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Experimental and Numerical Analysis on the Specific Storage of Low Permeable Sandstone for Supercritical CO₂

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Abstract: Experimental tests and numerical analysis were undertaken to investigate the specific storage of sedimentary rocks for supercritical CO₂. New laboratory system of constant flow pump permeability test was developed in order to measure permeability and specific storage of sedimentary rock in similar physical condition of deep aquifer where CO₂ tends to be in supercritical phase. To analyze experimental results, numerical analysis was established by extending the theoretical model of constant flow permeability to deal with two phase flow drainage displacement. For the examination of its applicability, experimental test was undertaken in which Ainoura sandstone used as core sample, injected with supercritical CO₂ under laboratory condition of 35°C constant reservoir temperature, 10 MPa initial hydraulic pressure, and 20 MPa constant overburden pressure. The experimental results show that, average specific storage of Ainoura sandstone is about 3.74×10^{-4} 1/Pa at a maximum CO₂ saturation of 0.55, and volumetric strain increases about 0.7%. The accuracy of specific storage measured was validated with ratio of the specific storage of the sandstone to the storage capacity of the pump used in the permeability test. This finding suggested that newly developed constant flow pump technique with its numerical analysis can be used to effectively obtain specific storage of sedimentary rock for CO₂ in supercritical phase.

INTRODUCTION

CO₂ sequestration in deep saline aquifers has become one of the most viable options to reduce the emission of anthropogenic greenhouse gases into the atmosphere. In Japan, deep sedimentary basins have been considered as suitable media for CO₂ sequestration since sedimentary formations comprise over half of Japan's total subsurface geology, with an estimated 146 billion Gt-CO₂ storage capacity (Ogawa et al. 2011; Nakanishi et al. 2009; Takahashi et al. 2009). However, prior to the commercialization of CO₂

sequestration projects in sedimentary basins, detailed studies were needed to understand the physics of water-CO₂ flow in this type of geological environment. In fact, CO₂-brine multiphase flow data is rather scarce compared to oil-water and CO₂-oil multiphase flow data generated for oil reservoirs over decades (Perrin and Benson 2010). Relative permeability and specific storage can be determined by two phase differential pressures and individual phase flow rates using Darcy's law. The injection ratio of the two-phase fluid is repeatedly alternated in order to determine the relative permeabilities at different saturation levels. A number of studies have been undertaken in measuring CO₂-water relative permeability and storativity in sedimentary rocks (Bennion and Bachu (2005), Perrin et al., (2009), and Shi et al. (2009). However, accurate, repeatable, and reliable relative permeability measurement with standardized and comparable laboratory experiments remains a research subject (Müller, 2011).

A standard experimental method used for permeability measurement is constant flow pump permeability test. This method is introduced by Olsen et al. (1985) as an improved version of the transient flow method introduced by Brace et al. (1968). The principle work of the flow pump permeability is to generate a constant rate of flow to precisely control pore fluid transport processes in a specimen. The flow of fluid through the specimen generates a hydraulic head that transiently increases and subsequently stabilizes to steady state with a constant head gradient imposed across the specimen. By using Darcy's law, the corresponding permeability value can be determined based on the steady state data collected. However, it would take several hours to reach steady state for the condition of a large specimen or a large pump system. Morin and Olsen (1987) developed a mathematical model for transient pressure response by considering the storage capacity of the specimen. This enables the determination of the permeability in early testing time. Later, Esaki et al. (1996) introduced the effect of storage capacity of the pump system to enhance the accuracy and efficiency of the mathematical model. Song et al. (2004) then introduced a more efficient and less tedious method to Esaki's et al. (1996) numerical simulation. In general, the constant flow pump permeability method is employed to measure the hydraulic conductivity and specific storage of rocks at laboratory conditions. In this study, we endeavor to develop laboratory system of the constant flow pump permeability method and its numerical analysis in order to measure permeability and specific storage of rock under the injection of CO₂ in supercritical phase. The validity and applicability of this technique are also included in this paper.

Experimental System

Figure 1 shows the schematic diagram of flow pump permeability test in the new developed experimental system. The new experimental system consists of several developed apparatus such as temperatures controllers, pressure controllers, and flow controllers. The aim of this instalment is to generate and stabilize high temperature and high pressure desired to create reservoir condition where CO₂ is in supercritical state. The main difficulty in creating a reservoir condition with high pressure and high

temperature is the vulnerability of the experimental system to the unstable temperature associated with seasonal weather and heat induced by the experiment apparatus. Such condition will affect the physical property of CO₂ leading to inaccuracy of experimental measurement. Therefore, the external and internal lab temperatures were attempted to control by constructing a greenhouse chamber and installing several devices and apparatus such as those are listed as follows: thermostatic room, greenhouse chamber, hemathermal circulation tank, and constant temperature water tank, temperature controller for pressure vessels, thermocouple and heater bars, remote control, and data acquisition system. Strain gauges were employed to measure deformation of rock specimen (Figure 2). The strain gauges were attached laterally and longitudinally on rock specimen.

