lateral extent of an injected CO₂ plume.

H12C-05

Relative Permeability Properties of CO₂ and Brine in Reservoir Rocks.

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This paper presents the results of laboratory experiments of relative permeability to brine and CO₂ on core samples of reservoir rocks. A new experimental facility in the Department of Energy Resources Engineering at Stanford University has been developed to replicate reservoir conditions ($T > 40^{\circ}\text{C}$, $P > 80$ bars) during CO₂ injection. We are able to co-inject brine and CO₂ continuously at flow rates anticipated in the storage reservoir near and distant from the injection well. Saturation distributions of each phase within the core are determined using X-ray CT scanning. Using this apparatus, experiments in which CO₂ and brine are co-injected at different proportions have been carried out over a range of flow rates. The results are analyzed in conjunction with sub-core scale capillary pressure measurements, analysis of the corresponding thin sections and numerical simulations.

H12C-06

Simulating Coupled Fluid Mechanical Processes During CO₂ Injection

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The geologic sequestration of CO₂ is one of the technologies being considered for mitigating impact of anthropogenic emissions of CO₂. Because of vast quantities of fluid needed to be injected in the subsurface and the relative lack of pore space, geo-mechanical effects and their impacts will be an important consideration in the evaluation of sequestration sites. Numerical models to predict the geo-mechanical effects must be capable of accurately representing the coupled thermal-hydrologic-mechanical (THM) processes and solving the resulting nonlinear system of equations in a very efficient manner. We present application of our approach to characterize the geo-mechanical impact of large-scale CO₂ injection. The approach is based on the coupled solution of THM processes presented by Bower and Zyvoloski (1997). Accurate representation of permeability-displacement behavior in faults presents significant challenges. We have developed a general permeability-displacement approach that is applicable not only to general rock types but also to accurately represent faults in large grid blocks. The approach allows use of a complex model near injection wells and a relatively simple model in the far field to accurately capture the geo-mechanical effects. Our numerical studies are focused on understanding how faults/fault gouges behave during large-scale CO₂ injection including coupled fault displacements and CO₂ migration. The results will show why a coupled approach is needed for accurate representation of geo-mechanical effects during large-scale CO₂ injection. Reference: Bower, K. M., and G. Zyvoloski, 1997, "A Numerical Model for Thermo-Hydro-mechanical Coupling in Fractured Rock," Int. J. Rock Mech. Min. Sci. Vol. 34, No. 8, pp. 1201-1211.

H12C-07

Pore-Scale Analysis of Microtomography Images of the Rock in Geosequestration Research

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Accumulation of carbon dioxide and other green-house gases in the atmosphere impacts the Earth energy balance and leads to global warming. Injection of gas in deep geologic formations is one of possible ways to mitigate the unwanted climate changes. Success of such program is determined by the capability of trapping the injected gas under the surface for a geologic time. This process is very complex: it includes interaction of multiphase flow and geochemistry processes across multiple scales. In particular, it is very important to clearly understand the consequences of injection at the pore scale and how these processes upscale to entire aquifer. Synchrotron-based microtomography at the Advanced Light Source line 8.3.2 at Lawrence Berkeley National Laboratory produces high-resolution three-dimensional images of the pore space of the rock of interest. A suite of methods of analysis of petrophysical properties of the rock provide a glimpse into the multiphase flow properties of the rock and the impact of injection and geochemical processes their modification. We present some results obtained by the method of Maximal Inscribed Spheres developed at Lawrence Berkeley National Laboratory. This work follows up presentation Eos Trans. AGU, 86(52), 2005 Fall Meet. Suppl., Abstract H33A-1381

H12C-08

Non-modal analysis of the onset of density-driven convection in porous media: Effect of medium heterogeneity

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In the context of geological sequestration of carbon dioxide (CO₂), trapping of the injected CO₂ due to dissolution is expected to play a dominant role in the short term. Due to its importance, a lot of work has recently been done to understand the length and time scales associated with the convective motions leading to the dissolution of injected CO₂. Recently, Rapaka et. al. (J. Fluid Mech., vol 609, p285-303, 2008) have used *non-modal stability theory* to obtain rigorous estimates of the growth rates of perturbations in homogeneous porous media. Non-modal stability theory is a mathematically rigorous extension of the traditional normal-mode (eigenvalue) approach to time-dependent systems. We present an extension of this method to account for heterogeneity of the permeability field. In particular, we study the two simplified problems of vertical heterogeneity (layering) and horizontal heterogeneity. We present results for the distribution functions of the critical time as a function of the mean, variance and correlation length of the permeability field.

Authors (2008), Title, Eos Trans. AGU, 89(53), Fall Meet. Suppl., Abstract xxxxx-xx

Hydrology [H]

H13K

MC:2002 Monday 1340h

Multiscale Science of Geologic CO₂ Sequestration III: Monitoring

Presiding: J Kaszuba, University of Wyoming; J Birkholzer, Lawrence Berkeley National Laboratory

H13K-01

Changes in Shallow Groundwater Chemistry Following CO₂ Injection at the ZERT Field Site, Bozeman, Montana

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Approximately 300 kg/day of food-grade CO₂ was injected through a perforated pipe placed horizontally 2-2.3 m deep during July 9-August 7, 2008 at the MSU-ZERT field test to evaluate atmospheric and near- surface monitoring and detection techniques applicable to the subsurface storage and potential leakage of CO₂. As part of this multidisciplinary research project, we collected 80 samples of water from 10 shallow observation wells (1.5 or 3.0 m deep) located 1-6 m from the injection pipe, and from two distant monitoring wells. The samples were collected before, during and following CO₂ injection. Field determinations of pH, alkalinity, conductance and DO, showed rapid, systematic and significant to major changes following CO₂ injection, especially in samples collected from the 1.5 m wells. The collected samples are being analyzed in the laboratory for major, minor and trace cations, metals and anions, as well as DOCs, VOCs and some isotopes. Results obtained to date show major increases in the concentrations of Ca (from 90 to 200 mg/L) Mg (25 to 70 mg/L), Fe (5 to 1000 ppb) and Mn (5 to 1200 ppb) following CO₂ injection. Dissolution of carbonate minerals and desorption-ion exchange resulting from lowered pH values of groundwater (from 7.0 to 5.8) following CO₂ injection, are the likely processes responsible for the observed increases in the concentrations of cations and anions. The concentrations decreased temporarily following the four significant precipitation events that occurred during sampling. The DOC values obtained are generally about 4 mg/L, with a range from 2.6 to 6.9 mg/L; the variations do not seem related to CO₂ injection. The injection of CO₂, however, is clearly responsible for the lowered pH values, mobilization of metals and increases in the values for Ca, Mg, alkalinity and conductance of shallow groundwater

H13K-02

Geoelectric Monitoring Studies for the Carbon Dioxide Geological Storage

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Self-potential (SP) anomalies of negative polarity are frequently observed near deep wells. These anomalies appear to be caused by an underground electrochemical mechanism similar to a galvanic cell: the metallic well casing acts as a vertical electronic conductor connecting regions of differing redox potential. Electrons flow upward through the casing from a deeper reducing environment to a shallower oxidizing environment, and simultaneously a compensating vertical flow of ions is induced in the surrounding formation to maintain charge neutrality. If the redox potential in the deeper region is then increased by injecting an oxidizing substance, the difference in redox potential between the shallower and deeper regions will be reduced, resulting in an SP increase near the wellhead. We have been monitoring earth-surface SP during gas injection tests at various sites in Japan. When air was injected into a 100-meter well within a geothermal field, a remarkable simultaneous increase in SP centered on the wellhead was observed. A small but unmistakable SP increase also took place near the wellhead when CO₂ was slowly injected, which we believe was caused by local pH reduction at depth resulting from dissolution of the injected CO₂ in the aquifer fluid. SP changes were also observed in Yubari, geological sequestration test site in Japan, where one well injected CO₂ into a coal bed and the fluid containing CH₄ was produced from a nearby well. The CO₂ content of the fluid was also monitored. SP increased substantially around the injection wellhead, but no significant SP changes attributable to the injection were observed near the production wellhead. This is consistent with the observation that CO₂ did not break through into the production well during the experiment. We believe that SP measurements at the earth surface represent a new and promising technique for sensing the approach of CO₂ to well casings deep within the subsurface.

H13K-03

Gas Membrane Sensor Technique for in-situ Downhole Detection of Gases Applied During Geological Storage of CO₂

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The geological storage of CO₂ in deep saline aquifers is regarded as a possible technology for the reduction of anthropogenic greenhouse gases. However, comprehensive research is still needed to better understand the behaviour of CO₂ during and after storage. Therefore, we developed and applied a new, innovative geochemical monitoring tool for the real time and in-situ determination of CO₂ and other gases in the underground and in bore holes. The method uses a phase separating silicone membrane, permeable for gases, in order to separate gases dissolved in borehole fluids, water and brines. Argon is used as a carrier gas to conduct the collected gases through capillaries to the surface. Here, the gas phase is analyzed in real-time with a portable mass spectrometer for all permanent gases. In addition, gas samples may be collected for detailed investigations in the laboratory. Downhole extraction and on-line determination of gases dissolved in brines using this gas membrane sensor (GMS) technique was successfully applied at the scientific CO₂SINK test site in Ketzin, Germany (sandstone aquifer). GMSs together with temperature and pressure probes were installed in two approx. 700m deep observation holes, drilled in 50m and 100m distance from the CO₂ injection well. Hydraulic pressure in the observation wells rose gradually during injection of CO₂. Increasing reservoir gas concentrations of helium, hydrogen, methane, and nitrogen as well as the arrival of the added krypton tracer were determined shortly before the injected CO₂ appeared. The breakthrough of CO₂ into the observation well, in 50m distance, was recorded after 531.5 tons of CO₂ were injected.

