Research Program

Energy Resources Program

Ernest L. Majer

510/486-6709
elmajer @lbl.gov

The Energy Resources Program (ER) within ESD is responsible for two major program areas: Oil and Gas Exploration and Development, and Geothermal Energy Development.

OIL AND GAS EXPLORATION AND DEVELOPMENT

Multidisciplinary research is being conducted in reservoir characterization and monitoring, optimization of reservoir performance, and environmental protection. Using basic research studies as a source of innovative concepts, ER researchers seek to transform these concepts into tangible products of use to industry within a time frame consistent with today's rapid growth in technology. Reservoir characterization and monitoring involve development of new seismic and electromagnetic techniques focused at the interwell scale. Field acquisition, laboratory measurements, and numerical simulation play important roles in the development activities. Optimization of reservoir performance is focused on simulation-based methods for enhancing reservoir management strategies. Emphasis is placed on the integration of geophysical data, production data, and reservoir simulation. The next major step in research will focus on methods to optimize performance through integration of monitored geophysical data, production data, and reservoir simulation.

International and national concern about the variable climatic effects of greenhouse gases produced by burning of fossil fuels is increasing, while it is also recognized that these fuels will remain a significant energy source for the indefinite future. In response to these concerns, ER has initiated research focused on development of technologies that will minimize the impact of fossil-fuel usage on the environment. Methane hydrates constitute a huge potential fuel source, with lower carbon emissions than coal or oil. ER researchers are developing and evaluating possible methods for producing gas from such deposits.

Geophysical data acquisition and inversion methods developed in the ER program are also being applied in a new ESD program, the Geologic Carbon Sequestration Program, described elsewhere in this volume.

Principal research activities include:

- Development of 3-D electromagnetic processing methods for oil and gas recovery
- Development of microwell seismic technology, including instrumentation, acquisition, and processing
- Applications of seismic methods for characterization of fractured reservoirs
- Development of joint electrical-seismic inversion methods for oil and gas exploration and monitoring
- Use of passive (microearthquake) and active seismic imaging for understanding and controlling enhanced geothermal systems (EGS)
- Laboratory measurement of the seismic properties of poorly consolidated sands
- Evaluation of seismic stimulations methods and their application to different classes of oil reservoirs
- Improved inversion methods for reservoir characterization, with a focus on combining production and geophysical data
- Application of x-ray computed tomography (CT) and nuclear magnetic resonance (NMR) imaging to study multiphase flow processes
- Pore-to-laboratory-scale study of physical properties and processes, with a focus on controlling phase mobility, predicting multiphase flow properties, and increasing drilling efficiency
- Development of new methods to mitigate environmental effects of petroleum refining and use
- Enhancement of refining processes using biological technologies
- Numerical simulation of subsurface methane hydrate systems

Since 1994, the major part of the Oil and Gas Exploration and Development program has been funded through the Natural Gas and Oil Technology Partnership Program. Begun in 1989, the partnership was expanded in 1994 and again in 1995 to include all nine Department of Energy multiprogram laboratories, and has grown over the years to become an important part of the DOE Oil and Gas Technologies program for the national laboratories. Partnership goals are to develop and transfer to the domestic oil industry the new technologies needed to produce more oil and gas from the nation's aging, mature domestic oil fields, while safeguarding the environment.

Partnership technology areas are:

- Oil and gas recovery technology
- Diagnostics and imaging technology
- Drilling, completion and stimulation
- Environmental technologies
- Downstream technologies

GEOTHERMAL ENERGY DEVELOPMENT

There are two main objectives of ER's geothermal energy development program: (1) to reduce uncertainties associated with finding, characterizing, and evaluating geothermal resources, and (2) to develop and understand the enhancement of current geothermal systems to significantly increase production, i.e., Enhanced Geothermal System (EGS). The ultimate purpose is to lower the cost of geothermal energy for electrical generation or direct uses (e.g., agricultural and industrial applications, aquaculture, balneology). The program encompasses theoretical, laboratory, and field studies, with an emphasis on multidisciplinary approaches to solving the problems at hand. Existing tools and methodologies are upgraded, and new techniques and instrumentation are developed for use in the areas of geology, geophysics, geochemistry, and reservoir engineering. Cooperative work with industry, universities, and government agencies draws from Berkeley Lab's 25 years of experience in the area of geothermal research and development.

In recent years, DOE's geothermal program has become more industry-driven, and the Berkeley Lab effort has been directed toward technology transfer and furthering our understanding of the nature and dynamics of geothermal resources under production.

At present, the main research activities of the program include:

- Geothermal Reservoir Dynamics: development and enhancement of computer codes for modeling heat and mass transfer in porous and fractured rocks, with specific projects such as modeling the migration of phase-partitioning tracers in boiling geothermal systems; modeling of mineral dissolution and precipitation during natural evolution, production, and injection operations; and geophysical-signature prediction of reservoir conditions and processes
- ***Isotope Geochemistry:*** identification of past and present heat and fluid sources, development of natural tracers for monitoring fluids re-injected into geothermal reservoirs, better understanding of the transition from magmatic to geothermal production fluids, and enhancement of reservoir-simulation methods and models by providing isotopic and chemical constraints on fluid source, mixing, and flow paths
- ***Geochemical Baseline Studies:*** documentation of geothermal-fluid behavior under commercial production and injection operations (e.g., field case studies), with specific emphasis on The Geysers field in Northern California
- ***Electromagnetic Methods for Geothermal Exploration:*** development of efficient numerical codes for mapping high-permeability zones, using single-hole electromagnetic data and surface magnetotelluric data.

Future research will concentrate on the development of innovative techniques for geothermal exploration and assisting in a reassessment of geothermal power potential in the U.S. The emphasis will be on expanding existing fields, prolonging their productive life, and finding new “blind” geothermal systems, i.e., those that do not have any surface manifestations, such as hot springs or fumaroles, that suggest the presence of deeper hydrothermal systems.

FUNDING

Within ER, The Oil and Gas Exploration and Development program receives support from the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Energy Technology Laboratory, the National Petroleum Technology Office, and the Natural Gas and Oil Technology Partnership, under U.S Department of Energy Contract No. DE-AC02-05CH11231. Support is also provided from industry and other sources through the Berkeley Lab Work for Others program. Industrial collaboration is an important component of DOE Fossil Energy projects.

The Geothermal Energy Development program receives support from the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Power Technologies, Office of Wind and Geothermal Technologies, of the U.S. Department of Energy.

Uncertainty Analysis Using Stochastic Rock-Physics Models and Markov Chain Monte Carlo Methods

Jinsong Chen
Contact: 510/486-6842, jchen@lbl.gov

RESEARCH OBJECTIVES

Rock-physics models are needed for reservoir parameter estimation using marine seismic amplitude-versus-angles (AVA) and controlled source electromagnetic (CSEM) data. They are typically derived in practice from suitable nearby borehole logs. However, since relationships between reservoir parameters and geophysical attributes are often nonlinear and non-unique, the derived rock-physics models are inevitably subject to uncertainty. Traditional methods for analyzing uncertainty in rock-physics models are performed by varying a small subset of the rock-physics parameters while keeping others unchanged. In essence, those methods explore the marginal effects of the parameters on reservoir parameter estimation and are valid only when the parameters being investigated are independent of those kept unchanged. Since rock-physics parameters often depend on each other, the utility of such approaches is limited. Additionally, the above methods only analyze the effects of uncertainty in the estimated rock-physics parameters without considering uncertainty in the rock-physics model outputs. The goal of this study is to develop a flexible, robust, and integrated approach for analysis of uncertainty in rock-physics models.

APPROACH

A Bayesian framework and stochastic rock-physics models are used to combine marine seismic AVA and CSEM data for reservoir parameter estimation. The outputs of rock-physics models are considered as random functions of reservoir parameters, and the parameters of rock-physics models are considered as random variables. Markov chain Monte Carlo (MCMC) methods are used to draw many samples from the joint posterior probability density functions. The obtained samples from MCMC methods are used to estimate reservoir parameters as well as uncertainty information in the estimation.

ACCOMPLISHMENTS

A Bayesian model based on a layered reservoir model has been developed to jointly invert seismic AVA and CSEM data, and the model has been applied to a synthetic reservoir with high gas saturation. Figure 1 shows the estimated posterior probability density functions (pdfs) of water saturation, shale content, and porosity using stochastic rock-physics models at various noise levels.