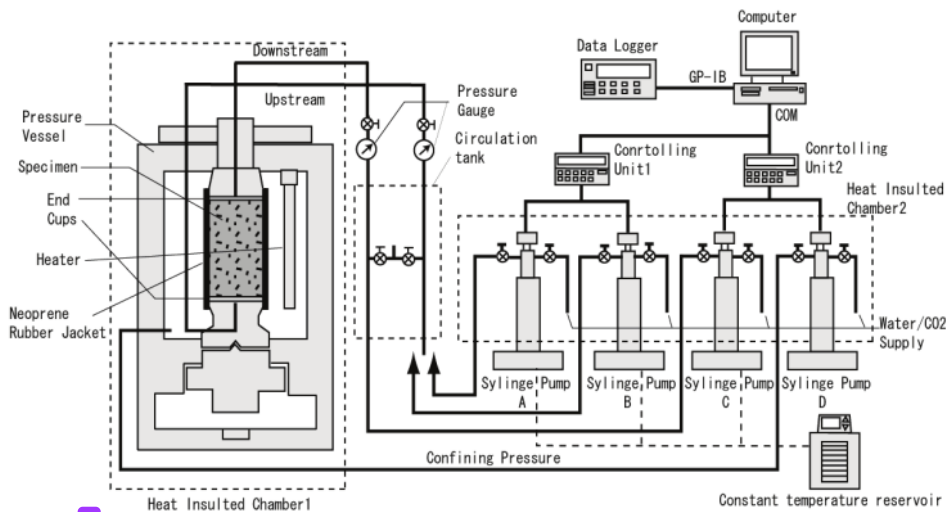


FIG. 1. Schematic diagram of constant flow pump permeability with newly developed laboratory system.

Rock specimens used in the experiments are Ainoura sandstones obtained from Sasebo, near Nagasaki Prefecture in Japan. The sandstone is a low permeable but high porosity rock, expected to be suitable for CO₂ storage. The pore size distribution of the Ainoura sandstone was investigated by using Pore Analyzer (Porosimeter) Micromeritics Auto pore III. Figure 3 presents the pore size distributions of the Ainoura sandstone specimens. It can be seen that two different Ainoura samples (Ainoura 1 and 2) exhibited a bi-modal pore size distribution, indicating heterogeneous pore siliciclastic characteristics. Due to the larger microporosity of Ainoura 1 than Ainoura 2 (Table 1), Ainoura 1 appeared to contain a much finer grain matrix than Ainoura 2. Figure 4 presents CO₂-water capillary pressure of Ainoura 1 and 2 specimens. By fitting the capillary pressure data with the Van Genuchten (1980) equation, the irreducible water saturation, S_{wr} , threshold capillary pressure, P_o , and pore size index, m .

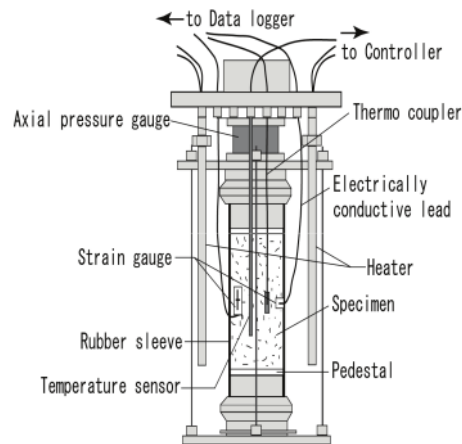


FIG. 2. Rock specimen with strain measurement devices and temperature controller.

Table 1. Pore Size Characteristics in the rock specimens

Specimen	% Micro-pores	% Mesopores	% Macropores	Median pore size (μm)	Porosity	IFT (mN/m)	P ₀ (kPa)
Ainoura 1	64.7	22.1	13.1	1	12.6	32.1	27
Ainoura 2	51.06	29.4	19.6	1.2	15.46	32.1	25

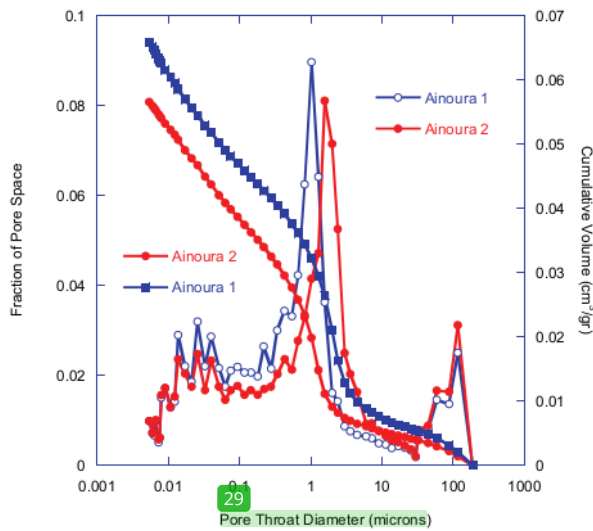


FIG. 3. Pore size distribution of Ainoura sandstone specimens.

