

## Methane gas as a renewable energy: Sustainable hydrocarbon energy resources development by carbon recycling CCS

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From a commonsense standpoint, methane gas is a kind of fossil fuel that is a limited energy resource. It is believed that the underground methane was generated mostly by thermal decomposition of oil, coal or other organic matter - fossil. However, large amounts of habitat of methanogens (archaea) have been found recently in wide range of underground environments. Many types of methanogens produce methane from organic acids which are also fossil origin. However, CO<sub>2</sub>-reducer methanogens produce methane from hydrogen and CO<sub>2</sub> which may be derived from the mantle or hydrothermal reaction in deep rocks instead of the direct seepage of deep methane in the unproven Thomas Gold hypothesis (abiogenic petroleum origin). Methane produced from abiogenic deep CO<sub>2</sub> and hydrogen predominant in deep igneous rocks and in basaltic oceanic crust is not a fossil energy resource but a renewable energy resource. In particular, the methane hydrate in deep oceanic crust is a renewable energy resource.

The stored CO<sub>2</sub> in underground reservoirs by the CO<sub>2</sub> capture and storage (CCS) will be converted into methane by the underground CO<sub>2</sub>-reducer methanogens. Then, the converted methane will be available as a renewable energy while CO<sub>2</sub> emission into the atmosphere is suppressed. The sustainable hydrocarbon energy development becomes reality by the worldwide systematic deployment of this CO<sub>2</sub>-recycling CCS.

Keywords: CCS, carbon recycling, renewable energy, methanogen, fossil energy, methane hydrate

## Classification and global circulation treatment of carbon circulation system for greenhouse gas

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Present strategy of global warming affairs with carbon by domestic and foreign researchers is treated to simple education, science and technology to reduce promptly the use and stop for global carbon dioxides from fossil fuel producing after the previous industrial revolution (ca. 150 years ago).

However, strong precept of lesson on 3.11 nuclear power plant accident is lacking of our long terrestrial historical knowledge only to stop and waste shortly in our short human activity, though artificial energy of nuclear power development spent the wisdom of all human society to be maintained.

Author proposes now dynamic global carbon cycle to control completely by human activity, not only global fossil carbon-dioxides with recycling but also extraterrestrial carbon cycling including the solar and solar system sources, which as a breakthrough innovation (not only stop and waste) .

The carbon cycle on Earth reveals major three types as follows:

- 1) Long-period global carbon cycle: coal carbon of long geologic period in millions years period.
- 2) Short-term local biological carbon cycle: carbon of short biological system of a year period.
- 3) Industry-originated carbon dioxides: carbon from industrial carbon dioxides for major cause of global warming.

Before industrial revolution (150 years ago), the above 1) and 2) circulation systems of carbon are largely ruled on the Earth planet.

However, after the industrial revolution (about 150 years ago), the above 3) short carbon system are produced and mixed at the above 1) and 2) carbon systems. Therefore the complex 1) 2) and 3) carbon systems are mainly produced the recent global warming now.

Dynamic stable systems of three carbon systems are required at human society to control the above 3) carbon system completely, by suitable science and technology developments in our fields.

This is mainly because old geologic period to form coal carbon (from ca.3 to 4 million years ago) which has been largely changed the contents of carbon dioxides, should be controled the carbon circulation system on global human treatment (not only to stop as garbage idea).

Keywords: carbon circulation system, dynamic stable system, short carbon cycle, long carbon cycle, industrial revolution, T-ET carbon cycles

## Evaluation of CO<sub>2</sub> Mineral Trapping Rates in Aquifers based on experimental studies

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In this study, experiments on CO<sub>2</sub>-water-rock interaction have been conducted to elucidate the rock dissolution rate and to investigate long-term dissolution and precipitation phenomena in CO<sub>2</sub> reservoirs. The dissolution experiments are conducted by using semi-open experimental system constructed for this study. As the rock samples, in addition to the basalt which is considered as a suitable candidate rock formation for geochemical trapping of CO<sub>2</sub>, tuffaceous sandstone (Hayama group: Kanagawa Prefecture) and three green tuff rocks (Tsugawa formation: Niigata Prefecture, Ushikiri formation: Shimane Prefecture, Daijima formation: Akita prefecture) from the Quaternary igneous rocks widely distributed in Japan were used.