H13K-04

Isotopic Approaches to Evaluate the Fate of Injected CO₂ in Two Geological Storage Projects in Mature Oilfields in Canada

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Monitoring and verification of CO₂ storage is an essential component of geological storage projects. We present evidence from two enhanced oil recovery projects in Canada that geochemical and isotopic techniques can be successfully used to trace the fate of injected CO₂. Geochemical and isotopic data for fluids and gases obtained from multiple wells at the International Energy Agency Greenhouse Gas Weyburn CO₂ Monitoring and Storage Project (Saskatchewan, Canada) and from the Penn West Pembina Cardium CO₂-Enhanced Oil Recovery Monitoring Pilot (Alberta, Canada) were collected before and throughout the CO₂ injection phase. Carbon isotope ratios of injected CO₂ in the Weyburn project were significantly lower than those of background CO₂ in the reservoir. In contrast, carbon isotope ratios of injected CO₂ at Penn West's Pembina Cardium CO₂-Enhanced Oil Recovery Monitoring Pilot were markedly higher than those of background CO₂. After commencement of CO₂ injection, the concentrations and carbon isotope values of CO₂ and HCO₃⁻ in fluids and gases repeatedly obtained from monitoring wells were determined. Increasing CO₂ and HCO₃⁻ concentrations in concert with carbon isotope values trending towards those of the injected CO₂ revealed effective solubility and ionic trapping of injected CO₂ at several monitoring wells at both study sites. In addition, changes in the oxygen isotope values of reservoir fluids provided independent evidence for dissolution of injected CO₂ in the produced waters. We conclude that geochemical and isotopic monitoring techniques can play an essential role in verification of CO₂ storage provided that the isotopic composition of the injected CO₂ is distinct.

H13K-05

Midwest Regional Carbon Sequestration Partnership Appalachian Basin Test Site: Developing a Sequestration Site from Concept through Injection

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The Midwest Regional Carbon Sequestration Partnership (MRCSP) Appalachian Basin Field Test is located at the First Energy Generation Corp. RE Burger Power Plant in Belmont County, Ohio. The goal at this site is to injection up to 3000 tonnes of carbon dioxide in up to three separate geologic formations. We present the development of this injection plan as more data was collected and added to the system. In addition, we present initial injection results. Site characterization consisted of a regional geological assessment and a 2D seismic survey. A test injection well (FEGENCO 1) was completed in early 2007 and data collected from this well, included geophysical wireline logs and core samples, were used to develop an injection plan. Two previously identified injection targets were analyzed, the Devonian Oriskany Sandstone and the Silurian Clinton Sandstone. Both of these sandstones are regional sequestration targets across the Midwestern United States. In addition to these, a third injection target was identified after drilling. The Silurian Salina Group is regionally extensive throughout most of the Midwest and consists of carbonate and evaporate layers. In the FEGENCO 1 well, one of the subgroups was found to have higher porosity dolomitic stringers sandwiched between anhydrite layers. Wireline data and field samples were used to better understand the geologic model

and predict the porosity and permeability distribution of the interval. Injection is expected to be completed by Fall 2008. This work was done as part of the Midwest Regional Carbon Sequestration Partnership (MRCSP); DOE/NETL Cooperative Agreement No. DE-FC26-05NT42589

H13K-06

Coupled flow and mechanical simulation as an aid in monitoring overburden pressure during geologic carbon sequestration

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The development of monitoring technologies for carbon sequestration is a key element to reducing risks to acceptable levels. A key need for such technology is the development of simulation tools that can help interpret the monitoring data that are gathered. As carbon dioxide is injected into a formation, the pressures within that formation and in overlying and underlying rock formations change, both due to the transmission of pressure within the fluid and rock mediums. As demonstrated in previous studies, the overburden pressure response depends on the permeability of the target formation and surrounding rock layers and the geomechanical properties of surrounding strata. The pressure response in an overlying reservoir layer may be an indicator of the presence of absence of carbon dioxide migration outside of the target reservoir. In this study, several hypothetical cases of fractures in the cap rock were considered. A "monitoring layer" in the overburden is defined as a highly permeable layer above the primary caprock. This study tests the variation in pressure response in the monitoring layer in the presence and absence of a fracture in the caprock. An overburden response during injection of carbon dioxide was simulated by considering coupled flow and deformation behavior within the nearby rock strata. Since the pressure response is perpetrated through the brine much faster than the CO₂, single-phase flow assumptions were used to simplify the calculations. Several parameters, such as fracture and matrix permeabilities, elastic moduli, layer thicknesses, and fracture location were varied to test the effect on pressure response. The results from this and further studies can be useful in the development of monitoring techniques for evaluating long-term performance of carbon storage sites.

H13K-07 INVITED

Lessons Learned from Ongoing Field Tests of Geologic CO2 Sequestration

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We present lessons learned □ an attempt to describe what we know and do not know □ based on ongoing field tests of geologic carbon sequestration. The Southwest Regional Partnership on Carbon Sequestration, funded by the U.S. Department of Energy and managed by DOE's National Energy Technology Laboratory, is conducting three separate field tests of geologic sequestration that include extensive monitoring and analysis of the fate of injected CO₂. The CO₂ injection sites include the Aneth oilfield in southern Utah, the coalbed "fairway" in the San Juan basin in northern New Mexico, and the SACROC oilfield in the Permian basin of west Texas. Results of the ongoing sequestration field tests are both encouraging and problematic. At the San Juan basin coalbed injection test, we forecasted coalbed swelling following injection to be detectable at the surface. Tiltmeter results indicated subsidence, not uplift, and poroelastic models of the site suggest that swelling is likely occurring, but cleat compaction may be responsible for the net subsidence. In a similar context, initial poroelastic models of the Aneth, Utah injection site suggested minimal rock strain would be induced by the 100,000 tons of CO₂ injected over the past year, but this forecast is belied by daily microearthquakes recorded at the site (albeit very small events: M -1 to 0). On the other hand, our initial multiphase flow models of the Aneth site provided forecasts of CO₂ migration that turned out to be extremely consistent with observed tracer test results, suggesting that our estimated permeability distributions and other model parameters were effective to some extent. These field tests suggest that probably the greatest challenges are (1) verification or confirmation of trapping mechanisms, and (2) monitoring of processes in the "intermediate zone," the section of strata above the sequestration formation topseal unit and below the upper 100 m of the section, (3) developing meaningful geologic characterization with sparse data, (4) developing meaningful risk assessment frameworks, especially quantification of consequences of "failed" sequestration. For example, regarding verification of trapping mechanisms, we developed detailed, quantitative forecasts of trapping mechanisms, including hydrostratigraphic trapping, solubility trapping, residual gas trapping and mineral trapping, for the SACROC field, using models calibrated with 30 years of past CO₂ injection data. However, verifying these forecasts is next to impossible.

Authors (2008), Title, Eos Trans. AGU, 89(53), Fall Meet. Suppl., Abstract xxxxx-xx

Hydrology [H]

H14C

MC:2002 Monday 1600h

Multiscale Science of Geologic CO₂ Sequestration IV: Leakage and Geomechanics

Presiding: R Pawar, Los Alamos National Laboratory; D DiCarlo, The University of Texas at Austin

H14C-01

Direct Numerical Simulation of Fluid Driven Geomechanical Events during Carbon Sequestration

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Geologic storage of supercritical CO₂ introduces significant stress perturbations into the target formation and overburden. In some cases, this could activate existing fractures and faults, or drive new fractures through the caprock. We will present results of a recent investigation to identify conditions that will activate existing fractures/faults or make new fractures within the caprock using a range of computational tools. Understanding the geomechanical sources of risk to successful CO₂ containment involves a wide range of scales. For example, at the largest scale, bounding fault stability must be considered. Many proposed sequestration targets are bounded by impermeable fault zones that are presumed to become flow paths if they slip. Such geologic features are activated at the field scale by pore pressure elevations. In contrast, fluid driven fracturing events that may introduce new flow paths across a caprock occur at smaller scales. Determining whether the creation of new fractures in the caprock leads to CO₂ leakage, however, requires knowledge of how the new fracture intersects and interacts with prior networks of fractures within the caprock. A range of computational tools have been developed at LLNL to consider these scenarios at the most appropriate scales and including the most appropriate physical models. For example, the Livermore Distinct Element Code (LDEC) has been used to simulate the mechanical deformation of fracture networks extending up to 100m. Frac-HMC was developed to evaluate network permeability evolution at the scale of many tens of meters incorporating both mechanical and chemical effects. Massively parallel, non-linear continuum codes that use a smeared fault representation have also been developed to predict activation of multiple faults at the field scale. Most recently, LDEC has been extended to simulate hydraulic driven fracture events at the scale of individual fractures. We will present results spanning the range of small scale fluid driven fracturing to field scale fault activation. We will discuss our emerging framework for integrating these results across multiple scales to build understanding of how CO₂ storage integrity is potentially controlled by small scale processes. For example, fluid driven fracturing can lead to percolation across the caprock, but evaluating the probability of such an event involves understanding the relationship between the new fracture and the prior fracture network. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

H14C-02

Coupled Reservoir-Geomechanical Analysis of CO₂ Injection Performance at In Salah, Algeria

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In Salah Gas Project in Algeria has been injecting nearly one million tonnes CO₂ per year over the past three years into a water-filled strata at a depth of about 1,800 m. Unlike most CO₂ storage sites, the permeability of the storage formation is relatively low and comparable thin with a thickness of about 20 m. To mitigate high injection pressures, the In Salah Gas Project decided to use long-reach (about 1 to 1.5 km) horizontal injection wells. In this study we are using field data and coupled reservoir-geomechanical numerical modeling of CO₂ injection to analyze geomechanical responses and to assess the effectiveness of this approach for CO₂ storage in relatively low permeability formations. We use satellited-based measurements of surface uplift to constrain our coupled reservoir-geomechanical model. We conduct sensitivity studies to investigate whether the observed uplift can be explained by poro-elastic expansion within the 20-m thick reservoir or if there is a significant contribution from pressure changes within the adjacent caprocks. We investigate whether surface deformations from InSAR can be useful for detection of a permeable leakage paths through the overlying caprock layers.