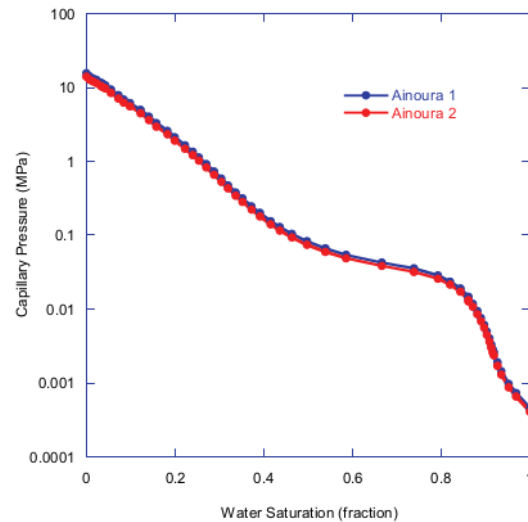


FIG. 4. Capillary pressure of Ainoura sandstone specimens.

Figure 5 illustrates the experimental system of the injection of supercritical CO₂ into the Ainoura sandstone by using the flow pump permeability test. To maintain the physical characteristic of CO₂ in the supercritical phase, the temperature of rock specimen, syringe pumps, syringe pipes and pressure vessels were maintained at 35°C, 36°C, and 38°C, respectively. After the temperatures stabilized, the confining pressure of 20 MPa and the pore pressure of 10 MPa were subjected on the specimen. The supercritical CO₂ within 3 μl/min flow rate was injected into the rock specimen and the pressures generated by the injection were measured continuously. The precision of the measurement can be seen from the record of the pressure in outlet and inlet of the rock specimen for every two seconds. Therefore, the pressure data of around 24 million during the experiment can be obtained. The measurement of the generated pressure was performed by employing the pressures of two gauges with resolution of 50 cmH₂O or 0.0049 MPa, manufactured by Research Institute Tokyo.

During the injection, the generated hydraulic pressures in the upstream and downstream of the Ainoura sandstone specimen including its volumetric strain were measured (Figures 6 and 7). The flow of the injected CO₂ generated an increase in pore pressure while the differential pressure was dropping. Meanwhile, the increase in the pore pressure propagated a deformation on the Ainoura sandstone specimen. The negative direction of the increasing strains indicates such an expansion of the Ainoura sandstone specimen.

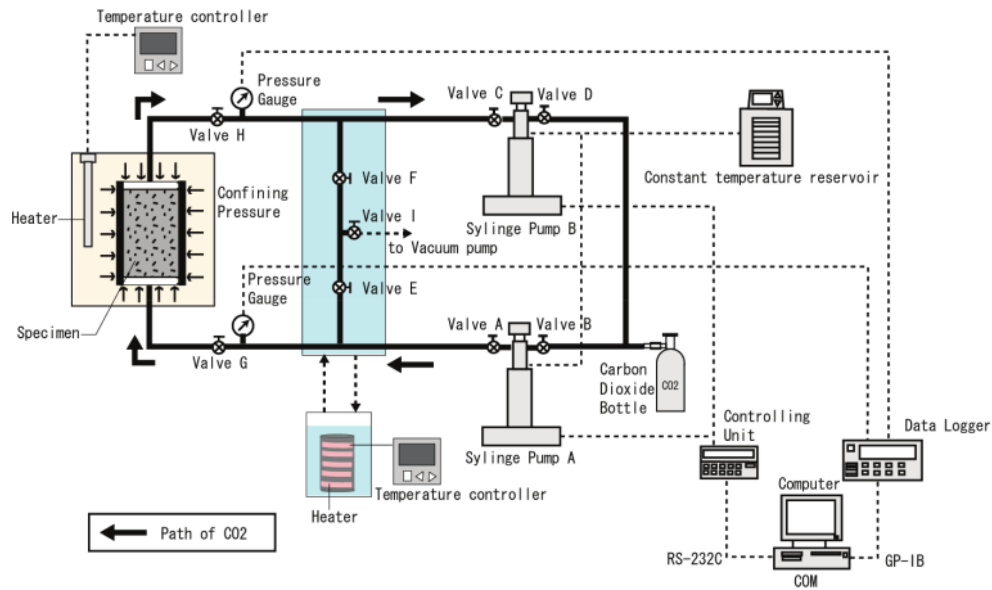
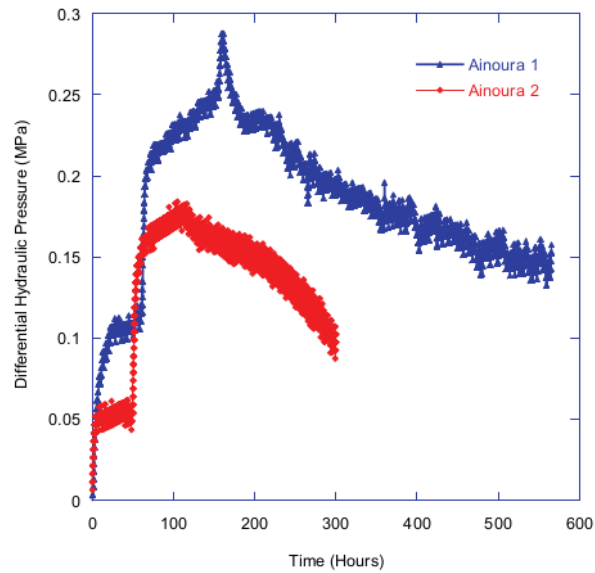


FIG. 5. Schematic diagram of experimental test of CO₂ injection.