From the eight-month-period of experiments, the facts found were that the composition of formation water will converge at the point where the rock dissolution and precipitation of secondary mineral are balanced and CO<sub>2</sub>-water-rock interaction proceeds under a certain formation water composition. For this reason, the determination of rock dissolution rate (element release rate) under a certain formation water composition inherent in each rock sample is indispensable in order to predict the long-term progress of the reaction within CO<sub>2</sub> reservoirs.

Si release rate under a certain formation water composition that indicates the dissolution of silicate minerals from each rock sample is  $29.8 \times 10^{-2}$  mmol/kg-rock/day for basalt,  $7.77 \times 10^{-2}$  mmol/kg-rock/day for Tsugawa green tuff,  $5.44 \times 10^{-2}$  mmol/kg-rock/day for Ushikiri green tuff and  $33.1 \times 10^{-2}$  mmol/kg-rock/day for Daijima green tuff at the temperature of 50°C.

The simulations on long-term CO<sub>2</sub> fixation efficiency (mineral trapping) in the CO<sub>2</sub> reservoir by using Ca, Mg and Fe release rates calculated from experiments were conducted. On the assumption that 1: CO<sub>2</sub> injection rate to be 2,000 ton/day 2: injection time period to be 50 years (total amount of injected CO<sub>2</sub> is 36,500,000 t) 3: target aquifer porosity 20% 4: CO<sub>2</sub> density 500 kg/m<sup>3</sup> 5: injected CO<sub>2</sub> to groundwater volume ratio 1:2, the time required for mineral fixation of 36,500,000 tons of CO<sub>2</sub> is simulated to be about 180 years for basalt, about 5,100,000 tons of CO<sub>2</sub> fixed as a carbonate mineral in 200 years for Tsugawa green tuff, about 22,000,000 tons of CO<sub>2</sub> fixed in 200 years for Ushikiri green tuff and 3,900,000 tons of CO<sub>2</sub> fixed in 200 years for Daijima green tuff. at the temperature of 50°C.

These results indicate that the mineral trapping rate in CO<sub>2</sub> reservoir is much faster than the results of previous studies and that geochemical trapping (mineral trapping) is an important mechanism not only for long-term (10<sup>3</sup> - 10<sup>4</sup> years) security but also for shorter-term (- 10<sup>2</sup> years) security of CO<sub>2</sub> aquifer storage and is a significant indicator for the selection of potential storage candidate site.

Keywords: CCS, CO<sub>2</sub> geological storage, water-rock interaction, Green-Tuff, Basalt

## Time-lapse field experiment using seismic ACROSS at the air injection into the shallow ground in Awaji Island-I

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### 1.Introduction

In the monitoring of physical states of CO<sub>2</sub> storage zones in CCS, oil-gas reservoirs, fluid flow along subduction zones and migration of magma bodies, the development of time lapse or 4D observation methods are very important. However, any effective methods for the above objectives have not been established yet. We have developed an effective method for time-lapse measurements using seismic ACROSS (Accurately Controlled and Routinely Operated Signal System) and seismic arrays. We showed nice imaging results by simulation using a few seismic sources assuming ACROSSs and seismic array (Kasahara et al., 2011; Hasada et al. 2011).

Although temporal changes have been reported in the past ACROSS observations, the separation of true changes in the deeper ground from near-surface changes are not enough. In order to prove the usefulness of our method, we carried out a field test using air injection into the ground as the artificial cause, ACROSS seismic sources and multi-receiver array.

### 2.Field experiment and processing

We carried out field experiment near the Nojima Fault System in Awaji Island, Japan from February 20 to March 10, 2011. We simultaneously used two seismic ACROSS; ACROSS-V with vertical rotational axis for the eccentric weight mass owned by Nagoya University and a newly built seismic ACROSS-H having the horizontal rotational axis to generate vertical and horizontal vibrations. The ACROSS-H generated 10-30Hz and the ACROSS-V generated 10-35Hz, though the latter unit has a potential up to 50Hz.

