Evaluation of In-situ Reservoir Blocking by Sodium Carbonate Gel Formed from Sodium Metasilicate Solution and Injected CO₂ for CO₂ Sequestration

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Preventing channeling flows during enhanced oil recovery targeting heterogeneous or fracture type reservoirs and leakage flows from saline aquifers containing CO₂ remains a challenge. This study evaluated the potential of in-situ gelation as a blocking agent in a heterogeneous reservoir using the reaction between aqueous solution of sodium metasilicate (Na₂SiO₃·9H₂O; S-MS) and dissolved carbon dioxide (CO₂). Both Raman and scanning electron microscopy/energy dispersive X-ray (SEM-EDS) spectroscopy revealed that the gel was a sodium carbonate type (S-C-gel). Physical characterization of the S-C-gel including the gelation time, gel strength and stability, were investigated in respect of S-MS concentration, temperature, salinity (NaCl), divalent ion concentration (calcium, Ca²⁺) as well as CO₂ injection pressure. Gelation time after CO₂ gas injection was around 1 to 24 h depending on temperature and pressure. Gel strength increased with higher S-MS concentration (≤ 10 wt%) and CO₂ gas pressure (≤ 5.5 MPa). Threshold pressure gradient (TPG) and gas permeability of the sandstone core filled with in-situ gel increased by 2.6 times and decreased about 1/10, respectively, compared with the water saturated core. These promising findings herein could be extended to CO₂ sequestration.

Keywords
Heterogeneous reservoir, In-situ blocking, Sodium carbonate gel, Sodium metasilicate solution, Carbon dioxide sequestration

1. Introduction

A heterogeneous reservoir is defined as a reservoir consisting of thief zones, high permeability layers and/or fractures. These features result in one of major problems during water-flooding recovery processes as far as oil recovery is concerned, because the injected water is passed through the high permeability zone (short-cut zone) without sweeping the low permeability zone. On the other hand, carbon dioxide (CO₂) leakage through the low sealing potential of fractures and high fault zone remains the main problem for carbon capture and storage (CCS) in underground geological formations. Consequently, both these processes require conformance improvement treatments, given the substantial volume of trapped oil within the low permeability zones or the need for sealing agent, to prevent CO₂ gas leakage from the underground reservoir. Conformance improvement, also referred to as water shutoff, can be divided into mechanical and chemical methods. The mechanical approach relies on horizontal and multi-lateral wells to increase the contact reservoir zone, or liner and cement squeezes to block short-cut flow. Such mechanical techniques often fail because water leaks past the packing devices to the outside of the plugging zone. The chemical approach, as considering in this study, uses chemical solutions to form gel in the high permeability zones. Several methods have been investigated with various results, including phenol-formaldehyde resins, chemical precipitation, inorganic gels (sodium silicate gels), and gelling polymer. Study of the cross-linked polymer for water diversion developed two methods to evaluate gel characteristics including beaker tests and gel evaluation. Both gelation time, defined as the time at which viscosity starts to increase, and the viscosity of the polymer are the main influencing parameters. Furthermore, water shut-off treatment using polymer as starting material is severely challenged by any heterogeneity of the candidate formation. Therefore, alternative approaches consisting of blending polymer with silica gel have been proposed. However, the detailed mechanisms and chemical analyses of this silicate gel are poorly understood. Previous studies suggest that formation of the gel requires a foreign material (gelling activator) which is not carbon dioxide (CO₂). Sodium metasilicate (S-MS) are commonly used as powder detergents and industrial applications.
contain an optimum portion of alkali and soluble silica, so is a suitable gel precursor\textsuperscript{10}. S-MS solution has viscosity similar to that of water, so is suitable for injection into an oil-bearing matrix, with formation areas located far from the injector\textsuperscript{11}. In addition, the solution must be thermally stable in high temperature reservoirs, and gelation time must be controllable under the same conditions\textsuperscript{12}. Furthermore, S-MS is inorganic and does not present hazards such as low flash or flammability. The alkali content the S-MS modifies interfacial tension of the trapped oil, so S-MS solution has a long history as an alkaline flooding agent for light/medium/heavy oil formations\textsuperscript{13}.