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 FIG. 6. Measured differential pressure between upstream and downstream side of the rock specimen during CO₂ injection.

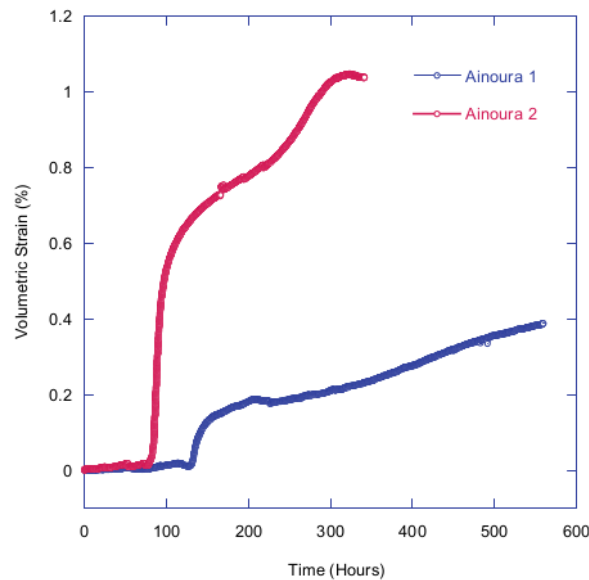


FIG. 7. Measured volumetric strain of the rock specimen propagated by CO₂ injection.

Theoretical model

The mathematical model of the flow pump permeability test is described as one-dimensional transient flow of a compressible fluid through a saturated porous and compressible medium. This model combines the principle of fluid mass in a deformable matrix and Darcy's law for laminar flow through a hydraulic isotropic rock matrix (Zhang et al. 2000). Since the experimental system of CO₂ injection into low permeable rock saturated with water can be described as a two-phase flow drainage displacement process, volumetric balance can be employed. It can be assumed that, after breakthrough, the flow rate of the injected fluid is equal to the total flow rate of the displaced and displacing fluid. Due to the compressibility of the displacing fluid (which is a liquid-like gas), the assumption was that the flow rate of the injected fluid was the total of the displaced fluid, and the displacing fluid, at time t minus volume, was absorbed within the compressible pump system per unit time interval. The system included the entire space of the flow pump cylinder, the space in the lower pedestal, and the tubing connecting the flow pump to the test cell. This assumption also led to change of the boundary conditions of the model shown in Eq. 4 and 4a. The schematic diagram and boundary conditions associated with the modified mathematical model are depicted in Figure 8.

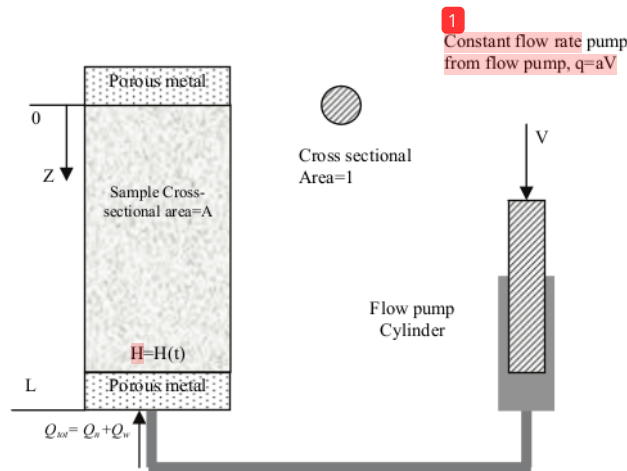


FIG. 8. Schematic diagram of the theoretical model of constant flow pump permeability test.

The governing equation is defined as follows:

$$\frac{\partial^2 H_n}{\partial z^2} - \frac{S_s}{k} \frac{\partial H_n}{\partial t} = 0 \tag{1}$$

Initial condition:

$$H(z,0) = 0 \quad 0 \leq z \leq L \tag{2}$$

Boundary conditions:

$$z = 0, \quad H(0,t) = 0 \quad t \geq 0 \tag{3}$$

$$z = L, \quad Q(t) = Q_n(t) + Q_w(t) \tag{4}$$

$$Q(t) = \left(\frac{k_{rn}}{\mu_n} \frac{dH_n}{dz} \rho_n + \frac{k_{rw}}{\mu_w} \frac{dH_w}{dz} \rho_w \right) gKA \tag{4a}$$