The one hour unit of each ACROSS signal comprised of 32 repetitions of 10-35Hz (or 10-30Hz) sweep and 400-seconds transition. The rotational directions were switched every one hour. By division of observed data by designed source spectrum, the transfer functions between the source and each receiver were obtained.

The geophones are placed in the 1km square region near the Nojima fault system. The injection point was at the center and two ACROSSs were at NE and SE of the region, respectively. 80ton air in total with 21MPa was injected into the Osaka formation at 100m depth between February 26 and March 3. 32 3-components and one 800m-borehole geophones were used. Although we used four different types of geophones, it does not matter because the frequency bands used in this experiment are >10Hz. In this presentation, we show only the results obtained by the ACROSS-H. For the ACROSS-H, we can synthesize vertical and horizontal forces by combining the clockwise and anticlockwise rotations (Kasahara et al. 2011b). Vertical and horizontal vibrations efficiently generate P and S waves, respectively.

In order to image the disturbed zone, we carried out time-reversal method (Kasahara et al.,2011a).

### 3.Results

Except for one malfunctioned unit, all surface and borehole geophones showed very large travel time and waveform changes. The largest changes were observed at stations #6 and #7 after one day.

The later phases change more than the first arrivals of P and S. Despite major P and S phases seen in travel-time vs. distance diagram, the interpretation for details appeared on each component is difficult. The change at 800m borehole is not easily interpreted because the borehole is away from the direct path from the source to the injection point. The results of time-reversal method to image disturbances zone due to the injection shows centralized during 8 hours after the start of injecting air and gradually migrates toward eastward.

### 4.Conclusions

By the injection experiment in Awaji Island using seismic ACROSS, we confirmed the effectiveness of our time-lapse method to image the disturbed zone. Although the behavior of air is not the same as supercritical CO<sub>2</sub>, the first step for the CCS and CO<sub>2</sub>-EOR can be obtained by the combination of seismic ACROSS and multi-receivers. This method is also applicable to monitor of seismogenic zones and volcanic areas.

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Keywords: CCS, Global Climate Change, Time Lapse, Back propagation, CO2-EOR, 4D

## Microseismic monitoring at CO<sub>2</sub> geological storage site - Initial data results observed at Cranfield in the U.S.-

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There are a lot of discussions on possible microseismicities induced by CO<sub>2</sub> geological storage. Case studies at CO<sub>2</sub> injection sites overseas suggest that the microseismicities caused by the CO<sub>2</sub> injection might be quite small should it occur, but monitoring at the injection field is necessary for ensuring safe carbon geological sequestration (CGS) and gaining public acceptance.

RITE has studied the microseismicities induced by CO<sub>2</sub> injection under partnership and collaboration with the Lawrence Berkeley National Laboratory and the Bureau of Economic Geology of the University of Texas. A long-term observation is underway to monitor the microseismicities at a large-scale CO<sub>2</sub> injection site in the U.S. Based on data and knowledge obtained by the observation, RITE will study a relationship between the CO<sub>2</sub> injection and microseismicities (presence/absence, scale and distribution of the microseismicities) and establish a technique of observing microseismicities, which will be demanded for prospective CGS demonstrations and practical implementation in Japan.

The observation site is located at an oil field of Cranfield in Mississippi. Enhanced oil recovery using CO<sub>2</sub> has been implemented there and approximately 3 M tons of CO<sub>2</sub> were injected. RITE deployed an observation network by installing 6 geophones within the radius of about 3 Kilometers in the field, and began the microseismic monitoring from December, 2011. This paper describes overview of the microseismic monitoring at the Cranfield site and the initial data results.

Keywords: CO<sub>2</sub> geological storage, Microseismic monitoring

## Numerical Simulation of the Effects of Sandstone channels Properties on the Seal Integrity in Geological Storage of CO<sub>2</sub>

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Geological storage of CO<sub>2</sub> is one of the methods to mitigate the global warming. Several kinds of reservoir are suggested including depleted oil/gas fields, unminable coal seams, and deep saline aquifers, test and demonstration projects for which are underway. In Japan, saline aquifers without structural trapping are known to keep water soluble methane gas and are considered to be the targets of geological storage of CO<sub>2</sub>.