SIGNIFICANCE OF FINDINGS

Uncertainty in both the outputs and the parameters of rock-physics models could have significant effects on the estimates of reservoir parameters in joint inversion of marine seismic AVA and CSEM data. The effects of uncertainty in rock-physics model parameters are generally less significant than those of uncertainty in rock-physics model outputs. The study also shows that an integrated approach is important for uncertainty analysis in reservoir parameter estimation, and that the developed Bayesian model in this study provides a consistent and effective method for uncertainty analysis.

RELATED PUBLICATIONS

Chen, J., and T. Dickens, Effects of uncertainty in rock-physics models on reservoir parameter estimation using marine seismic AVA and CSEM data. SEG Expanded Abstracts (in press), 2007.

ACKNOWLEDGMENTS

This work was supported by the ExxonMobil Upstream Research Company and by the U.S. DOE Assistant Secretary for Fossil Energy, Office of Oil and Natural Gas, under Contract No. DE-AC02-05CH11231.

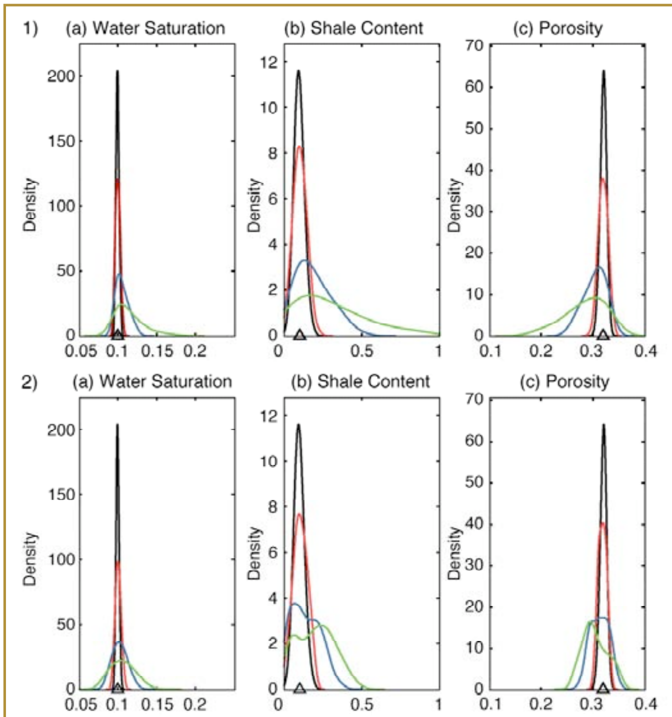

Figure 1. Estimated posterior pdfs of water saturation, shale content, and porosity, where the black, red, blue, and green curves are the estimated posterior pdfs when the outputs of rock-physics models are exact, and have errors equivalent to the coefficients of variations of 1%, 3%, and 5%, respectively.

Measurements and Observations of Water, Gas, and Heat Flow in Hydrate-Bearing Sands

T.J. Kneafsey, L. Tomutsa, Y. Seol, R.C. Trautz, G.J. Moridis, and T.A. Ghezzehei
Contact: T.J. Kneafsey, 510/486-4414, tjkneafsey@lbl.gov

RESEARCH OBJECTIVES

Natural deposits of gas hydrates (i.e., methane molecules enclosed within a solid water lattice at elevated pressure and cool temperatures), thought to contain vast amounts of recoverable natural gas, are present in the subsurface in permafrost areas and at or below the ocean floor. To capture this gas, the solid hydrate must be destabilized (by lowering pressure, increasing temperature, or adding an inhibitor), causing it to dissociate into methane gas and water, and extracted from the earth using a production well. Modeling of these processes is needed to estimate the economics of gas production, but the modeling must be based on correct physics. Our objectives are to perform experiments that provide visualizations and quantifications of water and gas flow through hydrate-bearing samples to improve conceptual and numerical models of gas production from hydrates.

APPROACH

We form methane hydrate in samples contained in x-ray transparent vessels, while controlling the water content, pressure, and temperature. We then quantify water and gas flow through the resulting system using pressure, temperature, and flow measurements, and use x-ray computed tomography to visualize changes and water flow.

ACCOMPLISHMENTS

We have performed a number of flow tests for sands having different characteristics at several hydrate saturations and have estimated relative permeabilities based on our measurements. We have performed and monitored dissociation tests by depressurization and thermal stimulation, and have performed flow tests through samples that had heterogeneities in hydrate saturation and prescribed heterogeneities. Also, we are developing a procedure for measuring the characteristic curve of hydrate-bearing sand.

SIGNIFICANCE OF FINDINGS

Our measurements are being incorporated into numerical simulations of gas production from hydrates, adding confidence to the modeling. Visualizations have taught us about the roles of heat and mass transfer in the technique used to make hydrate samples and their influence on resulting measurements. Additionally, the impact of parameter changes caused by the presence of hydrate in a porous medium can be seen directly in our visualizations, allowing better understanding and modeling of the processes.

RELATED PUBLICATIONS

Kneafsey, T.J., L. Tomutsa, G.J. Moridis, Y. Seol, B.M. Freifeld, C.E. Taylor, and A. Gupta, Methane hydrate formation and dissociation in a core-scale partially saturated sand sample. LBNL-59087. Journal of Petroleum Science and Engineering, 56, 108–126. 2007.

Gupta, A., T.J. Kneafsey, G.J. Moridis, Y. Seol, M.B. Kowalsky, and E.D. Sloan Jr., Methane hydrate thermal conductivity in a large heterogeneous porous sample. LBNL-59088. J. Phys. Chem. B; ASAP Web Release Date: August 2, 2006; DOI: 10.1021/jp0619639, 2006.

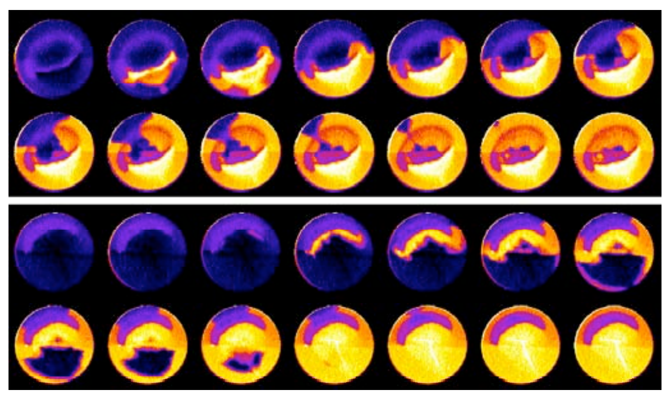

Figure 1. Sequence of CT scans showing water flow through a sample having coarse sand on the top and fine sand on the bottom. The brighter regions in the first scan show higher saturations of hydrate. In later scans, the brighter regions show the initial hydrate saturation with increasing saturations of water.

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Energy Technology Laboratory, under the U.S. DOE, Contract No. DE-AC02-05CH11231.

Frequency-Dependent Seismic Response of Underground Reservoirs

Valeri Korneev
Contact: 510/486-7214, vakorneev@lbl.gov

RESEARCH OBJECTIVES

The main objective of this project is the development and application of the new advanced technology of hydrocarbon reservoir imaging, supported by a frequency-dependent reflectivity model. Reaching this objective requires an understanding of how seismic waves interact with fluid-saturated rocks.

APPROACH

This project involves development of theory and processing algorithms, laboratory experiments, and verification of results, using field data provided by industrial partners. For theory development with respect to frequency-dependent reflections, a computer code was developed that models (in two and three layers) the reflections caused by P- wave incidence. Fluid flow effects are modeled by application of Biot's porous rock theory and generalized to the double porosity theory.

ACCOMPLISHMENTS

Modeling results were compared with observations in laboratory data and field experiments. It was determined that the modeled effects are too small to explain the data. On the other hand, analyses of permeability values consistently demonstrate that measurements at field scale (hundreds of meters to kilometers) show an increase of several orders of magnitude compared to values obtained at laboratory scales. This behavior finds an explanation in the self-similar distribution of fractures in rock and implies that fluid-related effects in fractures have to be considered for realistical models of rock. An analytical solution has been obtained with respect to the phase velocity of a Stoneley guided wave for an infinite fracture filled with viscous fluid. The solution suggests differentiating between oil and water fluids at low frequencies. It also suggests that at seismic frequencies, resonances are possible at reservoir scales (Figure 1). Resonant frequencies depend on fluid parameters and fracture dimensions.