- where
- H = hydraulic pressure, MPa,
 - H_w = hydraulic pressure of water, MPa,
 - H_n = hydraulic pressure of CO₂, MPa,
 - k = hydraulic conductivity, cm/s,
 - z = vertical distance along the specimen, cm,

t	=	time from the start of the experiment, s,
S_s	=	specimen's specific storage, 1/Pa,
K	=	intrinsic permeability of the specimen, cm^2
k_{rw}	=	relative permeability of water, fraction,
k_{rn}	=	relative permeability of CO ₂ , fraction,
L	=	the length of the specimen, cm ,
μ_w	=	dynamic viscosity of water, $Pa.s$,
μ_{nw}	=	dynamic viscosity of CO ₂ , $Pa.s$,
ρ_w	=	density of water, gr/cm^3 ,
ρ_n	=	density of CO ₂ , gr/cm^3 ,
A	=	the cross-sectional area of the specimen, cm^2 ,
$Q(t)$	=	flow in the specimen at time t , cm^3/s ,
$Q_n(t)$	=	flow of CO ₂ in the specimen at time t , cm^3/s ,
$Q_w(t)$	=	flow of water in the specimen at time t , cm^3/s ,
g	=	gravity acceleration, cm/s^2

Since $\frac{dH_w}{dz} = \frac{dH_n}{dz} - \frac{dH_c}{dz}$, equation (4a) can be described as:

$$Q(t) = \left(\frac{k_{rn}}{\mu_n} \rho_n + \frac{k_{rw}}{\mu_w} \rho_w \right) \frac{dH_n}{dz} gKA - \frac{k_{rw}}{\mu_w} \rho_w \frac{dH_c}{dz} gKA \quad t > 0$$

Hence, the displacing non-wetting fluid pressure gradient becomes:

$$\frac{dH_n}{dz} = \frac{Q(t)}{\left(\frac{k_{rn}}{\mu_n} \rho_n + \frac{k_{rw}}{\mu_w} \rho_w \right) gKA} + \frac{\frac{k_{rw}}{\mu_w} \rho_w \frac{dH_c}{dz}}{\left(\frac{k_{rn}}{\mu_n} \rho_n + \frac{k_{rw}}{\mu_w} \rho_w \right)}$$

In which

$$\int_0^t Q(t) dt = \int_0^t q dt - C_e H_n(L, t)$$

$$\therefore Q(t) = q - C_e \frac{dH_n(L, t)}{dt}$$

Where

q	=	flow rate into the upstream of the specimen at time t , cm^3/s ,
C_e	=	storage capacity of the flow pump system, i.e., the change in volume of the permeating fluid in upstream permeating system per unit change in hydraulic head, cm^3/cmH_2O

The complete analytical solution is give as follows:

$$h(z,t) = \frac{qL + \left(gKA \frac{k_{rw}}{\mu_w} \rho_w L \frac{dH_c}{dz} \right)}{KA g \left(\frac{k_m}{\mu_n} \rho_n + \frac{k_{rw}}{\mu_w} \rho_w \right)} \left\{ \frac{z}{L} - 2 \sum_{n=1}^{\infty} \frac{\exp \left[- \frac{K \left(\frac{k_m}{\mu_n} \rho_n + \frac{k_{rw}}{\mu_w} \rho_w \right)}{S_s} \beta_n^2 t \right] \sin(\beta_n z)}{L \delta \beta_n \cos(\beta_n L) \left[L \left(\beta_n^2 + \frac{1}{\delta^2} \right) + \frac{1}{\delta} \right]} \right\} \quad (5)$$

Where $\delta = \frac{C_e}{AS_s}$, and β_n is the root of following equation:

$$\tan \phi = \frac{1}{\delta \beta^2} \left(k_{rw} + k_m \frac{\mu_w \rho_n}{\mu_n \rho_w} \right)$$

The roots were obtained using the Newton Raphson Method (Carslaw and Jaeger 1959).

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The hydraulic gradient distribution within the specimen can be further derived by differentiating Eq. (5) with respect to variable z:

$$i_n(z,t) = \frac{qL + \left(gKA \frac{k_{rw}}{\mu_w} \rho_w L \frac{dH_c}{dz} \right)}{KA g \left(\frac{k_m}{\mu_n} \rho_n + \frac{k_{rw}}{\mu_w} \rho_w \right)} \left\{ \frac{1}{L} - 2 \sum_{n=1}^{\infty} \frac{\exp \left[- \frac{K \left(\frac{k_m}{\mu_n} \rho_n + \frac{k_{rw}}{\mu_w} \rho_w \right)}{S_s} \beta_n^2 t \right] \cos(\beta_n z)}{L \delta \beta_n \cos(\beta_n L) \left[L \left(\beta_n^2 + \frac{1}{\delta^2} \right) + \frac{1}{\delta} \right]} \right\} \quad (6)$$

CO₂-water relative permeability

History curve matching between the pressure gradient from the experiment and the corresponding data of pressure gradient based on theoretical model was conducted. Once they become matched, the unknown parameters of k_m , k_{rw} , and S_s were determined. To examine their uncertainties, a sensitivity analysis of the parameters k_m and k_{rw} towards the differential pressure was undertaken. Pressure gradient is insensitive when the parameter k_{rw} was above 0.5 at all values of the parameter k_m . On the other hand, at all values of the parameter k_{rw} , pressure gradient is insensitive at the parameter k_m above 0.1. This means the parameter k_m is more sensitive than k_{rw} .