If the seal capacity and the continuity of the layer located immediately above the reservoir are sufficient, all injected CO<sub>2</sub> is expected to be stored within the reservoir. However, even if the global permeability of mudstone seal seems low enough, the presence of intra-layer sandstone would significantly degrade the seal integrity. The past work by the authors indicated that the presence of those sandstone channels have large effects on the long-term fluid behavior by numerical simulations adopting the double porosity model to the seal layers.

In this study, we will present the results of numerical simulations to indicate the effects of the presence and properties of sandstone channels such as the volume fraction, spacing and permeability (or permeability ratio to the mudstone seal) on the seal integrity. The results include the behavior of CO<sub>2</sub> injected into a deep saline aquifer at a depth of 1000 m, dissolution and residual gas trapping, and pressure buildup and propagation. The global permeability of seal layers is set to be low enough to keep CO<sub>2</sub> below if they are homogeneous porous media. Calculations are carried out using the "STAR" general-purpose reservoir simulation code with the "SQSCO" equation of state.

Keywords: Geological storage of CO<sub>2</sub>, saline aquifer, double porosity model, numerical simulation

## Study of measuring method for supercritical CO<sub>2</sub> threshold pressure on several mudstone

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Geological storage of CO<sub>2</sub> is one of the measures to mitigate global warming. As the density of supercritical CO<sub>2</sub> is lower than the formation water, in order to safety storage of CO<sub>2</sub>, it is necessary impermeable layer at the top of the aruifer. Impermeable layer such as mudstone indicates a specific threshold pressure, at the CO<sub>2</sub> injection pressure is greater than the threshold pressure, CO<sub>2</sub> penetrates into seal layer. Therefore, appropriate evaluation of the threshold pressure is important for the safety and economic efficiency of CO<sub>2</sub> geological storage. As the testing methods of threshold pressure, there are step by step method, residual pressure difference method, dynamic fluid method. In this paper, we propose a new method of constant ejection rate method that test time is short and to evaluate the threshold pressure from the differential pressure change. Mudstone used in the tests are collected in the outcrop of the number of domestic places, were formed into a cylindrical shape. In all tests, pore pressure is 10MPa, temperature is 40 °C, and so CO<sub>2</sub> becomes supercritical phase. In preparatory stage of each test, permeability measurements were performed by steady state method

The test I were carried out by step by step method as the confining pressure is 20MPa. Water ejection has started at the differential pressure reaches to 1.24MPa and 70 hours elapsed from the start of CO<sub>2</sub> injection through 0.1MPa pressure increments every six hours. Threshold pressure is 1.24MPa to be evaluated.

The test II were carried out at constant ejection rate method as the confining pressure is 12MPa. The flow rate of ejection was controlled 0.004ml/min constant. Pore pressure of ejection side is decreased rapidly after 4 hours, showed a differential pressure 0.71MPa after 10 hours. The differential pressure change of 0.59MPa from 0.12MPa to 0.71MPa is considered equivalent for the threshold pressure.

The test III were carried out at constant ejection rate method as the confining pressure is 20MPa. Strain changes were observed at the injection side and ejection side. The threshold pressure was evaluated as 0.85MPa. At the same time of the reduction of ejection pressure, compressive strain were observed at both side of injection and ejection. This suggests that the threshold pressure is expressed with the CO<sub>2</sub> arrival to the end face and pore pressure was reduced uniformly. Expansive strain was observed at injection side 3 hours after the reduction of ejection pressure and expansive strain was observed at enjection side 11 hours after the reduction of ejection pressure. This suggests that the increase of pore pressure at the point of CO<sub>2</sub> reached causes expensive strain.

The permeability of test I, test II and test III, were 3.1, 9.3 and 6.5 ×10<sup>-6</sup> darcy respectively. The relationship between threshold pressure and reciprocal permeability showed a clear positive correlation on both logarithm. Threshold pressure of supercritical CO<sub>2</sub> was higher than the threshold pressure of N<sub>2</sub>. Whereas the threshold pressure has been evaluated 70 hours after in the step by step method, could be evaluated 10 hours after in constant ejection rate method. The possibility of shortening the test time has been confirmed. Observation of the strain is useful in the interpretation the behavior of CO<sub>2</sub>.