Using S-MS solution for conformance has been extensively studied. The silica gel is formed as the S-MS solution is acidified to a pH value of less than 10, but controlling the gelation time remains challenging\textsuperscript{14}. In the field, formation of S-MS silicate gels can be achieved using precursors or activators such as organic acid, urea, or multivalent cation\textsuperscript{15}. However, these precursors tend to lower the acidity of the solution and so promote the formation of the gel. Various potential gelling activators including CO\textsubscript{2}, hydrogen chloride (HCl) and amides were reviewed\textsuperscript{16}. CO\textsubscript{2} is easy to handle and adjust, and can be injected into a well and spread in a reservoir due to the higher relative permeability than liquid fluids. Moreover, using captured CO\textsubscript{2} as the precursor is also an alternative approach for carbon sequestration.

These features of S-MS solution suggest dual purpose uses of blocking of the channeling passes in the reservoir and as chemical agent for alkaline flooding (Fig. 1).

Both these functions could subsequently enhance oil recovery given that S-MS solution remaining after alkaline flooding could become the starting material for channel plugging. In theory, S-MS solution is injected as the alkaline flooding agent into the formation. The oil is produced from the high permeability zones. After water breakthrough, CO\textsubscript{2} is injected and the reservoir is shut-in. Subsequent in-situ gelation would plug short-cut passes, reducing the permeability. As a result, sweep efficiency, and thus oil recovery, is improved.

The present study aims at preparing, characterizing and evaluating the potential of in-situ gel as plugging/binding agent in heterogeneous reservoirs. Aqueous S-MS solution is used as the starting material and CO\textsubscript{2} as precursor. Both physical and chemical analyzes were considered to investigate the mechanisms of gel formation. Permeability tests were performed to validate the in-situ gel formation in porous media.

### 2. Materials and Measurement Methods of Gel Properties

#### 2.1. Materials

A lyophilized powder of sodium metasilicate nonahydrate (Na\textsubscript{2}SiO\textsubscript{3}·9H\textsubscript{2}O, S-MS), supplied by Junsei Chemical Co., Ltd. (Japan), was used as the starting material. CO\textsubscript{2} gas, 99.9 % pure, was used as precursor. To investigate the effects of salinity and the water hardness on gel formation, solutions of sodium chloride (NaCl) and calcium chloride (CaCl\textsubscript{2}) at different concentrations were used. Both salts were purchased from Junsei Chemical Co., Ltd. (Japan) and used as received.

#### 2.2. Gel Formation

Six S-MS solutions with concentrations ranging from 1 to 10 wt% were prepared using distilled water. The physio-chemical properties of each solution including pH, density and viscosity were measured using a pH meter (AS800), pycnometer and viscometer (Brookfield DV-I Prime) at room temperature (25 °C). Gel formation was conducted at 25 °C using 45 mL of S-MS solution (1-10 wt%) with various shut-in times (up to 6 h) and CO\textsubscript{2} pressures (2.0, 4.0 and 5.5 MPa) in a high pressure cell (Fig. 2).

Gel samples formed in the cell at each shut-in time, CO\textsubscript{2} pressure, and S-MS concentration was taken im-
Immediately after depressurization, and the gel properties such as pH, apparent viscosity, and gel strength were measured. Subsequently, beaker testing was carried out to screen and classify the gel sample based on its properties.

2.3. Measurement Methods of Gel Properties

2.3.1. Chemical Characterization

Chemical characterization used scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM-EDS) to investigate the surface morphology of the gels and determine the elemental composition of the gel, and Raman spectroscopy to confirm the chemical structure of the gel formed.

Prior to analysis, the gel samples were dried at 110 °C for 24 h. SEM photographs were taken with a low vacuum high sensitive scanning electron microscope (Hitachi, Ltd., SU 3500). Raman spectra were obtained by microscopic laser Raman spectroscopy (LabRAM ARAMIS, HORIBA, Ltd., Japan) applying the 532 nm line of an Ar laser.

2.3.2. Physical Characterization

Gel characterization, consisting of gel viscosity, gelation time, gel strength and gel stability, was conducted by changing the S-MS solution concentration, CO₂ gas pressure, and temperature.

Gel viscosity: The rheological properties were measured using a viscometer (Brookfield Viscometer DV-I) with shear rate from 5 to 100 RPM. The apparent viscosity was then plotted against the elapsed time (viscosity-time curve) for gelation time determination.

Gelation time: Gelation time, defined as the time at which the apparent viscosity deviated, was determined from the intercept of the extrapolations from the two straight line sections of the viscosity-time curve. Gelation time was determined from the six solutions from which the hardest gels were obtained and the thermal effects on the gel properties were investigated at 25, 40, and 55 °C. The gelation time of the hardest gel was also evaluated in glass beads as porous media at room temperature, 25 °C (Beaker tests, Fig. 3).