H14C-03

Leaking And Non-leaking Systems: Study Of Natural CO₂ Accumulations For Geological Sequestration

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The potential risks of geological CO₂ storage must be understood and geologists are required to predict how CO₂ may behave once stored underground. As natural geological accumulations of carbon dioxide occur in many basins in Italy and volcanic and seismically active areas allow CO₂ rich fluids to migrate to the near surface, many of these areas have been investigated in order to study long-term geochemical processes that may occur following geological storage of anthropogenic CO₂. A study representing an example of "leaking" system is the Solfatara crater (Campi Flegrei, Southern Italy) characterised by

the presence of both CO₂ rich-waters and fumarole. Soil gas flux measurements show that the entire area discharges between 1200 and 1500 tons of CO₂ a day. Most part of analysed waters is the effect of a mixing between a shallow meteoric water and a deep thermal Na-Cl end-member and/or seawater, resulting in sodium-chloride waters. A high dissolved CO₂ content (max value 566.28 cc/l) is also present. Furthermore, the Campi Flegrei frequently undergo bradyseism related to the elastic response of the shallow crust to increasing pressure within a shallow magma chamber. The study of this phenomenon could be useful to detect ground deformation linked to geomechanical changes in a geological CO₂ reservoir. In contrast, an example of "non-leaking" system is the Pisticci oil and gas Field (Southern Italy) where a great variety of hydrocarbons traps are formed by horst and tilted blocks in the Mesozoic carbonate substratum covered by an almost continuous sequence of Lower Pliocene marls and Middle Pliocene-Pleistocene marly blue clays. Soil gas surveys were performed after a MD 4.5 earthquake and two years later to test the permanence of the gas distribution pattern. CO₂ distribution in soil gas seems not to be affected by changes in stress, as suggested by the average values of both surveys. The principal aim of our research has been to evaluate and mitigate risks for local populations as the studied areas are densely populated. To date, the obtained results suggest that gas uprising is generally well localised around restricted areas, often controlled by local tectonics (faults and/or fractures). This implies that, in the frame of geological CO₂ sequestration, it is necessary to carefully assess the presence of pathways (fault and/or fractures) that might allow the migration of CO₂ out of the reservoir.

H14C-04

Numerical analysis of wellbore integrity: results from a field study of a natural CO₂ reservoir production well

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An important aspect of the risk associated with geological CO₂ sequestration is the integrity of existing wellbores that penetrate geological layers targeted for CO₂ injection. CO₂ leakage may occur through multiple pathways along a wellbore, including through micro-fractures and micro-annuli within the "disturbed zone" surrounding the well casing. The effective permeability of this zone is a key parameter of wellbore integrity required for validation of numerical models. This parameter depends on a number of complex factors, including long-term attack by aggressive fluids, poor well completion and actions related to production of fluids through the wellbore. Recent studies have sought to replicate downhole conditions in the laboratory to identify the mechanisms and rates at which cement deterioration occurs. However, field tests are essential to understanding the *in situ* leakage properties of the millions of wells that exist in the mature sedimentary basins in North America. In this study, we present results from a field study of a 30-year-old production well from a natural CO₂ reservoir. The wellbore was potentially exposed to a 96% CO₂ fluid from the time of cement placement, and therefore cement degradation may be a significant factor leading to leakage pathways along this wellbore. A series of downhole tests was performed, including bond logs and extraction of sidewall cores. The cores were analyzed in the laboratory for mineralogical and hydrologic properties. A pressure test was conducted over an 11-ft section of well to determine the extent of hydraulic communication along the exterior of the well casing. Through analysis of this pressure test data, we are able to estimate the effective permeability of the disturbed zone along the exterior of wellbore over this 11-ft section. We find the estimated range of effective permeability from the field test is consistent with laboratory analysis and bond log data. The cement interfaces with casing and/or formation are the most likely pathway by comparison of test data to cement core analysis. The results of this work demonstrate that field pressure tests can be an effective means to estimate effective permeabilities along existing wellbores, thus providing an important tool for managing the risk of geological CO₂ sequestration.

H14C-05

Assessing the Leakage Potential of CO₂ into Ground Water Resources at SACROC, West Texas

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In this paper we apply CO₂-PENS to characterize the long-term CO₂ storage performance at SACROC, specifically with respect to potential impacts on ground water resources located above the CO₂ injection depth. CO₂-PENS is a hybrid process/system model composed of a library of process level modules linked together to allow simulations to span a range of spatial and temporal scales. SACROC is the oldest CO₂-EOR operation in the Permian Basin and has been operational for over 35 years. We describe how CO₂-PENS is used to couple CO₂ migration at three different scales; from a reservoir module, through a borehole leakage module, and into an aquifer impact module. Both the reservoir module and wellbore leakage module are based on abstractions of the underlying physics of multiphase fluid flow, using reductions in complexity to allow fast Monte-Carlo simulation while capturing the behavior of these processes. Finally, we show how results from CO₂-PENS can be used to delineate areas that are susceptible to ground water impacts from CO₂ leakage and discuss how this information can be used in risk analysis.

H14C-06

Prediction of Groundwater Quality Changes in Response to CO₂ Leakage from Deep Geological Storage

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If carbon dioxide stored in deep saline aquifers were to leak into overlying aquifer containing potable groundwater, the intruding CO2 would lower groundwater pH and could enhance the solubility of hazardous inorganic constituents present in the aquifer minerals. As an effort to evaluate risks associated with geologic sequestration of CO2, this work assesses these potential effects using reactive transport modeling. A systematic geochemical evaluation of more than 38,000 groundwater quality analyses from aquifers throughout the United States provided the prerequisites for reactive transport modeling. For example, galena (under reducing conditions) and cerussite (under oxidizing conditions) control aqueous Pb (lead) whereas arsenopyrite component in pyrite controls aqueous As (arsenic) generally under reducing conditions. Reactive transport simulations are performed which focus on the chemical evolution of Pb and As in the groundwater after the intrusion of CO2. The simulations use representative mineralogies for shallow potable aquifers in the USA and two measured mineralogies for deep confined aquifers. The resulting concentrations of Pb and As in the groundwater are then compared to the EPA specified health-based limits for drinking water. A significant increase of aqueous Pb and As occurs, although in most situations they remain below health-based limits. Sensitivity studies are also conducted for variation in hydrological, geochemical and mineralogical conditions and several critical parameters. The results indicate that aquifers containing more carbonate (through pH buffer) and clay minerals (by adsorption) are less vulnerable to CO2 intrusion. Adsorption/desorption from minerals surface significantly impact the mobilization of Pb and As. Adsorption dampens the effect of galena and arsenopyrite dissolution by removing Pb and As from aqueous phase under reducing conditions. Under oxidizing condition desorption is primarily responsible for increasing the concentration of aqueous Pb while precipitation of cerussite downstream stabilize aqueous Pb concentration.

H14C-07

CO2 Leak Detection Using Hyperspectral Plant Signatures During the 2008 ZERT CO2 Sequestration Field Experiment in Bozeman, MT

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Geologic sequestration of carbon dioxide is a potential method of mitigating increased carbon dioxide (CO2) emissions into the atmosphere. However, the impact CO2 sequestration would have on various physical, chemical, and biological systems needs to be explored in order to effectively utilize this technique long term. We used plants to detect the effects of releasing pure carbon dioxide gas through a 100-meter long horizontal well, varying between 1 and 2.5 meters below ground surface, at a flow rate of 0.3 tons per day for 29 days, beginning on July 9th, 2008. An influx of CO2 gas into the soil induces stress on vegetation, which can be observed in their visible to near infrared reflectance spectral signatures. We followed the health of a 20 meter by 30 meter patch of plants (including alfalfa, dandelion, Timothy grass, Tall fescue grass, and Orchard grass), located along and across the injection well. 68 sites were chosen within the plant patch where spectral signatures were measured almost daily with an Analytical Spectral Devices "FieldSpec Pro" spectrometer. These sites were located over the injection well and up to 10 meters off the well. This array gave us data both within and outside of the CO2 leak zone so we could normalize our observations for all other environmental factors affecting the plants. On August 5, 2008, airborne hyperspectral imagery was also acquired from a low flying aircraft using a hyperspectral camera developed by Resonon, Inc. of Bozeman, MT. In addition, other groups participating in the ZERT experiment measured various parameters of the field site, such as CO2 flux mapping, soil CO2 concentrations, atmospheric CO2 concentrations, plant isotopics, soil moisture, groundwater chemistry, and depth to groundwater. Four to five days after the start of the CO2 injection, stress was observed in the spectral signatures of plants within 1 meter of the well. After approximately ten days of CO2 injection, plants located up to 2.5 meters from the well exhibited at least moderate amounts of stress. Locations of plant stress corresponded to areas of high measured CO2 flux. In addition, correlating the spectral signatures to adjacent soil CO2 concentration probes gave a lower limit to the CO2 concentration needed to act as a stressor on the plants. During the injection, soil probes located over the well and at 2.5 meters north of the well measured CO2 concentrations that equilibrated at approximately 13.5% and 8% by volume, respectively. The spectra of the plants near these soil probes exhibited some degree of stress. The other soil probes farther from the well (at 5, 7.5 and 10 m) did not measure CO2 soil concentrations above 4% by volume, and none of the plants adjacent to those probes displayed discernable stress. We infer that the lower limit of CO2 needed to stress the plants over a period of 30 days is between 4% and 8% by volume.