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Keywords: Threshold Pressure, supercritical CO<sub>2</sub>, cap rock, nudstone



## Water saturation estimated by X-ray CT scan and mass balance methods during relative permeability measurements

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Predicting the migration of injected CO<sub>2</sub> is a major concern in carbon dioxide capture and storage (CCS) projects. The prediction requires relative flow properties between the injected CO<sub>2</sub> and water in a saline aquifer. Generally, numerical simulations of the multiphase flow in porous media use the relationship between the fractions of two fluids and their relative permeability, which is called a relative permeability curve. In CCS projects, the volumetric ratio between water and CO<sub>2</sub> in saline aquifers varies widely through the injection of CO<sub>2</sub>. Therefore, a simulation of the migration of CO<sub>2</sub> needs a relative permeability curve of water-CO<sub>2</sub> system.

If the relative permeability curve of water-supercritical CO<sub>2</sub> system can be obtained from laboratory measurements under the condition of the reservoirs, the migration of CO<sub>2</sub> in the reservoirs can be predicted more precisely. However, a very few studies have tried to measure accurate relative permeability curves of water-supercritical CO<sub>2</sub> system compared to those of water-oil system that has been usually measured in oil and natural gas development field. In many cases, outflow volumes of water and CO<sub>2</sub> from a rock sample are measured in a water-CO<sub>2</sub> separator. The separator should be pressurized so that CO<sub>2</sub> remains as supercritical state. Experiments without a pressurized separator fail to measure the accurate volume of CO<sub>2</sub> due to the change of CO<sub>2</sub> phase from supercritical to gas. Furthermore, the pressure change causes degassing of water, which is the release of water-dissolved CO<sub>2</sub> into a non-pressurized gas. Therefore, we developed a temperature controlled and pressurized separator to observe the interfacial surface between water and supercritical CO<sub>2</sub> through a glass window. This enables us to estimate water saturation of a rock sample accurately using mass balance on the fluids passing through the sample.

In addition to the mass balance, X-ray CT scanner is often used to determine the water saturation. X-ray CT scan is used in many of recent studies for the measurements of relative permeability. This must be because the resolution of X-ray CT scanners has been improved in recent years. We measured the relative permeability of water-supercritical CO<sub>2</sub> system through the estimation of water saturation obtained by both mass balance and X-ray CT scan. This study shows the methodology of measuring the relative permeability curve of water-supercritical CO<sub>2</sub> system and discusses the results of the measurements.

The relative permeability curve is obtained by plotting the relative permeability values with respect to the degree of water saturation estimated by X-ray CT scan and mass balance methods. The values of water saturation are almost the same as each other though there is a little difference between those obtained by X-ray CT scan and mass balance methods. Consequently, both mass balance methods and X-ray CT scanning can estimate water saturation in a rock sample precisely. Ideally, both methods should be used simultaneously to cross-check the value of water saturation as shown in this study.

Keywords: relative permeability measurements, water-supercritical CO<sub>2</sub> system, water saturation, mass balance methods, X-ray CT scanning

## An attempt of evaluation of well integrity at Nagaoka site using ultrasonic logging and CBL data

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For the safety of CO<sub>2</sub> sequestration, injected CO<sub>2</sub> must be trapped in the underground and not be allowed to leak to the surface. Well integrity is one of the essential problems because potential leakage could occur along the well (Celia et al., 2004). Cement between the casing and the formation will be the first material exposed to CO<sub>2</sub> among the well components, so the state of cement in a CO<sub>2</sub> rich environment has been studied (e.g., Kutchko et al., 2007). It is important to measure and monitor the integrity of wells that are exposed to CO<sub>2</sub>. This paper reports well integrity examined by the ultrasonic and sonic logging at Nagaoka CO<sub>2</sub> injection site.

Ultrasonic tool is used to measure the internal condition of the casing, the thickness of the casing, and the acoustic impedance of the material outside the casing. Observed reflected wave was different at the part of iron and FRP casing. The amplitude of the first reflection at the part of FRP casing was smaller since the impedance contrast between casing and water is smaller. We evaluated the impedance of the cement from the analysis of the amplitudes of the multiple reflections.