SIGNIFICANCE OF FINDINGS

The Stoneley wave (slow wave) effect, which is not a part of any existing poroelastic theory, needs to be taken into account at field scales. For proper extraction of permeability information, the scattering of seismic waves caused by fracturing within heterogeneous systems needs to be included in reservoir models. Resonant characteristics of fluid-filled fractures are likely to cause nonlinear effects when seismic waves interact with reservoirs.

RELATED PUBLICATIONS

Korneev, V.A, Slow waves in fractures filled with viscous fluid. Geophysics (in press), 2007.

Silin, D.B., V.A. Korneev, G.M. Goloshubin, and T.W. Patzek, Low-frequency asymptotic analysis of seismic reflection from a fluid-saturated medium. LBNL-54955. Transport in Porous Media, 62, 283–305, 2006.

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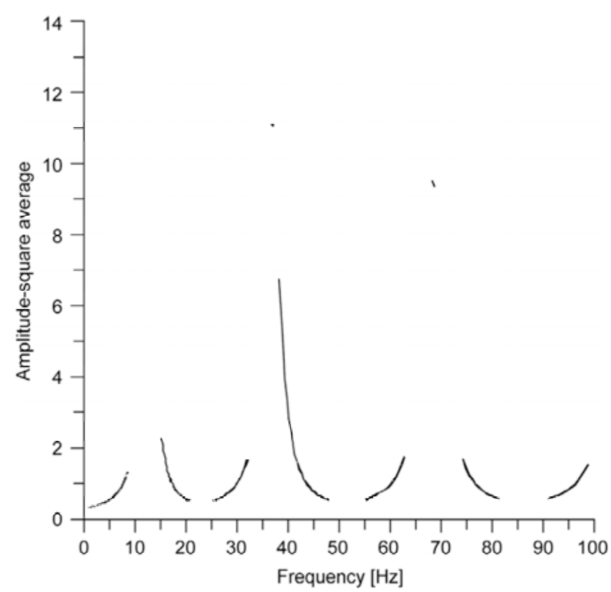


Figure 1. Resonant energy density for 4 m long, 1 mm thick water-filled fracture

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Energy Technology Laboratory, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

Comparison of Kinetic and Equilibrium Reaction Models in Simulating Gas Hydrate Behavior in Porous Media

Michael B. Kowalsky and George J. Moridis
Contact: Michael B. Kowalsky, 510/486-7314, mbkowalsky@lbl.gov

RESEARCH OBJECTIVES

Production of natural gas from hydrate accumulations may proceed by inducing dissociation, an endothermic reaction which results in the production of gas and water, using one or a combination of the following mechanisms: (1) depressurization; (2) thermal stimulation; and (3) inhibitors (such as salts and alcohols). The objective in this work is to evaluate, through numerical simulation, the importance of employing kinetic versus equilibrium reaction models to describe the hydrate dissociation reaction and thus predict the response of (methane) hydrate-bearing systems to external stimuli, such as changes in pressure and temperature.

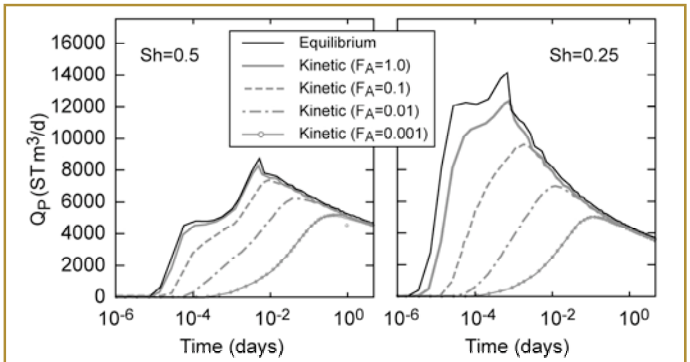


Figure 1. Effect of reaction area on early-time CH_4 production in Class 3 hydrate accumulation undergoing depressurization. The responses for two different initial hydrate saturations are shown in (a) and (b), respectively, for the equilibrium reaction model and for the kinetic reaction model with a decreasing amount of surface area available in the hydrate dissociation reaction.

APPROACH

Our numerical studies are conducted using TOUGH+HYDRATE (formerly TOUGH-Fx/HYDRATE), which models the nonisothermal hydration reaction, phase behavior, and flow of fluids and heat under conditions typical of natural hydrate deposits in complex formations. It includes both equilibrium and kinetic models of hydrate formation and dissociation, and can handle any combination of the possible hydrate dissociation mechanisms mentioned above. The code accounts for heat and up to four mass components (i.e., water, CH_4, hydrate, and water-soluble inhibitors such as salts or alcohols) that are partitioned among four possible phases (gas, liquid, ice, or hydrate phases, which may exist individually or in any of 12 possible combinations).

ACCOMPLISHMENTS

In this study, we (1) analyze and compare the responses simulated using both reaction models for natural gas production from hydrates in various settings, and for the case of depressurization in a hydrate-bearing core during extraction; and (2) examine the sensitivity to factors such as initial hydrate saturation, hydrate reaction surface area, and numerical discretization. An example of production in a Class 3 hydrate accumulation undergoing depressurization is shown in Figure 1 for different initial values of hydrate saturation, and for a decreasing amount of surface area available in the dissociation reaction for both kinetic and equilibrium reaction models. We find that for large-scale systems undergoing thermal stimulation and depressurization, the long-term predictions for both reaction models are remarkably similar, though some differences are observed at early times. However, for modeling short-term processes, such as the rapid recovery of a hydrate-bearing core, kinetic limitations can be important, and neglecting them may lead to significant underprediction of recoverable hydrate.

SIGNIFICANCE OF FINDINGS

Assuming validity of the most accurate kinetic reaction model that is currently available, use of the equilibrium reaction model appears to be justified in many cases and preferred for simulating the behavior of gas hydrates, given that the computational demands for the kinetic reaction model far exceed those for the equilibrium reaction model.

RELATED PUBLICATIONS

Kowalsky, M. B., and G. J. Moridis, Comparison of kinetic and equilibrium reactions in simulating the behavior of gas hydrates. LBNL-63357. Energy Conversion and Management, 48, 1850–1863, doi:10.1016/j.enconman.2007.01.017, 2007.

Moridis, G. J., and M. B. Kowalsky, Response of oceanic hydrate-bearing sediments to thermal stresses. LBNL-60150. SPE Journal, 12(2), 253–268, doi:10.2118/111572-PA, 2007.

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Energy Technology Laboratory (NETL), under the U.S. Department of Energy, Contract No. DE-AC02-05CH11231.

Gas Production from Oceanic Class 2 Hydrate Accumulations

George J. Moridis and Matthew T. Reagan
Contact: George J. Moridis, 510/486-4746, gjmoridis@lbl.gov:

RESEARCH OBJECTIVES

The objective of this study is to evaluate the production potential of marine Class 2 methane hydrate accumulations by means of numerical simulation, and to develop appropriate strategies for gas production.

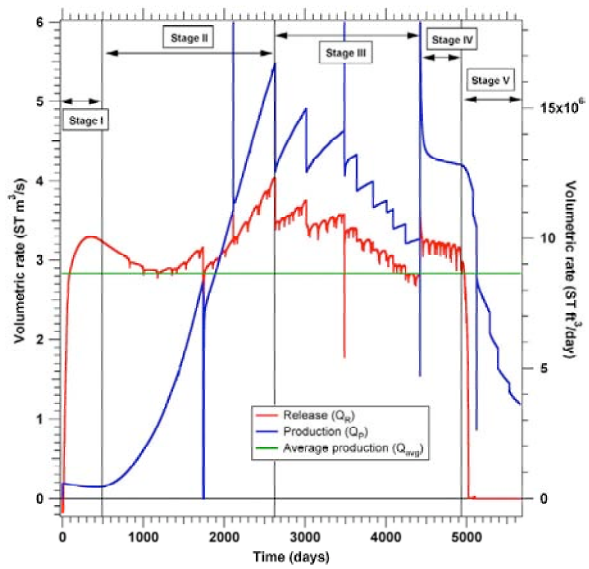


Figure 1. Rates of (a) hydrate-originating CH_4 release in the reservoir (red) and (b) CH_4 production at the well (blue) during production from a Class 2 oceanic hydrate deposit using a novel wellbore design. The stages of production and the average production rate (Q_{avg}, in green) over the simulation period are also shown.