H14C-08

Multispectral imaging of plant stress for detection of CO2 leaking from underground

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Multispectral imaging of plant stress is a potentially useful method of detecting CO2 leaking from underground. During the summers of 2007 and 2008, we deployed a multispectral imager for vegetation sensing as part of an underground CO2 release experiment conducted at the Zero Emission Research and Technology (ZERT) field site near the Montana State University campus in Bozeman, Montana. The imager was mounted on a low tower and observed the vegetation in a region near an underground pipe during a multi-week CO2 release. The imager was calibrated to measure absolute reflectance, from which vegetation indices were calculated as a measure of vegetation health. The temporal evolution of these indices over the course of the experiment show that the vegetation nearest the pipe exhibited more stress than the vegetation located further from the pipe. The imager observed notably increased stress in vegetation at locations exhibiting particularly high flux of CO2 from the ground into the atmosphere. These data from the 2007 and 2008 experiments will be used to demonstrate the utility of a tower-mounted multispectral imaging system for detecting CO2 leakage from below ground with the ability to operate

continuously during clear and cloudy conditions.

Authors (2008), Title, Eos Trans. AGU, 89(53), Fall Meet. Suppl., Abstract xxxxx-xx

Hydrology [H]

H23D

MC:Hall D Tuesday 1340h

Multiscale Science of Geologic CO₂ Sequestration V Posters

Presiding: G Thyne, Enhanced Oil Recovery Institute, University of Wyoming; J Birkholzer, Lawrence Berkeley National Laboratory

H23D-0984

Versatile and Sensitive Surface Monitoring of Deep Gas Leakage Using Soil Radon Concentration Dynamics

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The natural sealing integrity of reservoirs for carbon dioxide sequestration in deep geologic formations is a key issue for the relevance of storage projects, and public acceptance as well. Natural soil gas anomalies (He, Rn and carbon dioxide, etc.) are closely related to structural discontinuities which act as pathways for earth degassing. Accordingly they should be reliable targets for tracking gas regime modification at depth. Direct monitoring of carbon dioxide at ground surface needs gas sampling, at least of the order of magnitude of natural soil degassing. This is unacceptable as regard the search of the tiny first anomalies of the possible gas regime modification. Moreover, in such contexts, natural carbon dioxide gas flow, measured at 2 m depth, is at least one order of magnitude lower than surface flow, and is much smaller than seasonal surface natural variability. These factors rule out ground surface carbon dioxide monitoring for detection of very first anomalies of deep degassing. Monitoring of natural soil radon carried by rising gases allows getting over this difficulty. Since more than a decade we experienced solid state silicon detectors devices in contrasted environments, which allow permanent static records of radon concentration in soil air. Design of a new probe, using three radon sensors inside an elongated chamber inserted into the ground allows calculating the gas flow entering the device. Phase differences of radon signals recorded by the sensors, measured with a 15 minute step allows calculating both flow with a temporal resolution of some hours and mean vertical gas velocity at depth with a resolution of some weeks. Difficulties linked with air dilution entering the chamber, low signal/noise ratios, weak flows, etc. are solved using analysis of the cross correlogram of the respective radon time series. In the absence of advection, the cross correlogram reveals chance correlations between the various signals or those resulting from homogenization process (especially diffusion), and the symmetry of the cross correlogram shows that these correlations take place upwards and downwards. In the presence of an upward advective flow, the cross correlogram shows a positive off-set larger than negative off-set, because correlations resulting from the gas entering the probe is added to the chance. In addition, modelling of radon signal with meteorological parameters (atmospheric pressure, rain and temperature, etc.) which have a primordial action on soil gas composition and then its radon concentration allows explaining most of the recorded variations of soil radon concentration. However, some transient events recorded in Southern Saskatchewan are suggested to be correlated with deep crustal stress change, related with seismic or micro-seismic events.

H23D-0985

Dynamics of CO₂ Concentrations and Fluxes During the 2008 ZERT Project Shallow Subsurface CO₂ Release

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From 9 July to 7 August 2008, 0.3 t CO₂ d⁻¹ were released from a 73-m long, ~2.5 m deep horizontal well located in an agricultural field at Montana State University, Bozeman, MT. We measured soil CO₂ fluxes using the chamber method on a grid repeatedly on a daily basis. Infrared sensors installed at 30 cm depth in the soil at 0, 2.5, 5, 7.5, and 10 m NW of the well and at 4 cm above the ground surface at 0 and 5 m NW of the well continuously measured soil and atmospheric CO₂ concentrations. Environmental parameters were concurrently monitored. CO₂ fluxes ranged from 5 to 7756 g m⁻² d⁻¹ and indicated that leakage broke through to the surface within 2 concentrations ranged from 350 to >2000 ppmv; anomalously high values were observed after five hours following the start of release and suggested flow of leakage-derived CO₂ along the ground surface. Soil CO₂ concentrations ranged from 0.1 to 14.0 vol.%; anomalously high values were measured after ~one, two, and seven days following the release start by sensors at 0, 2.5, and 5 m from the well, respectively. Low (<0.5 cycle d⁻¹), high (>4 cycle d⁻¹), and band pass (1-2 cycle d⁻¹) filters were applied to soil CO₂ concentration and environmental parameter time series and showed that variations in concentrations at (1) low frequency were strongly correlated with CO₂ release rate at 0-5 m sensors, (2) moderate (diurnal) frequency were strongly correlated with variations in soil moisture and temperature, barometric pressure, and wind speed at all sensors, and (3) high frequency were moderately correlated with variations in soil moisture and temperature, barometric pressure, and wind speed at all sensors and strongly correlated with abrupt unintended changes in CO₂ flow rate in the well at 0-5 m sensors. Implications for near-surface monitoring of geologic carbon storage projects will be discussed.

H23D-0986

Sensitivity Analysis of Key Parameters on the Behavior of CO₂ Injected Into a Deep Saline Aquifer

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Geological storage of CO₂ is one of the methods proposed for mitigating global warming. Numerical simulations can be useful for assessing its positive and/or negative impact, but to perform realistic simulations of injected CO₂ behavior we need field information such as geological structure, the hydrological and mechanical properties of the underground formations, the chemical properties of the native fluids, the subsurface distributions of pressure and temperature, the locations of faults, etc. These parameters are essential for designing numerical models, and model results may be sensitive to small variations in the values assumed. We carried out sensitivity analyses to study the effect of these key parameters on the long-term behavior of injected CO₂ using the "STAR" reservoir simulation code. We used the new "SQSCO2" fluid constitutive module (Pritchett, 2008) which represents the thermodynamics and thermo-physical properties of H₂O-NaCl-CO₂ mixtures over the range from liquid-CO₂ to supercritical-CO₂ conditions including the three-phase region (liquid CO₂, gaseous CO₂ and the saline aqueous phase containing dissolved CO₂). We constructed a simple two-dimensional model representing 2000 meters of alternating sandstone and shale layers based broadly upon the geological structure underlying the Tokyo Bay area in Japan. The thickness of each layer is 100 m in the base case. We simulated 50 years of injection followed by 1000 years of shut-in. The computed results include the amount of CO₂ trapped by the dissolution and residual gas mechanisms over time and the evolution of the CO₂ plume. The results indicate that the geothermal gradient, the injection depth, the permeabilities of the formations, the residual-gas saturation in the aquifer, the capillary pressure in the seal layer, the formation dip and the salinity of the native water all have significant impacts on long-term CO₂ behavior. Reliable information concerning these parameters is essential for predicting the eventual fate of CO₂ injected into saline aquifers.

H23D-0987

Fully Coupled Thermo-Hydro-Mechanical Numerical Simulation of Geologic Storage of Carbon Dioxide in Layered and Folded Geologic Media

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A series of numerical simulations using a fully coupled multiphase thermo-hydro-mechanical (THM) numerical model is performed to analyze groundwater and carbon dioxide flow, heat transport, and land deformation in geologic media due to carbon dioxide injection and to evaluate their thermo-hydro-mechanical stability for geologic storage of carbon dioxide. The geologic media are composed of a series of Jurassic sandstone aquifer (reservoir rock) and shale aquitard (cap rock) layers, which are layered and folded, over Precambrian metamorphic rocks at two oblique angles. Two different cases of boundaries between the sedimentary and metamorphic rocks are simulated to evaluate effects of geologic structures on geologic storage of carbon dioxide. One is a pair of faults, and another is a pair of unconformities. Four different locations of carbon dioxide injection are also simulated to evaluate an optimal location of injection in each boundary case. The numerical simulation results show that the layered heterogeneity and the geologic structures such as folds, faults, and unconformities have significant effects on the spatial distributions and temporal changes of groundwater pressure and saturation, carbon dioxide pressure and saturation, geothermal temperature, and land displacement vector. The free phase carbon dioxide (structural trapping), which is injected into sandstone, moves upward along the interfaces between the sandstones and shales and then accumulates under the anticlines. Over a long period of time, the free phase carbon dioxide (residual trapping) and the carbon dioxide dissolved in groundwater (solubility trapping) also move along the groundwater flow direction and then leak to the ground surface through the faults and unconformities. In case of the fault boundaries, such leakage becomes more accelerated and intensified. On the other hand, land deformation with ground surface uplift occurs during the injection period, and then the ground surface recovers to its initial state in accordance with the recovery of groundwater pressure after the injection period. Therefore it may be concluded that the layered heterogeneity and the geologic structures such as folds, faults, and unconformities cannot always be ignored if they are observed in actual geologic systems, and thus they must be properly characterized and considered when more rigorous and reasonable predictions of both long-term thermo-hydro-mechanical responses of the whole geologic systems to carbon dioxide injection and their storage stability are to be obtained. Further numerical studies of various geologic and hydrogeologic settings and field applications are recommended to arrive at more general conclusions concerning the effects of the layered heterogeneity and the geologic structures on multiphase fluid flow, heat transport, and land deformation due to carbon dioxide injection.