Figure 9 presents the CO₂-water relative permeability of the Ainoura specimens. As expected, the relative permeability of water decreased while that of CO₂ increased. Overall, CO₂ relative permeability at irreducible water saturation was observed to be lower than what we expected as it was about 15% of the water relative permeability at conditions of 100% water saturations. This indicated a lower displacement efficiency of the saturated water by the injected CO₂ in Ainoura Sandstones. The result may be attributed to heterogeneous bi-modal pore distribution of Ainoura Sandstones. Similarly, Bennion and Bachu (2005) observed that the pore size distribution of reservoir rocks critically affects CO₂-water relative permeabilities. In this kind of pore system, the CO₂-water displacement results in channel flow, yielding a non-uniform CO₂ flow in the system. All these imply that CO₂ flows preferentially in the smaller pores of the bi-modal pore system. Another factor that might be contributing to low relative permeability of CO₂ is the capillary pressure effect. By having a large fraction of micropores (more than 50%), the Ainoura specimens yielded relatively high capillary pressure. Therefore, its irreducible water saturation was high at about 45%. Capillary pressure acts as a barrier for CO₂ flow when very low injection rates applied.

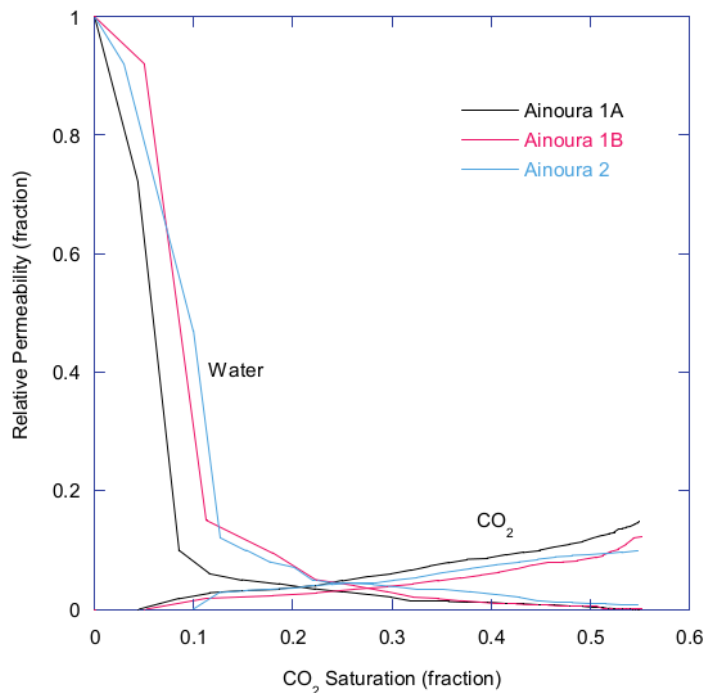


FIG. 9. End-point relative permeability of Ainoura sandstones versus CO₂ saturation.

Specific storage versus saturations

Figure 10 presents the change of specific storage of the Ainoura specimens with increasing CO₂ saturation. It was observed that the injection of CO₂ increased the specific storage of the specimen. Such a transient increase of specific storage beyond certain saturation was found at the same period of the significant increase of volumetric strain of the specimen. This suggested that the change of specific storage beyond certain CO₂ saturation is more pronounced as a mechanical response rather than just a hydraulic process. The specific storage increased by about 0.0004, 0.0003, and 0.0005 1/Pa for Ainoura 1A, 1B and 2, respectively.

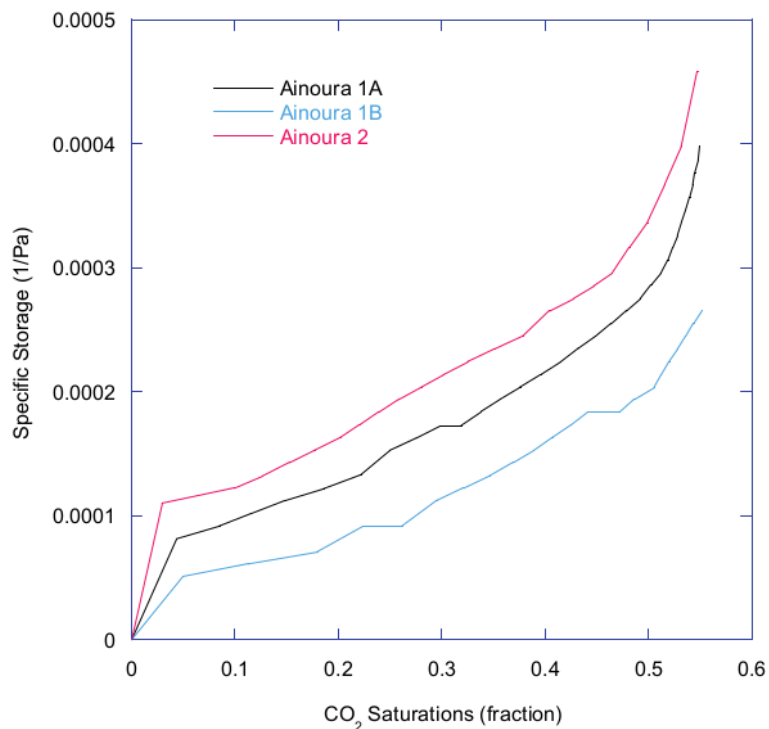


FIG. 10. Specific storage versus CO₂ saturation in Ainoura sandstone.