APPROACH

Class 2 hydrate deposits comprise two zones: a hydrate-bearing layer (zone) overlying a zone of mobile water. The simulated system is based in the "Tigershark" area, located in the Alaminos Canyon Block 818 of the Gulf of Mexico. Data from an exploration well in 2,750 m of water indicated the presence of a thick, sandy hydrate layer, and preliminary calculations suggested that the base of the gas hydrate stability zone occurs at or slightly below the base of the hydrate-bearing layer. A zone of mobile water underneath the hydrate zone was assumed.

Depressurization-induced dissociation, enhanced by the near-incompressibility of water, was selected as the most promising gas production strategy because of its simplicity and its technical and economic effectiveness. The code used in this study is TOUGH+HYDRATE, developed by Berkeley Lab staff with support from DOE-NETL. The code can describe any combination of hydrate dissociation mechanisms and can account for four mass components partitioned among four possible phases (gas, liquid, ice, hydrate), with a total of 15 unique phase states. Well configurations and the rates of mass production evolved during the course of production in response to system behavior.

ACCOMPLISHMENTS

For all strategies, production at the well is characterized by distinct cycles, with the end of each cycle marked by cavitation (rapid pressure drop at the well), caused by either secondary hydrate blockages near the well or by the increasing participation of lower-density gas. This is remedied by warm seawater injection to destroy any barrier (if it exists) and a reduction in the total mass production rate. Gas production continuously increases during each cycle, with a corresponding reduction in the water production.

Three productions strategies were evaluated. The first involved a well completed through the hydrate layer and into the zone of mobile water, with the entire interval perforated for production. This method resulted in the highest gas production rates, but was subject to flow blockages by secondary hydrate and ice formation. The second involved a well with a production interval below the base of the hydrate. This strategy was also subject to secondary hydrate formation, and production rates were limited because of a well design that did not access gas accumulated at the top of the formation. The third method combined features of the first two cases, using a combination of electrical heating to initiate production, warm seawater circulation during continued production, and finally the application of a novel well design that combines warm seawater injection with a long production interval to access the remaining hydrate and avoid blockages and cavitation (see the related publication). Production with this process proceeds in five stages (see figure): initiation, increasing production, depletion, enhanced recovery (through the novel well design), and exhaustion. This approach is clearly superior to conventional production techniques for production under the Tigershark reference conditions.

SIGNIFICANCE OF FINDINGS

This study demonstrates that gas can be produced at high rates over long periods from Class 2 oceanic hydrate deposits, using conventional technology, and can be further enhanced through the use of novel production methods.

RELATED PUBLICATION

Moridis, G.J., and M.T. Reagan, Gas production from Oceanic Class 2 hydrate accumulations. LBNL-62757. OTC 18866, Offshore Technology Conference, Houston, Texas, USA, April 30–May 3, 2007.

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Energy Technology Laboratory (NETL), under the U.S. Department of Energy, Contract No. DE-AC02-05CH11231.

BERKELEY LAB

A Strategy for Gas Production from Oceanic Class 3 Hydrate Deposits

George J. Moridis and Matthew T. Reagan
Contact: George J. Moridis, 510/486-4746, gjmoridis@lbl.gov

RESEARCH OBJECTIVES

The main objective of this study is to evaluate the natural gas production potential of marine Class 3 accumulations and to determine the factors and conditions affecting it. Class 3 accumulations are composed of a single zone bounded by confining strata, the hydrate-bearing interval, with no underlying zone of mobile fluids. Previous studies have dismissed such formations as targets for gas production, but an appropriate choice of production techniques allows us to re-examine their viability.

APPROACH

The code used in this study is TOUGH+HYDRATE, developed by Berkeley Lab staff with support from DOE-NETL. The model system is based on the "Tigershark" area of the Gulf of Mexico, where an exploration well in 2,750 m of water has indicated the presence of an 18.25 m thick sandy hydrate layer. Previous studies have used thermal stimulation, via electrical heating or warm water injection, to induce dissociation in Class 3 deposits, but with disappointing results. A new strategy uses constant-pressure production to overcome the very low initial effective permeability of the deposit, the uncertainty over the evolution of effective permeability over time, and the lack of a mobile water zone.

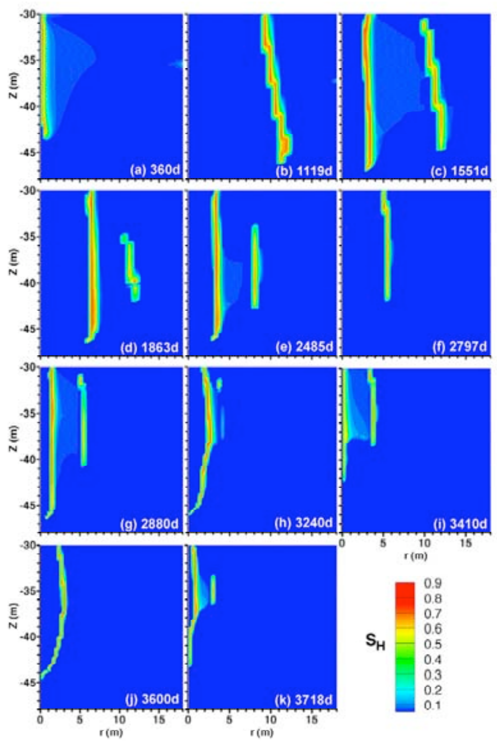


Figure 1. Evolution of spatial distribution of hydrate saturation, S_H, during gas production from a Class 3 oceanic hydrate deposit, showing the traveling barriers of secondary hydrate.

ACCOMPLISHMENTS

The simulated gas production occurs in a cyclical pattern. Each cycle consists of a long stage of increasing gas production, followed by sharp production decline, followed by a recovery. This pattern is repeated until the cessation of production. Peak production regularly exceeds 5 ST m^3/s of CH_4 (15 MMSCFD), and the average gas production during the 6,000-day simulation period is 2.61 ST m^3/s (8.10 MMSCFD), with a total of 1.37×10^9 ST m^3 (4.84×10^{10} ST ft^3) produced, all of which originated from the hydrate. Recovery from each production decline occurs naturally, without heating or fluid injection. Unlike conventional natural gas reservoirs, water production decreases continuously with time.

The figure shows the evolution of the spatial distribution of hydrate saturation over time and provides an explanation for the cyclical pattern. The precipitous drops in production are caused by the evolution of *traveling dual barriers* of secondary hydrate around the well. As gas flows to the well, a cylindrical sheath of secondary hydrate forms next to the wellbore (Panel a). Because the inner radius of the barrier is exposed to intense depressurization, it dissociates, but additional hydrate forms on its outer radius, resulting in the appearance of a traveling barrier (b). A second barrier forms (c) and moves outward through the previously described process (d). The outer barrier is now shielded from steep pressure gradients and begins to dissociate through contact with the warmer fluids behind it (d through f). As time advances, new barriers appear and disappear (g though k). Each of the temporary drops in production occurs when a new inner secondary hydrate barrier is formed.

SIGNIFICANCE OF FINDINGS

This study demonstrates that large volumes of hydrate-originating natural gas can be produced at high rates over long periods from Class 3 oceanic hydrate deposits. With straightforward depressurization-induced dissociation and gas production at constant pressure, secondary hydrate blockages can be "self-healing" without additional heating or fluid injection. Hence, these deposits may be viable candidates for future exploitation and development.

RELATED PUBLICATIONS

Moridis, G.J. and M.T. Reagan, Strategies for gas production from Oceanic Class 3 hydrate accumulations. LBNL-62758. OTC 18866, 2007 Offshore Technology Conference, Houston, Texas, U.S.A., April 30–May 3, 2007.

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Energy Technology Laboratory (NETL), under the U.S. Department of Energy, Contract No. DE-AC02-05CH11231.

SHEAR-STRESS MONITORING OF A FRACTURE USING SEISMIC WAVES

Seiji Nakagawa
Contact: 510/486-7894, snakagawa@lbl.gov

RESEARCH OBJECTIVES

Transmission and reflection of seismic waves can be used to monitor changes in in situ geological stresses on a fracture. For example, an increase in normal stress (compression) can be measured from an amplitude increase and a phase delay of the transmitted wave. For monitoring shear stress, a less well-known effect can be used: when an incoming wave is normally incident on a fracture, the amplitude and particle motions of the transmitted and reflected waves with mode conversions (compressional [P] waves to shear [S] waves, and vice versa) change as a function of the magnitude and direction of shearing on the fracture (Nakagawa et al., 2000). However, several questions need to be answered before this effect can be used as a practical tool for stress monitoring. These include: What is the quantitative relationship between the magnitude of shear stress and the conversions of the waves? What is the impact of normal stress acting together with the shear stress? What is the effect of nonlinear elastic and inelastic (frictional slips and failure) deformations of the fracture? And is there a precursor in the converted waves before a slip along a fracture?