Gel strength: The gel strength was evaluated from the gel strength code previously proposed based on visual observation as shown in Table 1. In the present study, flow behavior of the gel was observed by flipping 3 mL gel in a 10 mL glass tube.

Gel stability: This study mainly focused on the thermal stability by visual observation of the gel remaining ratio of the samples under constant temperatures (25 to 80 °C). The gel stability was then defined as the ratio (gel remaining ratio, %) of the remaining gel volume after the elapsed time divided by the total initial volume. The tests were carried out by continuous monitoring of sampled gel (10 mL) in the glass pipette during 1 month at 25, 55 and 80 °C.

2.3.3. Evaluation of In-situ Gel as a Blocking Agent

The gas permeability test was conducted by forming the gel in-situ in a Berea sandstone core fully saturated with water (Fig. 4).

S-MS solution 10 wt% was injected into a dried Berea sandstone core sample (length = 4.8 cm, diameter = 2.6 cm, permeability = 0.12 μm²), then in-situ gel was formed after CO₂ injection.

The flow rate and pressure drop were measured to evaluate the permeability of CO₂ gas from the measurement results using air, because this measurement was performed at almost atmospheric pressure so correction

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Table 1 Bottle Test-gel Strength Code

| Type | Definition |
|------|------------|
| 'A'  | No detectable gel formed |
| 'B'  | Highly flowing gel |
| 'C'  | Flowing gel |
| 'D'  | Moderately flowing gel |
| 'E'  | Barely flowing gel |
| 'F'  | Highly deformable non-flowing gel |
| 'G'  | Moderately deformable non-flowing gel |
| 'H'  | Slightly deformable non-flowing gel |
| 'I'  | Rigid gel |
| 'J'  | Ringing rigid gel |

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Fig. 3 Gel Formation Test in Porous Media

Fig. 4 Schematic Evaluation of Gas Permeability in Berea Sandstone Cores
for gas compressibility was not required.

3. Results and Discussion

3.1. Screening of the Baseline Concentration

Monitoring of the increase in acidity in the cell found that the gels are formed subsequent to pH reduction as shown in Fig. 5. This increase in acidity occurred in the primary solution and suggests the threshold concentration.

Dissolution of CO₂ lowers the pH from the initial pH of 13 to less than 5.9 regardless of the injection pressure. The acidity within the solution decreased slightly with S-MS concentration to an order of CO₂ gas injection pressure. Furthermore, the formed gel was weak at a concentration lower than 5 wt%, at which the pH was around 6, plausibly because of the low concentration of S-MS.

This observation contrasted with the finding at higher concentration (above 5 wt%) at which the pH ranged from 7 to 9.5. A similar result has previously been reported14). These results also show that the gel hardness increased with higher CO₂ injection pressure and S-MS concentration. Therefore, the gels prepared from 5, 8, and 10 wt% of Na-metasilicate concentration were selected for further analyses.

3.2. Spectral Characterization of Gel

SEM analysis, which shows the morphology of the gel, revealed a soft texture at low S-MS concentration (Fig. 6(a)). In contrast, the gel structure appeared either less loose (Fig. 6(b)) or coarser (Fig. 6(c)) at higher concentrations, suggesting stronger molecular bonding. Further information was conveyed by the Raman spectra of the samples (Fig. 7).

Characteristic bands (at 703, 1083 and 1424 cm⁻¹) were observed, all of which were more pronounced with higher concentration of S-MS. Peaks at 703 cm⁻¹ and 1082 cm⁻¹ were associated with the symmetric stretching of carbonate ions (CO₃²⁻), and the peak at 1424 cm⁻¹ was associated with the asymmetric stretching of the carbonate18),19). These assignments suggested that the gel formed was a carbonated rather than siliceous. Comparing the Raman spectra of the formed gels with commercial sodium carbonate (Na₂CO₃) found the peaks appeared at the same wavelengths.