The validity of the specific storage obtained from the numerical analysis was also examined. The accuracy of specific storage of rock specimen is much affected by the storage capacity of the pump system. Tominaga and Kameya (2003) introduced a dimensionless parameter, δ' , described as the ratio of the pump's storage capacity to the specimen's specific storage:

$$\delta' = \frac{\delta}{L} = \frac{C_e}{S_s AL} \quad (7)$$

They suggested that the parameter S_s would have sufficient accuracy if the ratio, δ , is less than 0.3. In the numerical analysis, the ratio of the obtained S_s to the measured C_e was found to be between 0.00078 and 0.0187 (Figure 11). This clearly shows the ratios are below the ambient ratio of 0.3. However, the ambient ratio to examine accuracy of specific storage may be insufficient to implement in a transient flow from CO₂-water drainage displacement system in reservoir condition. Further sensitivity analysis and poro-elasticity measurement to examine the accuracy of specific storage will be a subject of future investigation. Nonetheless, as preliminary validation to the numerical analysis, the use of ambient ratio of Tokunaga and Kameya (2003) was found acceptable.

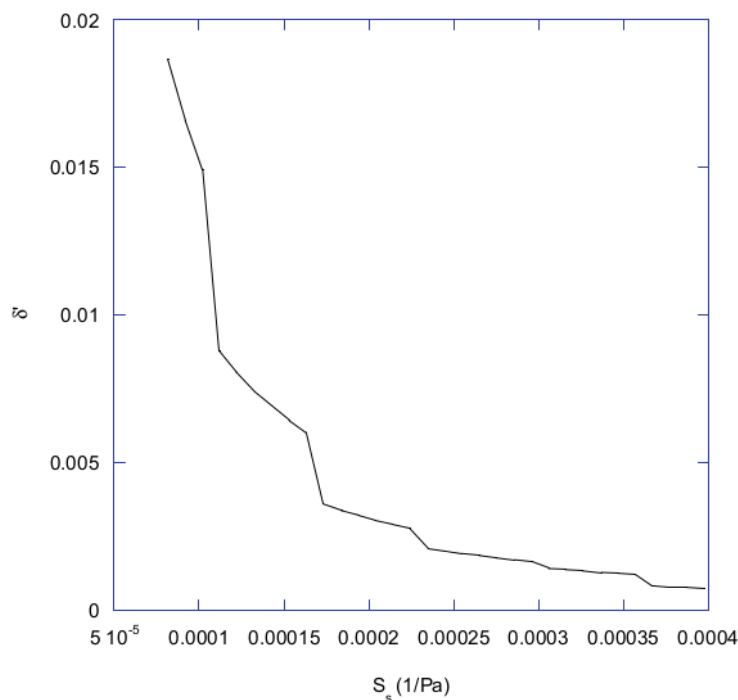


FIG. 11. Ratio of pump storage capacity to the Ainoura sandstone’s storage capacity.

Change of Bulk Compressibility of Ainoura Sandstones during CO₂ injection

Since the Ainoura sandstone specimen is a porous body subjected to internal pore pressure and external confining pressure, its bulk compressibility of the specimen can be determined based on matrix compressibility and the increased pore compressibility during the injection of CO₂ injection. The matrix compressibility (C_m) of the specimens is about $2.54 \times 10^{-5}/\text{MPa}$ for typical sandstone (Zimmerman, 1991). Figure 12 presents the transient increase of bulk compressibility occurs at the beginning of

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CO₂ injection. This corresponded to the transition from the displaced incompressible water flow to the displacing compressible CO₂, in the specimen pores. After this period, overall, bulk compressibility of the specimen decreased with increasing pore pressure. Above a certain pressure, the bulk compressibility reached a plateau that is independent of the pore pressure. Figure 12 also shows the tested Ainoura 2 has larger bulk compressibility than the tested Ainoura 1. This is due to higher fraction of macropores in the Ainoura 2 resulted in more flow of CO₂, leading to higher pore pressure generated.

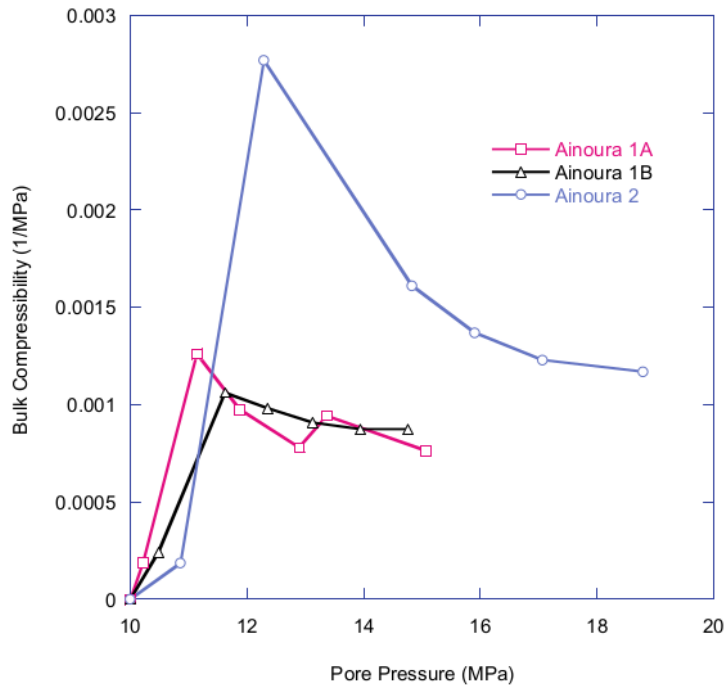


FIG. 12. Bulk compressibility changes as CO₂ injected into the rock specimens.