APPROACH

A series of laboratory seismic measurements was conducted on both a natural rock sample and synthetic (aluminum and steel) samples. (Sample diameter=10.16 cm, height=5.08 cm.) A single, through-going fracture was induced by tension in the rock sample. The metal samples contained a pair of sine-wave fracture surfaces (period=1.25 mm, peak-trough amplitude=250 mm). The fractures were quasi-statically sheared under a constant normal stress. The stresses were controlled using a small, bi-axial loading machine capable of applying up to 5.5 MPa of both normal and shear stresses on the fracture surface. For each combination of normal and shear stresses, converted S-waves and unconverted P-waves transmitted through a fracture from an incident P-wave (central frequency=500 kHz) were measured.

ACCOMPLISHMENTSC

From an experiment using a natural rock sample (granite), the amplitude of converted S-waves was found to increase nonlinearly with shear stress (Figure 1). Also, the magnitude of normal stress did not have a strong effect on the amplitude. At low normal stresses, P-waves also showed small increases in amplitude. However, at high stresses, the rate of increase reduced, and the amplitude decreased for large shear stresses. A metal sample exhibited a somewhat different behavior, because stable, large slips occurred during shearing, owing to the smooth (although not flat) surface of the machined fracture. Further, a cyclic shearing of the fracture resulted in hysteretic changes in the amplitude of converted waves, which exhibited larger changes when the fracture surface was sliding.

SIGNIFICANCE OF FINDINGS

The experiments indicate that the relationship between the shear stress on a fracture and the amplitude of shear-induced, mode-converted waves depends strongly on the mechanical properties of the fracture.

RELATED PUBLICATION

Nakagawa, S., K. T. Nihei, and L. R. Myer, Shear-induced conversion of seismic waves across single fractures. Int. J. Rock Mech. Min. Sci. & Geomech. Abstr., 37(1–2), 203–218, 2000.

ACKNOWLEDGMENTS

This work was supported by the Director, Office of Science, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences, and Biosciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

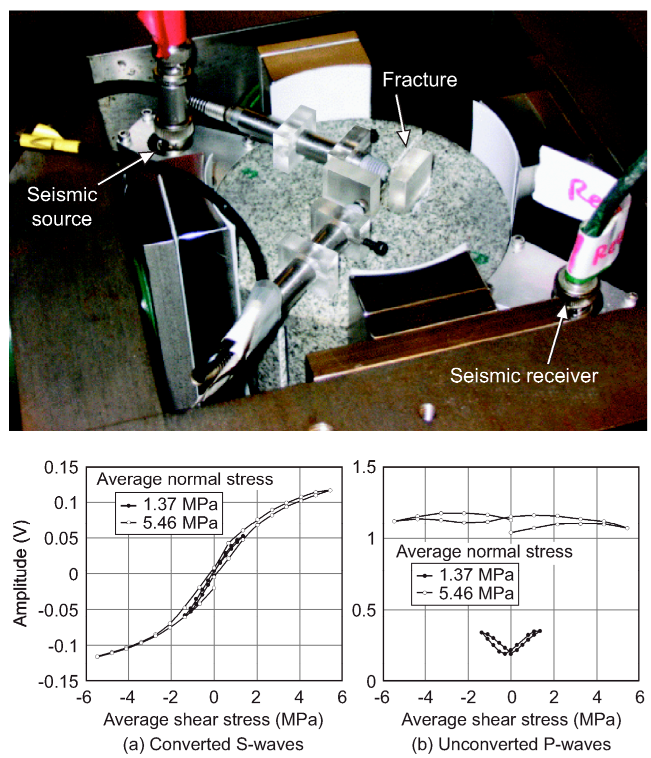


Figure 1. Laboratory seismic wave transmission experiment during bi-axial shear loading. Both normal and shear stresses were applied to a granite cylinder containing a single, through-going tensile fracture. Static deformations normal and parallel to the fracture were also monitored during the experiment.

Seismic Boundary Conditions for a Fluid-Filled Fracture

Seiji Nakagawa and Michael A. Schoenberg
Contact: Seiji Nakagawa, 510/486-7894, snakagawa@lbl.gov

RESEARCH OBJECTIVES

Rock is often permeated by compliant plane discontinuities (such as fractures and faults) that, depending on their permeability relative to the background, serve as either conduits or barriers to subsurface fluid. The fluid permeability of a fracture is often a key parameter, yet the quantitative relationship between permeability and its effect on seismic wave scattering is not fully understood. Strong scattering of seismic waves by a fracture is usually related to large permeability, because an open fracture with partial surface contacts has increased mechanical compliance (deformability). However, if a fluid-containing fracture is filled with debris, or a single fracture consists of a large number of microcracks, complex interactions between rock and pore fluid in the fracture result. In this research, a simple mathematical model that captures the essential nature of solid-fluid interaction within a fracture is developed, to predict the effect of hydraulic permeability and other fracture properties on seismic wave scattering.

APPROACH

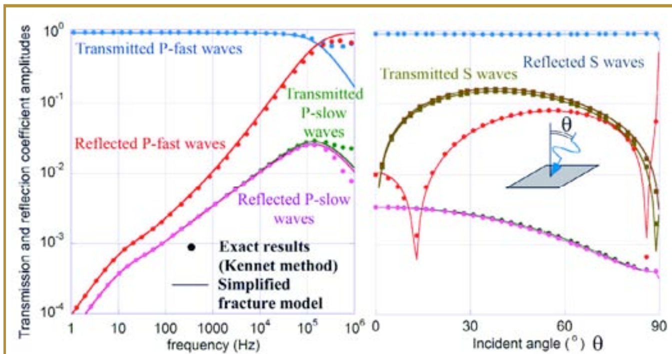
We envision a fracture as a thin, flat, homogeneous poroelastic layer surrounded by homogeneous halfspaces. Starting from the governing equations of poroelastic wave propagation, the spatial gradient in the fracture-normal direction of the displacement (or particle velocity), stress, and fluid pressure induced by seismic waves are expressed using other wave field variables. When accumulated (integrated) over the thickness of the fracture, this gradient produces a finite jump (discontinuity) in the field quantities. If this jump can be expressed using only the wave-induced displacement and stress (and pressure) on the surfaces of a fracture, we obtain a seismic boundary condition.

ACCOMPLISHMENTS

The derivation of the jump conditions resulted in a pair of compact, coupled matrix equations for fast and slow compressional (P) waves and a shear (S) wave. For a fracture thickness much smaller than the wavelength of propagating seismic waves, these jump conditions correctly capture frequency-dependent changes in the poroelastic behavior of a fracture. A further simplification of the equations resulted in a set of boundary conditions that are a function of five fracture characteristic parameters defined for a fluid-filled fracture (for a dry fracture, there are only two—normal and shear fracture compliances). One of the five parameters represents the fracture-normal hydraulic permeability, but none is related to the fracture-parallel permeability.

SIGNIFICANCE OF FINDINGS

The five characteristic parameters identified in the boundary conditions govern the scattering of seismic waves by a fluid-filled homogeneous fracture. Conversely, these are the only quantities that can possibly be determined from seismic measurements alone. An interesting and surprising finding is that the scattering of seismic waves is insensitive to the fracture-parallel permeability. (This was also confirmed using an independent numerical modeling.) A further investigation is under way to see if this is also true when the material properties within a fracture are heterogeneously distributed.



(a) Normal incidence frequency response. k_0=10^{-16} m.

(b) 1 kHz angle-of-incidence response. k_0=10^{-16} m^2

Figure 1. Transmission and reflection coefficients of a thin (1mm), homogeneous fracture for incident Biot's fast P-waves. The coefficients were computed using both an exact method and the simplified poroelastic fracture boundary conditions with the five characteristic fracture parameters.

RELATED PUBLICATION

Nakagawa, S., and M.A. Schoenberg, Poroelastic modeling of seismic boundary conditions across a fracture. LBNL-60862. J. Acoust. Soc. Am, 122(2), 831–847, 2007.