H23D-0988

Computational Study of the Scaling relationship between Fluid Flow and Fracture Stiffness

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To sequester CO₂ in the subsurface requires an understanding of the relationships among physical processes that occur on multiple length and time scales. If seismic techniques are to be developed to monitor the injection and containment phases of CO₂ sequestration, it is important to understand not only how the microscopic behavior affects macroscopic measurements, but to determine how measurements made on the laboratory scale may be relevant to the field scale. We performed a computational study to investigate the scaling behavior of the interrelationship between the fracture specific stiffness and fluid flow

through fractures to promote development of active seismic monitoring techniques to quantify the time-dependent changes in a subsurface CO₂ reservoir. The relationship between the hydraulic and seismic properties of fractures is based on the empirical relationship between fluid flow through a fracture and fracture specific stiffness. Experimental work has shown that fracture stiffness and fluid flow through a fracture both depend on the topology of the fracture. In our study, we focus on the role of fracture in expressing the fracture stiffness \square flow fluid relationship in scaling form. The geometry of fractures were simulated using stratified continuum percolation that constructs a hierarchical aperture distribution with a tunable spatial correlation length. Correlation lengths were varied from a quarter of the fracture length to the full length of the fracture. The dimensions of the fractures ranged from 1024 to 32 pixels (1 m to 0.03 m) to observe scaling behavior. The spatial and size distribution of the aperture and contact area for each simulated fracture were quantified along with percolation probabilities, percolation cluster statistics and other geometric parameters. The fracture specific stiffness was determined using an algorithm that models the asperities of the fracture surface on a regular lattice grid. The half-spaces and asperities deformed elastically. Each asperity in contact interacts with all of the other asperities, leading to a system of coupled linear equations. The fracture specific stiffness is extracted from the displacement-stress curves. Single phase fluid flow simulations were performed on each fracture for each increment of stress as well. The fractures were modeled as a bilateral network of pipes. The conductance of the pipes was calculated based on the analytic solution for flow between two parallel plates, the "cubic" law. Once the pressure at each location is calculated, the fluxes are easily calculated. The flow and stiffness calculations were performed at six different scales. By doing this, we are able to use the renormalization group framework to calculate percolation thresholds and critical exponents of the system and how they directly relate to the topology of the fracture. Acknowledgments: The authors wish to acknowledge Joe Morris for his assistance with code development. This work is supported by the Geosciences Research Program, Office of Basic Energy Sciences US Department of Energy (DEFG02-97ER14785 08) and is funded in part by the Geo-mathematical Imaging Group at Purdue University.

H23D-0989

Co-injection of SO₂ With CO₂ in Geological Sequestration: Potential for Acidification of Formation Brines

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Coal-fired power plants produce flue gas streams containing 0.02-1.4% SO₂ after traditional sulfur scrubbing techniques are employed. Due to the corrosive nature of H₂SO₄, it will likely be necessary to remove the residual SO₂ prior to carbon capture and transport; however, it may still be economically advantageous to reintroduce the SO₂ to the injection stream to mitigate the cost of SO₂ disposal and/or to get credits for SO₂ emissions reduction. This study examines the impact of SO₂ co-injection on the pH of formation brine. Using phase equilibrium modeling, it is shown that a CO₂ gas stream with 1% SO₂ under oxidizing conditions can create extremely acidic conditions (pH<1), but this will occur only near the CO₂ plume and over a short time frame. Nearly all of the SO₂ will be lost to the brine during this first phase equilibration, within approximately a decade, and the pH after the second is only 3.7, which is the pH that would occur from the carbonic acid alone. This suggests that although SO₂ will create low pH values due to the formation of H₂SO₄, the effect will have a very limited lifespan and a localized impact spatially. SO₂ is much more soluble than CO₂ and as the relative amount of SO₂ to CO₂ is very small, the SO₂ will quickly dissolve into the formation brine. The extent of H₂SO₄ formation is dependent on the redox conditions of the system. Several SO₂ oxidation pathways are investigated, including SO₂ disproportionation which produces both sulfate and the weaker acid, H₂S. Further modeling considers a time varying, diffusion limited flux of SO₂. Relative to the case of instantaneous phase equilibrium, this results in a smaller decrease in pH occurring over a longer duration. Our overall conclusion is that brine acidification due to SO₂ co-injection is not likely to be significant over relevant time and spatial scales.

H23D-0990

Feasibility Assessment of CO₂ Sequestration and Enhanced Recovery in Gas Shale Reservoirs

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CO₂ sequestration and enhanced methane recovery may be feasible in unconventional, organic-rich, gas shale reservoirs in which the methane is stored as an adsorbed phase. Previous studies have shown that organic-rich, Appalachian Devonian shales adsorb approximately five times more carbon dioxide than methane at reservoir conditions. However, the enhanced recovery and sequestration concept has not yet been tested for gas shale reservoirs under realistic flow and production conditions. Using the lessons learned from previous studies on enhanced coalbed methane (ECBM) as a starting point, we are conducting laboratory experiments, reservoir modeling, and fluid flow simulations to test the feasibility of sequestration and enhanced recovery in gas shales. Our laboratory work investigates both adsorption and mechanical properties of shale samples to use as inputs for fluid flow simulation. Static and dynamic mechanical properties of shale samples are measured using a triaxial press under realistic reservoir conditions with varying gas saturations and compositions. Adsorption is simultaneously measured using standard, static, volumetric techniques. Permeability is measured using pulse decay methods calibrated to standard Darcy flow measurements. Fluid flow simulations are conducted using the reservoir simulator GEM that has successfully modeled enhanced recovery in coal. The results of the flow simulation are combined with the laboratory results to determine if enhanced recovery and CO₂ sequestration is feasible in gas shale reservoirs.

H23D-0991

The Effect of CO₂ Injection on the Compressional Strength of Anhydrite

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Geological storage of CO₂ in clastic reservoirs and aquifers offers one of the most promising ways of disposing of anthropogenic carbon dioxide, hence reducing CO₂ emissions. The injection of CO₂ into such formations is expected to have a variety of coupled geochemical and geomechanical effects on both the host and caprock. To maintain reservoir integrity the behaviour of the caprock is particularly important. We performed triaxial compression experiments on anhydrite cores, obtained from the base of the Dutch Zechstein, which acts as a caprock to many onshore gas reservoirs. Our aim was to determine the effect of CO₂-saturated water on the strength of the material, compared to dry and water-wet conditions. Experiments were performed at 20 and 80°C, at effective confining pressures up to 50 MPa, pore fluid pressures of 15 MPa and a strain rate of ~10⁻⁵ s. The cores consisted of anhydrite and contained approximately 10-30 wt% dolomite. The material had a permeability of less than 0.02 mD and a porosity of ~0.3%. We have constructed the failure envelope for anhydrite under dry conditions, at room temperature and 80°C. At room temperature, the transition between brittle and brittle/ductile behaviour was observed at confining pressures of 5 to 10 MPa. Increasing temperature to 80°C weakened the material, pushing the brittle-ductile transition to higher confining pressures. We observed that the cores did not become sufficiently permeable to saturated CaSO₄ solution until near failure. The strength of the material was not significantly affected by the addition of saturated CaSO₄ solution. The presence of CaSO₄ solution saturated with CO₂ at a pressure of 15 MPa showed no significant effect on the material strength.

H23D-0992

Analysis of Geologic CO₂ Sequestration at Farnham Dome, Utah, USA

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The Farnham Dome in east-central Utah is an elongated, Laramide-age anticline along the northern plunge of the San Rafael uplift and the western edge of the Uinta Basin. We are helping design a proposed field demonstration of commercial-scale geologic CO₂ sequestration, including injection of 2.9 million tons of CO₂ over four years time. The Farnham Dome pilot site stratigraphy includes a stacked system of saline formations alternating with low-permeability units. Facilitating the potential sequestration demonstration is a natural CO₂ reservoir at depth, the Jurassic-age Navajo formation, which contains an estimated 50 million tons of natural CO₂. The sequestration test design includes two deep formations suitable for supercritical CO₂ injection, the Jurassic-age Wingate sandstone and the Permian-age White Rim sandstone. We developed a site-specific geologic model based on available geophysical well logs and formation tops data for use with numerical simulation. The current geologic model is limited to an area of approximately 6.5x4.5 km² and 2.5 km thick, which contains 12 stacked formations starting with the White Rim formation at the bottom (>5000 feet bgl) and extending to the Jurassic Curtis formation at the top of the model grid. With the detail of the geologic model, we are able to estimate the Farnham Dome CO₂ capacity at approximately 36.5 million tones within a 5 mile radius of a single injection well. Numerical simulation of multiphase, non- isothermal CO₂ injection and flow suggest that the injected CO₂ plume will not intersect nearby fault zones mapped in previous geologic studies. Our simulations also examine and compare competing roles of different trapping mechanisms, including hydrostratigraphic, residual gas, solubility, and mineralization trapping. Previous studies of soil gas flux at the surface of the fault zones yield no significant evidence of CO₂ leakage from the natural reservoir at Farnham Dome, and thus we use these simulations to evaluate what factors make this natural reservoir so effective for CO₂ storage. Our characterization and simulation efforts are producing a CO₂ sequestration framework that incorporates production and capacity estimation, area-of-review, injectivity, and trapping mechanisms. Likewise, mitigation and monitoring strategies have been formulated from the site characterization and modeling results.