Effect of Pressure Margin on Volumetric Strain

The injection of CO₂ into the rock specimen increased its pore pressure and volumetric strain. As the experiments were constantly set at 20 MPa confining pressure, only the pore pressure increased from the 10 MPa initial pressure. If the pressure margin is defined as the gap pressure of the pore pressure to the confining pressure, the pressure margin decreased during the injection. The pressure margin was analysed in this study since it is a considerable parameter that might cause hydraulic fracturing. The initiation of hydraulic fracturing will occur when the pore pressure equals the confining pressure (Jaeger et al., 2007).

Figure 13 illustrates the relationship between the pressure margin and the volumetric strains measured during the experiments. As seen, the pressure margin increased as the volumetric strain increased. Beyond a certain pressure margin, the volumetric strains increased significantly. The transient increase of volumetric strain occurs at the transition of the incompressible water flow to the compressible CO₂ flow in the specimen pores, as observed in the second phase of the experiment. After that, the CO₂ did breakthrough the specimen, generating a higher increase of the volumetric strain. Given the trend of curves in Figure 13, the flow of CO₂ would generate a significant increase in volumetric strain when the pressure margins are above -9 MPa and -8 MPa for the Ainoura 2 and 1 samples, respectively. This means that the increased volumetric strain of the higher porosity specimen would occur slower than that of the lower porosity specimen. However, in the case of the magnitude of the strains generated, the specimen with higher porosity yielded a larger volumetric strain compared to a lower porosity sample. As a result, the generated pore pressure in the higher porosity specimen took a shorter time to reach the confining pressure level. The results suggested the benefit of lower porosity Ainoura sandstones in which they would have a higher specific storage for CO₂ but would generate lower deformation. It is noted that the lower deformation observed was induced by the injection at the very low flow rate applied in the experimental test. The very low flow rate was selected to mimic laminar flow in deep underground.

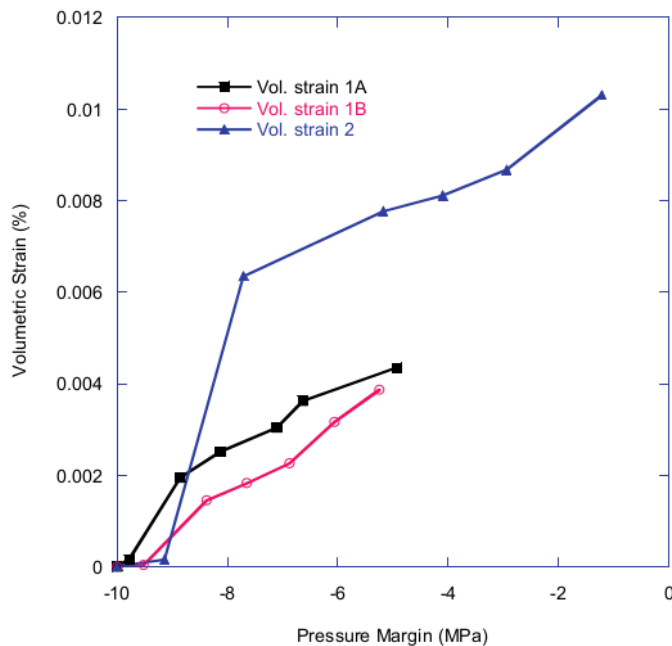


FIG. 13. Pressure margin of pore pressure to confining pressure versus volumetric of the specimens.

CONCLUSIONS

This paper presented experimental and numerical analysis to measure permeability and specific storage of sedimentary rock under the injection of CO₂ in supercritical phase. Conclusions from this study are as follows:

- Ainoura sandstone has lower CO₂-water displacement efficiency indicated by low CO₂ relative permeability (only 15% of the relative permeability of water at 100% water saturation).
- The average capacity of Ainoura sandstones for storing supercritical CO₂ is estimated to be 3.74×10^{-4} 1/Pa within the experimental conditions applied.
- Ainoura sandstone appears to be effective in retaining the flow of supercritical CO₂ as it takes considerably long time to migrate through the sandstone.
- The injection of CO₂ into the Ainoura sandstones has resulted in the increase of volumetric strains of the sandstones. Given by the direction of strains, the sandstones appear to be expanded during the injection;
- The expansion of the Ainoura sandstones is due to the decrease of effective pressure as the pore pressure induced by the injection increases and the confining pressure is set to be constant. The expansion initiates when the pressure margin between the pore pressure and the confining pressure is -9 MPa and -8 MPa for the Ainoura 1 and 2, respectively;

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