ACKNOWLEDGMENTS

This work was supported by the Director, Office of Science, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences, and Biosciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

Large Scale 3-D Electromagnetic Inversion Problems

Gregory A Newman and Michael Commer
Contact: Gregory A. Newman, 510/486-6887, ganewman@lbl.gov

RESEARCH OBJECTIVES

Large-scale controlled source electromagnetic (CSEM) 3-D geophysical imaging is now receiving considerable attention for mapping complex geological systems, with emphasis on mapping fluids associated with potential oil and gas and geothermal reservoirs and subsurface contamination. When combined with established seismic methods, direct imaging of reservoir fluids is possible. While modeling in 1-D is relatively easy, and trial-and-error 3-D forward modeling is straightforward, 3-D imaging is necessary in highly complex and subtle geological environments. Faster 2-D CSEM imaging technology has some relevance to this problem, but because of its assumption of 2-D geology, it cannot be relied upon for a consistent treatment of the 3-D imaging problem, especially when data are acquired specifically for a 3-D imaging experiment. Because of the size of the 3-D CSEM imaging, problem strategies are required that exploit computational parallelism and optimal meshing.

APPROACH

New techniques for improving both the computational and imaging performance for 3-D electromagnetic (EM) inversion have been developed. A nonlinear conjugate gradient algorithm is the framework of the inversion scheme. Full wave equation modeling for controlled sources is utilized for the data simulation and efficient gradient computation needed for the iterative model update. Improving the modeling efficiency of the 3-D finite difference method involves the separation of the potentially large model mesh, defining the set of model parameters, from the computational finite-difference meshes used for field solution. Meshing and thus overall mesh sizes can be reduced and optimized according to source, frequencies, and source-receiver offsets of a given input data set. Further computational efficiency is obtained by combining different levels of parallelization over the model and data spaces, avoiding the performance loss caused by computationally idle message-passing when increasing the number of parallel tasks. Image enhancement is achieved by model parameter transformation functions, which enforce bounds on the conductivity parameters and thus prevent parameter overshoots. Further, we have developed a remedy for treating distorted data within the inversion process. Data distortions simulated here include positioning errors and a highly conductive overburden, hiding the desired target signal. The methods are demonstrated using synthetic and field data.

ACCOMPLISHMENTS

In a recent imaging experiment, we have demonstrated the power of our 3-D imaging approach, where 32,768 tasks/processors were utilized on the IBM Watson Research Blue Gene/L supercomputer. Over a 24-hour period, we were able to image a large-scale field data set that previously required over four months of processing time on distributed clusters, based on Intel or AMD processors utilizing 1024 tasks on an Infiniband fabric. Results of the Blue Gene/L experiment showed that the broadside inline component data displays a systematic bias that could not be fit to a degree that is within the anticipated noise level of the measurements; other field components were satisfactorily fit. Modeling confirms that a likely explanation for this outcome is the need to incorporate conductivity that exhibits transverse anisotropy within the 3-D model. The speed at which the Blue Gene/L platform delivered this result is consistent with time frames required by practical exploration problems.

SIGNIFICANCE OF FINDINGS

We have made significant progress in reducing computational demands for solving large-scale, 3-D, electromagnetic imaging problems.

ACKNOWLEDGMENTS

Funding for this work was provided by the ExxonMobil Corporation, and by the Director, Office of Science, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences, and Biosciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

Enhanced Geothermal Systems (EGS) Using CO_2 as Working Fluid

Karsten Pruess
Contact: 510/486-6732, K_Pruess@lbl.gov

RESEARCH OBJECTIVES

The resource base for geothermal energy is enormous, but commercial production of geothermal energy is currently limited to high-grade hydrothermal systems, in which naturally present fracture networks permit fluid circulation, and allow geothermal heat to be produced by tapping these hot fluids through wellbores. The concept of "enhanced geothermal systems" (EGS) aims to extract geothermal energy from lower-grade resources by stimulating fracture permeability and extracting energy through a system of injection and production boreholes. Previous attempts to develop EGS in the U.S. and other countries have all employed water as heat transmission fluid and have met with limited success. At geothermal temperatures, water is a powerful solvent for many rock minerals, which poses great difficulties in achieving and maintaining water circulation at adequate rates.

The purpose of this research was to explore an alternative concept, first proposed by Donald Brown (2000), that would use carbon dioxide (CO_2) instead of water as a heat-transmission fluid.

APPROACH

Mathematical modeling capabilities for flow of water and CO_2 previously developed in the DOE geothermal and carbon management programs were enhanced to permit application to temperatures of up to 250ºC. These capabilities were used to evaluate and compare flow of water and CO_2 in injection and production wells, and to analyze heat sweep in fractured reservoirs.

ACCOMPLISHMENTS

At temperature and pressure conditions of interest, typically 200ºC and 500 bar, CO_2 has far greater compressibility and expansivity than water. As a consequence, for comparable injection conditions, a CO_2 production well would have far greater wellhead pressure than a water production well. This finding is very favorable, as it implies that EGS operated with CO_2 would require less pumping power than water to maintain fluid circulation, and may in fact achieve commercially adequate flow rates without artificial pumping. CO_2 also has very favorable heat extraction characteristics. Figure 1 compares numerically simulated heat extraction for a typical EGS that would be operated either with water or with CO_2. Heat extraction rates with CO_2 are seen to be approximately 50% larger than with water.

SIGNIFICANCE OF FINDINGS

Initial evaluation of this novel concept—operating enhanced geothermal systems with CO_2—has shown very promising results. In addition to favorable wellbore hydraulics and reservoir heat extraction, inevitable fluid losses during EGS operation provide opportunities for carbon storage in CO_2-driven systems, whereas in water systems, fluid losses present a severe economic liability.

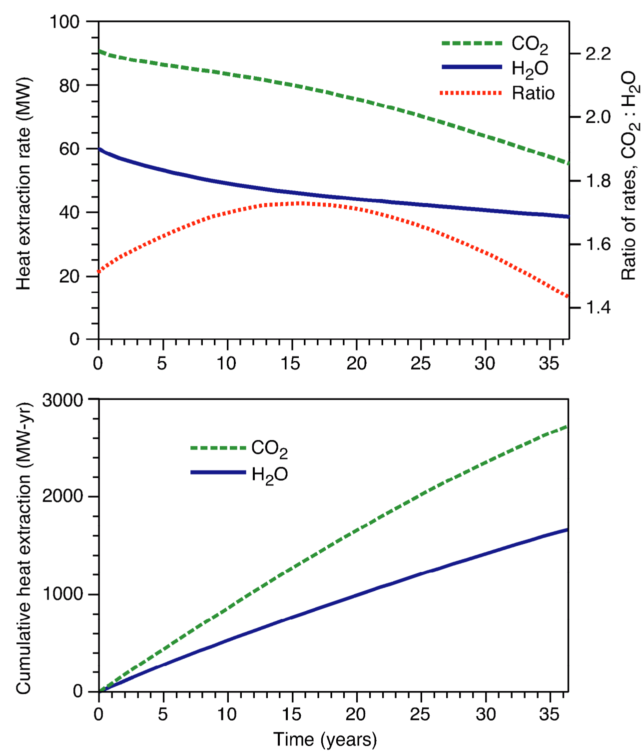


Figure 1. Rate of net heat extraction (top) and cumulative heat recovery (bottom) for a typical EGS reservoir, operated at (T, P) = (200°C, 500 bar)

RELATED PUBLICATIONS

Pruess, K. and M. Azaroual, On the feasibility of using supercritical CO_2 as heat transmission fluid in an engineered hot dry rock geothermal system. Proceedings, 31st Workshop on Geothermal Reservoir Engineering, Stanford University, Stanford, CA, January 30–February 1, 2006.

Pruess, K. Enhanced geothermal systems (EGS) using CO_2 as working fluid—A novel approach for generating renewable energy with simultaneous sequestration of carbon. Geothermics, 35(4), 351–367, August 2006.

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary for Energy Efficiency and Renewable Energy, Office of Geothermal Technologies, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

Oceanic Gas-Hydrate Instability and Dissociation under Climate Change Scenarios

Matthew T. Reagan and George J. Moridis
Contact: Matthew T. Reagan, 510/486-6517, mtreagan@lbl.gov

RESEARCH OBJECTIVES

The dissociation of accumulated oceanic gas-hydrate deposits and the release of large quantities of methane—a powerful greenhouse gas—over a short period of time could have dramatic climatic consequences, leading to further atmospheric and oceanic warming and accelerated decomposition of the remaining hydrates. This positive-feedback mechanism has been proposed as a significant contributor to rapid and significant climate changes in the past. However, the behavior of contemporary oceanic methane-hydrate deposits subjected to rapid temperature changes, like those predicted under future climate change scenarios, is poorly understood. In this study, we simulated the dynamic response of several types of oceanic gas-hydrate accumulations to temperature changes at the seafloor and assessed the potential for methane release into the ecosystem.