Therefore, the gel formation was probably triggered by aqueous dissociation of S-MS and formation of silicates as shown in Eqs. (1), (2), and (3),

\[
Na_2SiO_3 + 9H_2O \rightarrow 2Na^+ + SiO_3^{2-} + 9H_2O \quad (1)
\]

![Fig. 5 pH Monitoring during Gel Formation](image)

![Fig. 6 SEM Images Obtained from S-MS Solutions and CO₂ at Different Concentrations: (a) 5 wt%, (b) 8 wt%, (c) 10 wt%](image)

![Fig. 7 Raman Spectra of Gel Samples](image)
Equation (3) explains why crystalline silica (SiO\(_2\)) was not detected in the Raman spectra. Silica was instead converted to silicic acid. With the injection of CO\(_2\), the gas dissolves in the aqueous solution to yields carbonates,

\[
\text{CO}_{2(g)} \rightarrow \text{CO}_{2(aq)} \quad (4)
\]

\[
\text{CO}_{2(aq)} + \text{H}_2\text{O} \rightarrow \text{CO}_3^{2-} + 2\text{H}^+ \quad (5)
\]

The carbonates are plausibly responsible for increasing the acidity in the initial S-MS solution (Fig. 5). As the concentration of CO\(_2\) increases within the solution, with higher concentration of CO\(_3^{2-}\) ions, the carbonated gel (S-C-gel) is formed following the overall reaction,

\[
(2\text{Na}^+ + \text{SiO}_3^{2-} + 2\text{H}^+ + 9\text{H}_2\text{O}) + \text{CO}_2(aq) + \text{H}_2\text{O} \\
\rightarrow (2\text{Na}^+, \text{CO}_3^{2-}) + (4\text{H}^+, \text{SiO}_3^{2-}) + 9\text{H}_2\text{O} \quad (6)
\]

This reaction pathway was confirmed by EDS analysis (Fig. 8) which revealed that S-C-gel predominantly contained carbon (C, originating from CO\(_2\)), sodium (Na, from S-MS), and oxygen (O from both CO\(_2\) and S-MS). The concentration in silicon (Si) was decreased based on the chemistry leading to gel formation (Eq. (3)), so the decrease in Si is plausibly explained by Eq. (2). In fact, we believe that formation in silicate (SiO\(_3^{2-}\)) is reversible and could be altered by the concentration within the mother liquor (S-MS).

### 3.3 Physical Properties of Gels

#### 3.3.1 Gel Formation Time

Figure 9 shows the viscosity-time curves after CO\(_2\) injection for three selected solutions of S-MS.

Gelation time at constant CO\(_2\) gas pressure of 2 MPa was 1 to 4 h depending on the initial concentration of S-MS. Longer gelation time was required for the lower concentration (≤ 5 wt%) because of the lower reaction rate. Therefore, gelation time decreased at higher concentration. On the other hand, the S-C-gel became more viscous at higher concentration of S-MS solution. For example, the gel viscosities formed with 5 wt% and 10 wt% S-MS solution were about 40 mPa s and 180 mPa s, respectively (see Fig. 9).

Increasing the temperature from 25 to 55 °C, with same 10 wt% of S-MS solution under 5.5 MPa of CO\(_2\) injection pressure, revealed delay in gelation time up to 1 day at 40 °C and 55 °C compared to around 1 h at 25 °C with abrupt decrease of apparent viscosity (Fig. 10).

This result contrasted with previous findings that higher temperature resulted in significant decrease in the gelation time\(^{(1,5)}\). These studies also found a different type of gel forming. CO\(_2\) solubility is normally decreased at higher temperature\(^{(20)}\). CO\(_2\) solubility decreases with lower concentration of CO\(_3^{2-}\) ions, so the S-C-gel formation time becomes longer (Eqs. (4), (5), and (6)). Delay in gelation time will promote gel formation and replacement with formation water at longer distances from the injector in a deeper zone with high permeability. Nevertheless, clearly both gelation time and apparent viscosity are sensitive to temperature. Therefore, reservoir temperature will be an important operation parameter for application of in-situ gel formation.
Gelation time was also measured in the porous media consisting of glass beads 1–2 mm (average 1.5 mm) in diameter and saturated with 10 wt% of S-MS solution at room temperature of 25 °C. CO2 gas was injected and the pressure within the cell was maintained at 5.5 MPa (Fig. 11).

S-C-gel was generated in the pore spaces between grains and bonded the glass beads together (Fig. 12). As the S-C-gel was formed, the solution color became milky unlike water and the initial solution. The gelation time was 12 h, much longer than that of the solution only, because of the limited CO2 diffusion rate in the porous-media.

3.3.2. Rheological Properties of In-situ Gel

Figure 13 shows the relationship between apparent viscosity and shear rate of the S-C-gel samples generated from three solutions with S-MS concentrations of 5, 8, and 10 wt% under CO2 injection pressure of 5.5 MPa.