H23D-0993

Changes in Geophysical Observables Caused by Carbon Dioxide Injection Into Saline Aquifers

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An appropriate monitoring program is indispensable for an individual geological storage project to aid in answering various operational questions by detecting changes within the reservoir and to provide early warning of potential CO₂ leakage through the caprock. Such a program is also essential to reduce uncertainties associated with reservoir parameters and to improve the predictive capability of reservoir models. Repeat geophysical measurements performed at the earth surface show particular promise for monitoring large subsurface volumes. To appraise the utility of geophysical techniques for monitoring CO₂ injected into aquifers, we carried out numerical simulations of an aquifer system underlying a portion of Tokyo Bay and calculated the temporal changes in geophysical observables caused by changing underground conditions as computed by the reservoir simulation. We used the STAR general-purpose reservoir simulator with the CO₂SQS equation-of- state package (Pritchett, 2005) which treats three fluid phases (liquid- and gaseous-phase CO₂ and an aqueous liquid phase) to calculate the evolution of reservoir conditions, and then used various □geophysical postprocessors□h to calculate the resulting temporal

changes in the earth-surface distributions of microgravity, electrical self-potential (SP), apparent resistivity (from either DC or MT surveys), ground deformation and seismic observables. Of course, the applicability of any particular method is likely to be highly site-specific, but these calculations indicate that none of these techniques should be ruled out altogether. Some survey techniques (gravity, DC resistivity, MT) appear to be suitable for characterizing long-term changes, whereas others (seismic reflection, SP) are quite responsive to short-term disturbances.

H23D-0994

Pore-scale Investigation of Calcite Precipitation and Permeability Changes in Silicon- based Micromodels

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CO₂ sequestration in geologic formations is increasingly being studied as a strategy for limiting CO₂ emission to the atmosphere, but there are potentials for leakage from the reservoirs and uncertainties associated with the environmental impacts. Reactions between dissolved CO₂ and cations in groundwater (e.g., Ca²⁺) may cause mineral precipitations, and hence reductions in permeability. In this study, microfluidic pore structures etched into silicon wafers were used as two-dimensional model groundwater systems (micromodel) to study the mechanisms of mineral precipitation relevant to CO₂ sequestration. Solutions containing CaCl₂ and Na₂CO₃ were introduced into the micromodel through two separate inlets and they mixed along the center of the micromodel. Images of micromodel were taken using a microscope equipped with a digital camera at selected time points. Calcite formation rates and morphology along the mixing zone are determined from images and the impacts of water flowrates and solution concentrations on reaction rates are investigated. Impacts of pore-scale processes, i.e., mixing induced reaction, permeability changes, diffusion and dispersion, will be discussed.

H23D-0995

Solubility and Diffusivity of SO₂ for Co-injection With CO₂ in Geological Sequestration

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There are potential economic benefits to the co-injection of SO₂ with CO₂ in the context of geological sequestration, but the impact of this co-injection on the fate and migration of SO₂ and CO₂ is poorly understood. Previous modeling studies have shown that injection of SO₂ with CO₂ would create highly acidic conditions due to formation of sulfuric acid. However, little is known regarding the solubility of SO₂ under high pressure, high salinity conditions, and the kinetic limitations of SO₂ diffusion in a CO₂ phase. A method to estimate the phase partitioning of SO₂ under geological storage conditions was developed in this study. The method uses the Krichevsky-Ilinskaya equation to correct for high pressures and the Schumpe model for mixed electrolyte solutions. Henry's constants for a broad range of brine solutions were calculated at storage conditions of 100 bar pressure. The Henry's constant for SO₂ is 1.5 M/atm at 40°C and is 0.86 M/atm at 60°C. Under these same conditions, the Henry's constant for CO₂ is much smaller, roughly 0.01 M/atm (40°C to 60°C). Henry's constants increase with increasing pressure but decrease with increasing temperature. These effects can be observed by comparing the SO₂ Henry's constants under storage conditions with the value under ambient temperature and pressure conditions in pure water, 1.2 M/atm. To simulate diffusion through stationary CO₂, a non-steady state two-dimensional model of SO₂ diffusion through supercritical CO₂ was also created. A binary diffusion coefficient of 5×10^{-8} m²/sec was estimated based on the Takahashi correlation to account for high pressures, where a low pressure coefficient was determined using the Fuller estimation. Binary diffusion coefficients for polar compounds in supercritical CO₂ have been previously studied and are on the same order of magnitude as the binary diffusion coefficient estimated in this study. The system that was modeled is a cone-shaped system representing separate-phase CO₂ confined in a formation after injection. Boundary conditions consisted of a no-flux boundary at the top of the cone to account for the impermeable confining caprock, and a zero concentration boundary at the cone edge to simulate a worst case scenario for dissolution. The initial conditions considered a uniform concentration of one percent SO₂ everywhere in the cone. To numerically simulate the concentration profile throughout the cone, a time-split explicit difference method was applied. The diffusion modeling results show that contact between SO₂ and formation brine will be diffusion limited; after 3000 years approximately 75% of sulfur remains in the cone. In summary, while SO₂ is highly soluble in water, its slow diffusion through a supercritical CO₂ phase will likely inhibit its mass transfer.

H23D-0996

Pore-scale studies of multiphase flow and reaction involving CO₂ sequestration in geologic formations

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In geologic CO₂ sequestration, pore-scale interfacial phenomena ultimately govern the key processes of fluid mobility, chemical transport, adsorption, and reaction. However, spatial heterogeneity at the pore scale cannot be resolved at the continuum scale, where averaging occurs over length scales much larger than typical pore sizes. Natural porous media, such as sedimentary rocks and other geological media encountered in subsurface formations, are inherently

heterogeneous. This pore-scale heterogeneity can produce variabilities in flow, transport, and reaction processes that take place within a porous medium, and can result in spatial variations in fluid velocity, aqueous concentrations, and reaction rates. Consequently, the unresolved spatial heterogeneity at the pore scale may be important for reactive transport modeling at the larger scale. In addition, current continuum models of surface complexation reactions ignore a fundamental property of physical systems, namely conservation of charge. Therefore, to better understand multiphase flow and reaction involving CO₂ sequestration in geologic formations, it is necessary to quantitatively investigate the influence of the pore-scale heterogeneity on the emergent behavior at the field scale. We have applied the lattice Boltzmann method to simulating the injection of CO₂ saturated brine or supercritical CO₂ into geological formations at the pore scale. Multiple pore-scale processes, including advection, diffusion, homogeneous reactions among multiple aqueous species, heterogeneous reactions between the aqueous solution and minerals, ion exchange and surface complexation, as well as changes in solid and pore geometry are all taken into account. The rich pore scale information will provide a basis for upscaling to the continuum scale.

H23D-0997

Using a Natural Analogue to Investigate Chemical Reactions Associated with Carbon Dioxide Sequestration

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Capture and storage of carbon dioxide in deep underground geologic formations (geologic carbon sequestration) is currently the most advanced technology for reducing or mitigating anthropogenic carbon dioxide emissions. There are a number of scientific challenges associated with injection and storage of large amounts of CO₂ in geologic formations. Understanding the chemical reactions that can occur among reservoir rocks, aqueous fluids, and supercritical carbon dioxide □ other gasses is one of these challenges. Natural analogues to CO₂ sequestration are systems where carbon dioxide has been stored over geologic time scales. By studying these analogues we can determine important chemical reactions between the host rock and stored gases. The Moxa Arch is a structural feature located in the southern end of the greater Green River Basin, Wyoming. Carbon dioxide and methane were emplaced in Paleozoic rocks, including the 1000 feet thick Mississippian age Madison Limestone, of the Moxa Arch through natural processes. Concentrations of carbon dioxide in the emplaced gas in these formations vary in the region of the Moxa Arch from 70-95% and are as low as ~ 15% in gas producing areas outside of the Moxa Arch. Methane, hydrogen sulfide and helium comprise the balance of the gas compositions. Geochemical reaction path and reactive transport models based upon the mineralogy of 12 core samples collected from three wells completed in the Madison Limestone near the Moxa Arch will be presented. These models help identify potential geochemical reactions between reservoir minerals and stored gasses.

H23D-0998

Effects of density and mutual solubility of CO₂-brine system on CO₂ storage in geological formations

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The fluid properties of CO₂ injected into geological formations for subsurface storage are strongly affected by the specific formation conditions of pressure, temperature and salinity. The conditions affect fluid solubility and density, and their effects on subsurface CO₂ storage efficacy are investigated. Several common EOS and solubility models were investigated; their accuracy and applicability are briefly discussed. We evaluated the effects of gaseous/supercritical CO₂ phase density and H₂O solubility in CO₂. Results suggest that phase density estimated using different EOS or associated assumptions typically do not induce huge disparities in simulation results because of the low solubility of H₂O in gaseous/supercritical CO₂. However more experimental studies on the solubility of H₂O in CO₂ are needed, especially in the high pressure and temperature range. Simulation results also suggest that formations at higher temperatures are less efficient for CO₂ storage. We evaluated aqueous-CO₂ solution density at a broad range of pressure and temperature conditions using different EOSs. Results indicate that CO₂-dissolution in brines at high temperatures (>120° C) may reduce mass density to values lower than the original brine density, nullifying the dissolution trapping mechanism. The concept of equal density temperature is proposed here for the first time. In certain scenarios with temperature greater than the equal density temperature, CO₂ can escape the aqueous phase and be subject to buoyancy-driven migration (and potential escape from the formation) associated with separate phase CO₂. Simulation results are very sensitive to the density models selected. Predictions on the migration of CO₂ enriched brine with different models can yield opposite results.