APPROACH

The amount of methane hydrate currently residing in the deep ocean and along continental margins is estimated to be from 500 billion to over 3,000 billion tons. In oceanic deposits, hydrate stability depends on the pressure (imposed by the water depth) and temperature. Figure 1 presents a general schematic of the hydrate-phase boundary (red) and gas-hydrate stability zone (GHSZ) for oceanic hydrates (shaded area). An increase in water temperature at the seafloor (a shift from Temperature Profile 1 to Profile 2) lowers the position of the top of the GHSZ (A) and raises the position of the bottom of the GHSZ (B). Such a shift could induce hydrate dissociation and lead to the release of methane into the ocean and atmosphere.

We modeled oceanic hydrate deposits under a range of depths and temperatures, using the TOUGH+HYDRATE code. We simulated temperature increases of 1ºC to 5ºC over a 100 yr simulation period, representing possible changes in seafloor temperature as predicted by advanced climate simulators, and determined the dynamic evolution of dissociating hydrates and the transport of released methane through benthic sediments to the seafloor.

ACCOMPLISHMENTS

The simulations found that deep, cold hydrates at 1,000 m or greater depth are stable under all anticipated temperature-change scenarios, and methane release was insignificant. Warmer, shallower hydrate deposits, representative of Gulf of Mexico formations, exhibited a stronger response, producing significant aqueous and gaseous fluxes of methane. These fluxes, however, were within the range of anaerobic methane oxidation rates in benthic sediments. A shallow, cold hydrate deposit, representative of the arctic continental shelf, exhibited the strongest response, with explosive releases of methane, primarily in the gaseous phase, at rates that greatly exceed possible chemical or biological consumption.

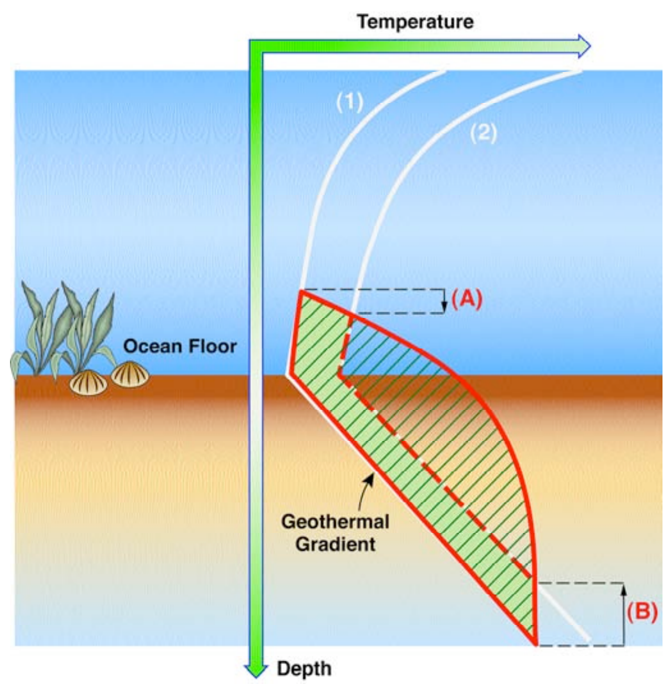


Figure 1. Schematic of the gas hydrate stability zone in the seafloor environment and its relationship to ocean temperature changes

SIGNIFICANCE OF FINDINGS

These results indicate that while many deep oceanic hydrate deposits are indeed stable under the influence of significant temperature variations, shallow deposits, such as those found in arctic regions or in the Gulf of Mexico, can undergo rapid dissociation and release large quantities of carbon. In arctic regions in particular, temperature changes are expected to be more pronounced, increasing the risk of hydrate destabilization even further. These simulations can provide a source term to global climate models, yielding a prediction of the possible effects of oceanic hydrate decomposition on global climate.

ACKNOWLEDGMENTS

This work was supported by Laboratory Directed Research and Development (LDRD) funding from Berkeley Lab, provided by the Director, Office of Science, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

A Numerical Model for Analysis of the Geomechanical Performance of Hydrate-Bearing Sediments

Jonny Rutqvist and George Moridis
Contact: Jonny Rutqvist, 510/486-5432, jrutqvist@lbl.gov

RESEARCH OBJECTIVES

Several methods are being considered for production of gas from hydrate-bearing sediments (HBS), including depressurization, thermal methods, and inhibitor injection. However, methane hydrate drilling and production operation may pose a significant hazard, because thermal and mechanical loading can result in hydrate dissociation and a significant pressure increase, with potentially adverse consequences on wellbore stability and the structural integrity and stability of the HBS. The objective of this research project is to build a simulator that can be used for scientific and engineering analyses of hydrate stability, including well bore and seafloor stability during production from oceanic hydrate formations.

APPROACH

The starting point of our approach is the TOUGH+HYDRATE simulator, which is an advanced code currently available for the simulation of multiphase flow system behavior in geological media containing gas hydrates. To consider geomechanical effects, the TOUGH+HYDRATE has been coupled with FLAC3D, a code widely used for soil and rock mechanics engineering, and for scientific research in academia. The two codes, TOUGH+HYDRATE and FLAC3D, are linked through a coupled thermal-hydrological-mechanical (THM) model of the HBS (Figure 1). Based on the coupled THM model, coupling functions are developed that serve to pass relevant parameters between the multiphase heat and transport analysis in TOUGH+HYDRATE and the geomechanical stress-strain analysis in FLAC3D.

ACCOMPLISHMENTS

The first version of the simulator is now fully operational and is currently being tested on a suite of problems related to geomechanical behavior of the HBS (Rutqvist and Moridis, 2007). The first involves hydrate heating as warm fluids from deeper conventional reservoirs ascend to the ocean floor, through uninsulated pipes intersecting the HBS. The second case describes system response during gas production from a hydrate deposit, and the third involves mechanical loading caused by the weight of structures placed on hydrate-bearing sediments at the ocean floor. Our simulation results indicate that the stability of HBS in the vicinity of warm pipes may be significantly affected, especially if the sediments are unconsolidated and more compressible. Gas production from oceanic deposits may also affect the geomechanical stability of HBS under the conditions that are deemed desirable for production. Conversely, the increased pressure caused by the weight of structures on the ocean floor increases the stability of hydrates.

SIGNIFICANCE OF FINDINGS

Methane hydrates occur naturally offshore in shallow depths below the ocean floor and onshore beneath the permafrost. They contain enormous quantities of methane gas, which if economically producible will have significant implications for U.S. energy security. The developed numerical model provides a state-of-the-art numerical tool for analyzing geomechanical hazards and optimizing production from hydrate-bearing sediments.

RELATED PUBLICATIONS

Rutqvist, J. and G. Moridis, Numerical studies on the geomechanical stability of hydrate-bearing sediments. OTC-18860, Presented at the 2007 Offshore Technology Conference held in Houston, Texas, U.S.A., April 30–May 3, 2007.

ACKNOWLEDGMENTS

This work was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Energy Technology Laboratory, under the U.S. Department of Energy, Contract No. DE-AC02-05CH11231.

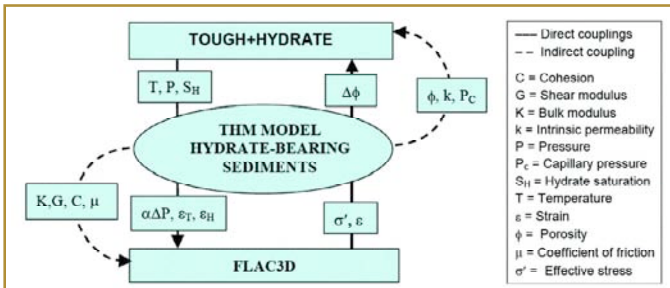


Figure 1. Linking of TOUGH+HYDRATE with FLAC3D for analyzing the geomechanical behavior of the HBS.

Simulations of Induced Seismicity by Injection and Production at The Geysers Geothermal Field

Jonny Rutqvist and Curtis M. Oldenburg
Contact: Jonny Rutqvist, 510/486-5432, jrutqvist@lbl.gov

RESEARCH OBJECTIVES

The Geysers Geothermal Field in Northern California is the site of the largest geothermal electricity generating operation in the world. It is also one of the most seismically active regions in California. At The Geysers, water injection into the steam-dominated reservoir is necessary to maintain reservoir pressure and economical production. However, an increasing injection rate over the years has also resulted in an increased level of seismicity, which has raised concern in the local communities. The purpose of this study is to investigate the causes and mechanisms of seismicity at the Geysers and to develop injection and production strategies that minimize induced seismicity while maximizing energy recovery.