CBL is used to measure the bond between the casing and the cement, and the bond between the cement and the formation. The bond between the cement and the casing can be evaluated from the amplitude of the first reflection. The time-lapse observation of the CBL showed that the amplitude became smaller after the cementing. This means that the bond became better. The waveform showed the reflection from the interface between the cement and the formation. By combining the results of ultrasonic tool and some numerical calculations we would extract more information about the formation.

We note other logging program at Nagaoka. About 40 times sonic logging at Nagaoka from the injection period to the post injection period showed temporal change with the correlation of CO<sub>2</sub> saturation. The sonic velocity decreased when CO<sub>2</sub> arrived at the observation well. Another logging program is the sampling of the formation water using cased-hole dynamics tester (CHDT). This logging provides the information on chemical reaction and permeability. These results would be used for the interpretation of the state of the materials near the well.

We investigated the well integrity of the observation well at Nagaoka by the combination of the above logging method. The analysis showed that there is no clear evidence of the CO<sub>2</sub> leakage at Nagaoka.

Keywords: CO<sub>2</sub> geological storage, well integrity, Nagaoka, sonic logging

## Impact of lithofacies and reservoir heterogeneity on distribution of CO<sub>2</sub> at Nagaoka Pilot Site

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The Nagaoka pilot site is located in the onshore area of Nagaoka oil field in the Niigata plain of central Honshu on the Japan sea side of central Japan. The onshore deep saline aquifer utilized for the CO<sub>2</sub> storage project is located near the city of Nagaoka. The pilot site and the target formations were selected based on the geological information. The selected formation was the early Pleistocene Haizume Formation, around 60m thick and 1100m below the ground surface. Cores of the Haizume Formation at Nagaoka from well IW-1 show realistic reservoir characterization with litho-stratigraphic architecture.

To geologically model and assess CO<sub>2</sub> distribution, 3D seismic schemes were applied in the Nagaoka site, which included 4D seismic monitoring. Seismic attributes analysis is a popular and important method to predict the distributions of reservoir rock properties such as lithofacies, porosity, density, and thickness. Although 3D seismic survey has been executed, the distribution of lithofacies and the heterogeneity in reservoir layers remain unclear across the Nagaoka site because there are only few wells drilled. Therefore, we described the geomodeling framework and simulation studies that were applied to micro and macro scale reservoir modeling with realistic litho-stratigraphic architecture at the Nagaoka site.

Lithofacies relations and much of the heterogeneity in Nagaoka aquifer reservoirs are related to the stacking of depositional sequences. To investigate the challenges of the spreading CO<sub>2</sub>, a detailed reservoir heterogeneity model was set up based on analysis of 45 cores. The Haizume Formation consists of predominantly sandstone, alternating beds of siltstone and sandstone, siltstone, sandstone-argillaceous, and conglomerate. The marine deposits and consists of numerous thin shales (siltstone to mudstone) form the majority of heterogeneities in Haizume Formation. Heterogeneity is mainly controlled by the distribution of sandstones embedded in numerous alternated facies. The CO<sub>2</sub> reservoir formation in this site (ca. 20m thick) is divided into some independent zone layers with millimetric to decimetric laminations of sandstone, siltstone and mudstone, silts and sands alternate, conglomerate within sequences from metric to a few meters. In such a heterogeneous formation, the connectivity of permeable rocks is clearly of major concern for predicting of CO<sub>2</sub> storage potential. The realistic modeling of these connectivities is thus required to plan future developments, to understand and predict CO<sub>2</sub> behaviors.

This paper presents the realistic modeling strategy that was applied to Nagaoka site. The modeling strategy is multi steps, with first a geologically constrained generation of facies distributions, and second, simulations of spreading CO<sub>2</sub> variations with the measured permeability within the facies distributions. These descriptions were incorporated into the model at a resolution, which ensured capture of the most significant heterogeneities. The detailed reservoir model matched well log and core performance in this site. The detailed reservoir model and results of simulation matched the monitoring data from well and field more closely than the previous large scale models. The modeling technique also allows accounting for larger scale constraints, such as field wide variations of facies frequencies and main directions of spatial continuity.