H23D-0999

Long-Term Dissolution Rate of Carbon Dioxide in Saline Aquifers

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The aim of geological CO₂ storage is the permanent removal of the injected CO₂ from the atmosphere. The buoyancy of the injected supercritical CO₂ and the possibility of leakage along fractures faults and old wells may lead to leakage of CO₂ back into the atmosphere over time. The brine density increases with increasing CO₂ concentration, and therefore dissolved CO₂ is unlikely to leak back into the atmosphere. The rate at which CO₂ dissolves into the brine is a key constraint on the duration of possible leakage. Due to the low solubility of the CO₂ in the brine a large volume brine is necessary to dissolve a given amount of CO₂. Gravity driven flow induced by the increased density of CO₂ saturated brine is necessary to contact this large volume of brine and therefore determines the long-term dissolution rate. We present high-order direct numerical simulations of the convective motion in the brine in homogeneous, horizontal, laterally-unbounded aquifers. At early time, before the plumes of saturated brine have reached the bottom, the overall dissolution rate is essentially constant due to rapid convective overturn. At late time the saturated brine forms a miscible gravity current propagating outward from the CO₂ source. Simple models of constant density gravity currents predict a power-law decay of the overall dissolution rate. Direct numerical simulations show a similar power-law decay but slightly lower rates of decay. We attribute this to temporal variations of the average density of the gravity current.

H23D-1000

Physical Properties of Low-Rank Coal Samples from the Powder River Basin, Wyoming

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We characterize the mechanical properties of coal samples from the Powder River Basin (Wyoming, USA) by conducting laboratory experiments. We present results from laboratory measurements of adsorption, static and dynamic elastic moduli, and permeability as a function of effective stress, pore pressure, and gas species. Notably, we observe that CO₂ adsorption causes the static bulk modulus to decrease by a factor of two, while simultaneously causing the dynamic bulk modulus to increase by several percent. Permeability of both intact and powdered samples decreases by approximately an order of magnitude in the presence of CO₂, which is consistent with observations of adsorption-related swelling of the coal matrix. Interestingly, CO₂ appears to change the constitutive behavior of coal; helium saturated samples exhibit elastic behavior, while CO₂ saturated samples exhibit viscous, anelastic behavior, as evidenced by creep strain observations.

H23D-1001

Semi-analytical models of CO₂ Injection into Deep Saline Aquifers: evaluation of the area of review and leakage through abandoned wells

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This presentation will provide a conceptual preview of an Area of Review (AoR) tool being developed by EPA's Office of Research and Development that applies analytic and semi-analytical mathematical solutions to elucidate potential risks associated with geologic sequestration of carbon dioxide into deep saline subsurface formations. These solutions can be applied to commercial scale injections of supercritical CO₂ and enable the zone of influence and potential endangerment to be mapped, thereby helping to delineate the AoR. We anticipate implementing the semi-analytical solutions into an open source computer modeling framework. The major risks to be evaluated by the AoR tool include: induced subsurface pressures that may force native saline waters into an underground source of drinking water (USDW), and the potential transport of CO₂ away from the injection center and out of the receiving zone. Both of these phenomena are influenced by leakage and compromises of the sealing layers, such as presented by abandoned wells or other subsurface penetrations. The semi-analytical solutions will be tested against numerical solutions (TOUGH2/ECO2N) and field data associated with the Kimberlina test injection site near Bakersfield, CA. The AoR tool will be used to simulate a hypothetical commercial scale injection and to evaluate if existing or potential USDW aquifers may be adversely impacted by short-term or long-term geologic sequestration activities. The AoR tool will be useful for permit applicants and regulators evaluating potential exposure and risks associated with geosequestration under the Underground Injection Control (UIC) program. This project will benefit from partnerships with Lawrence Berkeley National Laboratory and Princeton University.

H23D-1002

Geomechanical Characterization and Reservoir Simulation of a CO₂-EOR and Sequestration Project in a Mature Oil Field, Teapot Dome, WY

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Mature oil and gas reservoirs are attractive targets for geological sequestration of CO₂ because of their potential storage capacities and the possible cost offsets from enhanced oil recovery (EOR). In this work we develop a 3D reservoir model and fluid flow simulation of the Tensleep Formation using geomechanical constraints in advance of a proposed CO₂-EOR injection experiment at Teapot Dome Oil Field, WY. The objective of this work is to model the migration of the injected CO₂ as well as to obtain limits on the rates and volumes of CO₂ that can be injected without compromising seal integrity. In the present work we combine our previous geomechanical analysis, geostatistical reservoir modeling and fluid flow simulations to investigate critical questions regarding the feasibility of a CO₂-EOR project in the Tensleep Fm. The analysis takes in consideration the initial trapping and sealing mechanisms of the reservoir, the consequences of past and present oil production on these mechanisms, and the potential effect of the CO₂ injection on the reservoir and the seal. Finally we also want to assess the long-term recovery of the injection site and what will happen in the system once the oil production stops. The CO₂-EOR injection pilot will consist of the injection of 1 MMcf of supercritical CO₂ for six weeks. The preliminary simulation results indicate that the injected CO₂ will rapidly rise to the top layers, above the main producing interval, and will accumulate in the fractures (almost none will get into the matrix). Design optimization will be needed to ensure adequate spatial distribution of the CO₂ and sufficient time for CO₂ miscibility.

H23D-1003

Geochemical Detection of Carbon Dioxide in Dilute Aquifers

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Carbon storage in deep saline reservoirs has the potential to lower the amount of CO₂ emitted to the atmosphere and to mitigate global warming. Leakage back to the atmosphere through abandoned wells and along faults would reduce the efficiency of carbon storage, possibly leading to health and ecological hazards at the ground surface, and possibly impacting water quality of near-surface dilute aquifers. In this study we use the reactive transport simulations performed by the Nonisothermal Unsaturated Flow and Transport (NUFT) code to test the hypothesis that perturbations in water chemistry associated with a CO₂ gas leak into dilute groundwater are important measures for the potential release of CO₂ to the atmosphere. We address the relationships between CO₂ flux, groundwater flow, and detection time and distance. The CO₂ flux ranges from 10³ to 2 × 10⁶ t/yr to assess chemical perturbations resulting from relatively small leaks that may compromise long-term storage, water quality, and surface ecology, and larger leaks characteristic of short-term well failure. The simulation results show the CO₂ leakage into a dilute groundwater creates a slightly acid plume that can be detected at some distance from the leak source due to groundwater flow and CO₂ buoyancy. Detection of CO₂ leaks in aquifers by changes in pH and carbonate chemistry is readily available and well understood. Reactive transport modeling is a critical component to the design and effective performance of measurement, monitoring, and verification plans for carbon storage. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

H23D-1004

Hydrogeologic Effects on Design and Results for Multiple Midwest Regional Carbon Sequestration Partnership Test Sites

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In planning and monitoring CO₂ injection experiments at Midwest Regional Carbon Sequestration Partnership sites, it was found that the hydrogeologic framework had a significant influence on the test design and results. The test sites are located along major regional geologic structures in the Midwestern United States: the Appalachian Basin, the Cincinnati Arch, and the Michigan Basin. Factors such as injection target thickness, permeability, formation pressures, and injection depths had a significant impact on the tests. In the Appalachian Basin, the nature of the injection targets resulted in a flexible injection plan capable of testing the injectivity of multiple targets. At the Cincinnati Arch site, approximately 90 m section of Mt. Simon Sandstone is present with promising hydraulic properties. As such, this test was focused on examining the mobility of the CO₂ within the storage formation, since a supply of CO₂ may not be available to test maximum injection rates. At the Michigan Basin site, a large supply of CO₂ was available. This test involved a longer injection period and more detailed examination of the CO₂ distribution in the deep rock formations. In addition, it allowed more analysis of the hydraulic pressure response in the reservoir. This work was done as part of the Midwest Regional Carbon Sequestration Partnership (MRCSP); DOE/NETL Cooperative Agreement No. DE-FC26-05NT42589.

H23D-1005

Geochemistry and Age Dating of Ancient and Modern CO₂ -rich Hydrothermal Systems as Natural Analogues for CO₂ storage: Examples from Australia and Eastern Mediterranean

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We investigated physico-chemical conditions during mineral authigenesis in CO₂-rich ancient and recent hydrothermal environments in Eastern Australia (Gunnedah and Bowen Basins) and Turkey, respectively. We performed Rb-Sr and U-series dating of clay-carbonate associations and travertine veins

respectively to evaluate the degassing and storage history of CO₂. Intense carbonate veining and coal seam cleat mineralisation in the Gunnedah Basin took place as a result of heat and CO₂ release associated with magmatism during the breakup of Gondwana in the Late Cretaceous. Widespread carbonate veining and cementation in the Bowen Basin occurred as products of basin-wide CO₂ rich meteoric hydrothermal fluids during the Late Triassic extension. CO₂ has largely been used for carbonate precipitation (calcite, siderite, ankerite and dawsonite) in eastern Australian basins; however, some high proportion of CO₂ has been stored in coal seams as adsorbed molecules on coal. Significant CO₂ degassing is common in geothermal fields in Turkey, as manifested by recent deposition of travertine pools and terraces as well as travertine vein networks in damage zones of active major fault systems. Trace element geochemistry indicates that transient ascent of CO₂-bearing fluids during seismic strain cycles without significant interaction with basement and host rocks resulted in rapid precipitation of the vein travertine near the surface. Such veins and associated breccias formed by hydraulic fracturing in response to overpressure of CO₂-rich fluids. Correlation of high-precision U-series ages with global/regional climate events indicates that late Quaternary climate variability may have controlled the geothermal water circulation that regulates CO₂ accumulation and the generation of CO₂ over-pressurised reservoirs and their behaviour during seismic events.