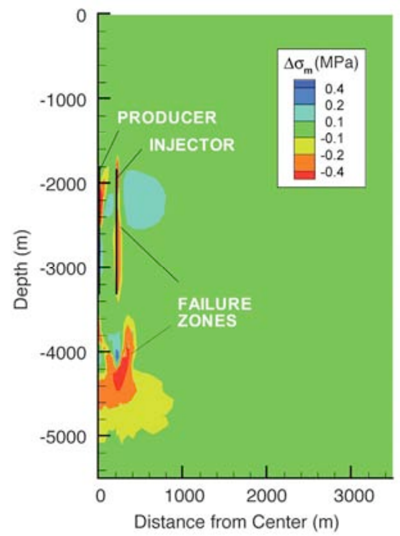


Figure 1. Simulated distribution of potential failure after 6 months of seasonally varied injection. Yellow and red contours show areas where effective stress changes have moved into a state of failure indicating high potential for induced seismicity.

APPROACH

The approach we take is numerical simulation, using coupled hydrogeomechanical models for the reservoir based on TOUGH2 and TOUGH-FLAC. Simulation results are integrated with observations from expanded seismic arrays and satellite-based surface strain at The Geysers. The coupled-reservoir geomechanical analysis described herein is used to calculate the time evolution of the three-dimensional stress field during steam production and cold-water injection, and to evaluate the potential evolution and distribution of microearthquakes (MEQs) using various failure criteria. An important aspect of the analysis is the concept of a rock mass that is critically stressed for shear failure, conditions under which very small perturbations of the stress field can trigger seismicity.

ACCOMPLISHMENTS

We have conducted numerical simulations to analyze the potential for induced seismicity during both steam production and cold-water injection. Figure 1 shows injection-induced high potential for failure indicated by orange and yellow contours occurring near the injection well and in a plume at a distance several kilometers below in the injection well. This is consistent with the pattern of injection-induced seismicity observed at The Geysers. Overall, our analysis shows that the most important cause for injection-induced seismicity at The Geysers is injection-induced cooling and associated thermal-elastic shrinkage, which changes the stress state in such a way that mechanical failure and seismicity can be induced. Specifically, the cooling shrinkage results in unloading and associated loss of shear strength in critically shear-stressed fractures, which are then reactivated.

SIGNIFICANCE OF FINDINGS

With an understanding of the mechanism of induced seismicity at the Geysers identified through numerical simulation, we are now in a position to simulate reservoir operations that minimize induced seismicity. The ability to reduce MEQs associated with enhanced geothermal systems (EGS) is important for public acceptance and for optimizing energy production.

RELATED PUBLICATIONS

Rutqvist, J., E. Majer, C. Oldenburg, J. Peterson J., and D. Vasco, Integrated modeling and field study of potential mechanisms for induced seismicity at The Geysers Geothermal Field, California. GRC Transactions, 30, 629–633, 2006.

Rutqvist, J. and C. Oldenburg, Analysis of cause and mechanism for injection-induced seismicity at the Geysers Geothermal Field, California Geothermal Research Council, Annual Meeting, Sparks, Nevada, September 30–October 3, 2007.

ACKNOWLEDGMENTS

This work was conducted with funding from the California Energy Commission (CEC) with matching funds from the Assistant Secretary for Energy Efficiency and Renewable Energy, Geothermal Technologies Program, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

Imaging Electronic and Atomic Redistribution During Redox Reactions at Surfaces

Glenn Waychunas, Ben Gilbert, Roger Falcone, Jillian Banfield, Klaus Attenkofer, and Robert Schoenlein
Contact: Glenn Waychunas, 510/495-2224, gawaychunas@lbl.gov, or Ben Gilbert, 510/495-2748, bgilbert@lbl.gov

RESEARCH OBJECTIVES

A number of important geochemical reactions are activated by electron transfer at mineral surfaces. Processes include reductive and oxidative dissolution, coupled redox-sorption of complexes, respiration of surface-associated biota, photoactivated reactions, and electrochemical cell reactions. Our research focuses on electron-transfer-initiated dissolution reactions, which are particularly important processes in the generation of acid mine drainage and resultant pollutant transfer in the environment. The crucial electron transfer step is faster than can be studied by currently available means, but the series of succeeding relaxation steps can be probed to elucidate much of the reaction mechanism.

APPROACH

In reductive dissolution processes, electrons are transferred from solution species via shared surface ligands to surface metal ions, like Fe(III), producing surface Fe(II) species. The Fe(II)-O bonds at the surface must lengthen from this reaction, introducing surface vibrations and a general expansion of the surface. This localized strain may act to limit the density of Fe(II) that can be produced at the surface, depending on the rate of electron transfer within the mineral. We wish to measure the rate of Fe(II) buildup on Fe oxide surfaces and the ultimate concentration limit, as well as surface and bulk conduction rates. We are conducting such experiments at the Advanced Photon Source at Argonne National Laboratory (and soon also at the Advanced Light Source at Berkeley Lab), starting with observations of Fe(II) creation from a photostimulated ligand attached to Fe oxide nanoparticles and single crystal surfaces. A femtosecond laser system activates electron transfer from the ligand, and the synchrotron x-rays detect the result at variable delay times after the excitation. This is a "pump-probe" experiment.

ACCOMPLISHMENTS

Our first static experiments at the APS beamline 13ID showed proof of concept with respect to electron injection from the organic ligand alizarin red into the (1-102) hematite surface. A significant amount of surface Fe(II) (Figure 1) is produced within minutes of exposure to the dye. We observe the Fe(II) by collecting surface diffraction data via the (10L) crystal truncation rod. Collection of complete data sets will enable refinement of the electronic configuration of the excited hematite surface, and this process can be done as a function of time-delay to detect the time-resolved progress of charge migration and relaxation. Our first time-resolved experiments at the APS beamline 11-ID showed measurable Fe(II) creation in 2 nm maghemite nanoparticles coated with eosin Y surface ligands with a time delay of 154 ns. In these experiments, the nanoparticles were studied in recycled suspension, and the near Fe K-edge x-ray absorption spectra was collected.

SIGNIFICANCE OF FINDINGS

The static results on large single crystal hematite surfaces demonstrate the sensitivity of the CTR method to the presence of reduced surface atoms. We observe that significant concentrations of Fe(II) are stable for long time periods, but that control of the injection process may be difficult. The time-resolved nanoparticle results show that varied ligands can have much different injection probability, and that beam damage (either from laser or x-rays) may have a significant effect on nanoparticle structure or chemistry. Both issues will be investigated in the next series of experiments.

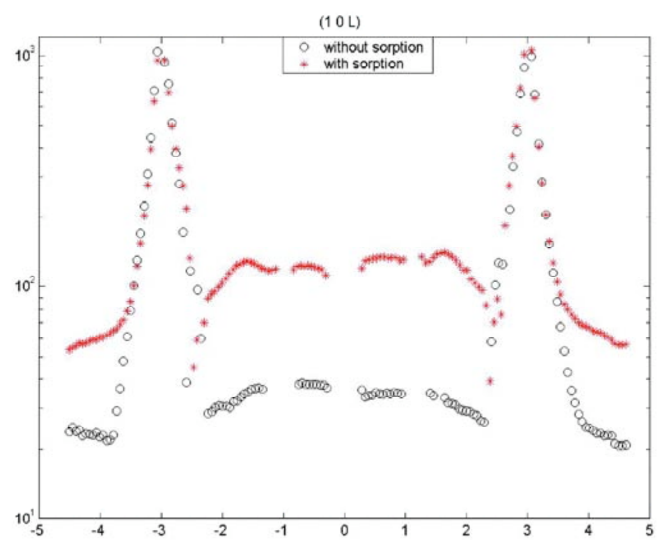


Figure 1. (10L) crystal truncation rod measured for the bare hematite (1-102) surface without alizarin red sorption (circles), and after 10-minute exposure to alizarin red solution in ambient light (red asterisks). The features at L=-1.6, 1.6 are due to surface Fe(II).

ACKNOWLEDGMENTS

This work was supported by the Director, Office of Science, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences, and Biosciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. We thank Dr. Lin Chen (Argonne National Laboratory Chemistry Division) for the use of the LITR-XAS facility at the Advanced Photon Source.