H23D-1006

Multi-channel Auto-dilution System for Remote Continuous Monitoring of High Soil-CO₂ Fluxes

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We describe a novel field instrument that takes input from up to 27 soil flux chambers and measures flux using the steady-state method. CO₂ concentrations are determined with an infrared gas analyzer (IRGA, 0- 3000 ppmv range) with corrections for temperature, barometric pressure, and moisture content. The concentrations are monitored during data collection and, if they exceed the range of the IRGA, a stepped dilution program is automatically implemented that allows up to 50-fold dilution of the incoming gas stream with N₂ supplied by boil-off from a large dewar. The upper concentration limit of the system with dilution is extended to at least 150,000 ppmv CO₂. The data are stored on a datalogger having a cellular modem connection that allows remote control of the system as well as transmittal of data. The system is designed to operate for six weeks with no on-site maintenance required. Longer periods are possible with modifications to allow on-site generation of N₂ from air. Example data from a recent CO₂ test injection at the Zero- Emission Research and Technology (ZERT) field site in Bozeman, MT are presented.

H23D-1007

Geologic CO₂ sequestration in saline aquifers accounting for dual permeability/porosity environments.

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The State of Minnesota, like many regions of the United States and beyond, has mandated significant reductions in CO₂ emissions by mid-century, and geologic CO₂ sequestration is recognized as one means by which to meet emissions goals. Unfortunately, the state, like many other regions, does not contain sedimentary basins that meet the currently established criteria for CO₂ sequestration in deep saline aquifers. That is, existing basins, though expansive, are shallower (e.g., the Mount Simon aquifer in Minnesota) or less permeable (e.g., the Midcontinental Rift System) than sedimentary units that are typically considered for sequestration. The field of karst hydrogeology recognizes the importance of multiple permeability/porosity systems in groundwater transport and storage. High permeability fracture networks permit rapid groundwater transport while the large, lower permeability matrix allows for significant storage. With this motivation, we develop a geologic CO₂ sequestration model, using TOUGH2 and TOUGHREACT, which accounts for the presence of multiple permeability/porosity structures. Capillary forces play an important role in these multiphase, multi-permeability and porosity systems. Our preliminary models investigate whether the Midcontinental Rift System could prove a viable candidate for geologic CO₂ sequestration, should suitable fracture networks (among other criteria) be located there.

H23D-1008

Coupled Effect of Wind and Rain on the Migration of CO₂ in the Vadose Zone under Wavy Topography

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The migration of CO₂ in the vadose zone depends on several factors that relate to the soil and fluid properties. Additionally, realistic boundary conditions are dynamic due to many factors that change on a diurnal or annual period. Therefore, the more we understand the effects of the time dependent boundary conditions, the better we can predict the whereabouts of a potential CO₂ leak. Along these lines, several studies have been performed by researchers to identify the physical mechanisms in the subsurface. The present study focuses on the effects of the interplay of the subsurface physics and the above surface phenomena using the TOUGH2 code. In a previous study by the authors, it was shown that winds of 5 m/s over a hill surface can significantly affect the underground paths followed by a CO₂ leak. The present study looks at the difference between a yearly-averaged continuous wind and a time varying wind pattern that has the same yearly average. Also, rainfall events have been accounted for, although several simplifying assumptions have been necessary to save on computational time. The results show that the wind patterns and the rainfall events can lead the CO₂ to show up in places where it is not expected, or can suppress at locations where it is expected. These findings point at the importance of continuous weather monitoring above the sequestration sites for

successful monitoring-mitigation practice.

H23D-1009

Geologic Storage at the Basin Scale: Region-Based Basin Modeling, Powder River Basin (PRB), NE Wyoming and SE Montana

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Carbon capture and storage from the over 2000 power plants is estimated at 3-5 GT/yr, which requires large-scale geologic storage of greenhouse gases in sedimentary basins. Unfortunately, determination of basin scale storage capacity is currently based on oversimplified geologic models that are difficult to validate. Simplification involves reducing the number of geologic parameters incorporated into the model, modeling with large grid cells, and treatment of subsurface reservoirs as homogeneous media. The latter problem reflects the focus of current models on fluid and/or fluid-rock interactions rather than fluid movement and migration pathways. For example, homogeneous models overemphasize fluid behavior, like the buoyancy of super-critical CO₂, and hence overestimate leakage rates. Fluid mixing and fluid-rock interactions cannot be assessed with models that only investigate these reactions at a human time scale. Preliminary and conservative estimates of the total pore volume for the PRB suggest 200 GT of supercritical CO₂ can be stored in this typical onshore sedimentary basin. The connected pore volume (CPV) however is not included in this estimate. Geological characterization of the CPV relates subsurface storage units to the most prolific reservoir classes (RCs). The CPV, number of well penetrations, supercritical storage area, and potential leakage pathways characterize each RC. Within each RC, a hierarchy of stratigraphic cycles is populated with stationary sedimentation regions that control rock property distributions by correlating environment of deposition (EOD) to CPV. The degree to which CPV varies between RCs depends on the geology and attendant heterogeneity retained in the fluid flow model. Region-based modeling of the PRB incorporates 28000 wells correlated across a 70,000 Km² area, 2 km thick on average. Within this basin, five of the most productive RCs were identified from production history and placed in a fourfold stratigraphic framework (second- through fourth-order cycles). Within the small-scale 4th-order sequences (30-150-m thick, 16 total), sedimentation regions, each corresponding to an EOD, are defined by thickness, lithology and core-calibrated well-log patterns. This talk illustrates the workflow by focusing on one of the 16 layers in the basin-scale model. Isopach maps from this sample layer conform to depositional patterns confirmed through definition of five core-calibrated, well-log defined sedimentation regions. Lithology distributions also conform to thickness trends in nearshore deltas, but not in offshore regions, where sand-rich and sheet-like, but thin-bedded sandstones are flanked by mud-rich intervals of equivalent thickness. These maps represent sedimentation patterns confined by basal erosional sequence boundary and basin-wide bentonite, yet containing up to seven high-frequency sequence boundaries. To illustrate over simplification problems in this same layer, a 14000 km² sample area is 600 km³ and using standard averaging methods, which are considered to be geologic in origin, the CPV is 16 km³. However, averaging increases connectivity with high CPV more uniformly distributed; significantly, the key mud belt region separating nearshore from offshore sandstones is not represented. Region-based modeling of this layer yields 13 km³ (110 Bbl). Furthermore, significant vertical leakage may exist from the 20000 well penetrations and faults and fractures along the western basin margin. This example illustrates the importance of accurately characterizing heterogeneity and distributing CPV using sedimentation regions.

H23D-1010

Evaluating the Effect of Gravity on CO₂ Plume Behavior in Deep Confined Saline Aquifers

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Previous modeling studies of CO₂ injection into deep saline aquifers have often neglected to account for gravity. Here, we assess the conditions under which this simplification may be valid. We considered the injection of CO₂ at a constant rate into a confined, homogeneous, isotropic saline aquifer via a single vertical well. We employed the TOUGH2/ECO2N software package to conduct numerical simulations of CO₂ injection. We conducted two sets of simulations, one in which gravity is included, and the other in which gravity is neglected. Other factors varied in the simulations include the relationship between relative permeability and brine saturation. Predicted pressures and vertically-averaged brine saturation profiles are used as bases of comparison between the two sets of simulations. The effect of gravity on the predicted pressures was found to be significant when relative permeability (k_r) varies non-linearly with brine saturation (S_w). However, if the relationship between k_r and S_w is linear or quasi-linear, gravity was found to have a small effect on model predictions of pressure in the formation. Results of vertically-integrated brine saturation profiles show slight increases in CO₂ plume extent when gravity is included, with a concomitant reduction in CO₂ storage efficiency. Results from this study suggest that gravity must be included in simulations of CO₂ sequestration if k_r varies non-linearly with S_w .

H23D-1011

Occurrence of Vaterite in CO₂ Well Bore Gas-rich Environments: a new Process of Carbonate Formation

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Vaterite scale (metastable hexagonal CaCO₃) occurs in well tubing from several southern California oilfields. The mineral forms in gas-rich well bore environments at depths ranging from 1.4 to 1.8 km. Some of the vaterite is extremely coarse-grained (up to 300µm crystals), much coarser than the micron-size crystals reported in the literature. This may be the reason for its long-term stability (years). Compared with calcite in adjacent parts of the tubing string, vaterite has extremely light carbon and oxygen isotopic composition that suggests it forms from a CO₂ rich and aqueous fluid depleted part of the system. The vaterite oxygen values are about 15 to 20 ‰ lighter than expected for equilibrium with the bulk pore fluid oxygen, suggesting that the oxygen within the vaterite has come from oxygen in the CO₂ gas rather than the pore water. Calcite scales in the same field typically have positive carbon values and relatively

positive oxygen values as would be expected from rapid crystallization of calcite due to CO₂ degassing from modified marine pore water. The vaterite appears to have crystallized by a fundamentally different process than the calcite scale and is interpreted to form as CO₂ gas interacts with small amounts of fluid on the tubing string. These occurrences suggest that vaterite may be much more common than presently known and that it may be expected to form in CO₂ rich well bore environments, such as from CO₂ sequestration.

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