S-C-gel appeared more viscous, with the rheological properties of non-newtonian fluid. Three gels exhibited shear thinning behavior of a pseudo plastic fluid similar to that of polymer solutions. The yield stress was evaluated to be 0.5 to 1.0 Pa. The apparent viscosity of the polymer solution decreases as shear rate increases because the polymer molecules can align with the shear field to reduce the internal friction1).

3.3.3. Gel Strength

Figure 14 shows the effects of S-MS concentration and CO2 gas pressure on the S-C-gel strength measured based on the gel strength code (Table 1).

The gel samples formed using 5, 8 and 10 wt% of S-MS at 25 °C could be classified (B) to (J) based on the ease of flow. Highly flowing gel (B) and non-flowing gel or strong gel (H) were formed from 5 wt% and 10 wt% solutions, respectively. The gel strength increased with higher S-MS concentration and CO2 gas injection pressure. S-C-gel strength can be expected to become stronger and more irreversible than under supercritical CO2 gas injection, because the higher CO2 solubility enhances gel formation in the solution. The gel was formed by chemical reactions that do not cause large changes in gel properties by depressurization. The viscosity of the gel is also not so sensitive to the pressure range (less than 5.5 MPa in this study), as the
drilling fluid viscosity is not sensitive to pressure. Even if depressurization does affect the gel property, our measurements show that the properties of the irreversible gel are formed chemically and remain stable against pressure change.

All S_C-gel samples formed from 10 wt% concentration were code “F” (high strength), whereas the S_C-gels formed from 5 wt% of S_MS solution were code “B” or “E”. Therefore, 10 wt% S_MS solution was selected for in-situ gel formation at 40 °C and 55 °C with CO2 gas injection pressure of 5.5 MPa to characterize the effects of temperature on the gel properties. The gel strength was reduced from high strength “F” at 25 °C to low strength “D” at 40 °C and 50 °C with low apparent viscosity of 50 to 80 mPa s.

3. 3. 4. Gel Stability

Figure 15 shows the results of remaining ratio (remaining gel volume %) versus time up to 35 days.

The remaining ratio-time curves decreased sharply before 200 h, especially in the cases of low S_MS concentration and high temperature. However, the stability was improved with the remaining ratio over 55 % after 200 h, even at a higher temperature (80 °C). The remaining ratio decreased with lower solution concentration. The remaining ratio of S_C-gel increased with lower temperature (Fig. 15).

3. 4. Parameters Influencing Gel Formation

3. 4. 1. Effect of Salinity

Different concentrations of sodium chloride (0.5-10 wt%) were added into Na-metasilicate to evaluate the effect on the gels (Fig. 16).

The pH of S_MS solution with 0.5 wt% concentration of NaCl was around 13 and slightly decreased with higher salinity. No precipitation was observed after adding the salt (0.5 to 10 wt%) to the S_MS solution. However, both gel strength and viscosity were decreased presumably due to the common ion effect or salting out effect. Gel viscosity is decreased with salinity higher than 3.5 wt% as shown in Fig. 16.

3. 4. 2. Effect of Divalent Ions

Different concentrations of divalent ion calcium Ca2+ (0.09 to 90.11 mmol/L) were added to the S_MS solution.

Precipitation was clearly observed by adding the calcium divalent ion as shown in Fig. 17. This precipitation was probably caused by the formation of calcium silicates.
CaCl₂(s) → Ca²⁺ + 2Cl⁻  \hspace{1cm} (7)

Ca²⁺ + SiO₃²⁻ + SiO₄⁴⁻ → CaSiO₃ + Ca₄(SiO₃)₂₂⁻ \hspace{1cm} (8)

S-MS solutions including Ca²⁺ at concentrations of 0.09 to 90.11 mmol/L were used for gel property measurements. Figure 18 shows the effect of Ca²⁺ concentration on gel viscosity and gel strength. Precipitation was increased with higher Ca²⁺ concentration, and intensively from 45 to 90.11 mmol/L with viscosity of 6 mPa s.

Based on these results, the S-MS solution is sensitive to hard brine containing divalent ions (Ca²⁺, Mg²⁺), so large preflush volume of soft water needs to be injected.

3.5. Blocking Performance of In-situ S-C-gel

The blocking performance of in-situ gel, formed from 10 wt% of S-MS solute under CO₂ gas injection, was measured by the core permeable gas flow test. Figure 19 shows the measurements of air flow rate versus pressure drop, Δp/L for dry core, core saturated with water and core including the in-situ formed S-C-gel.

The air flow rate increased linearly with the pressure gradient in dried core, whereas air flow started at the pressure gradient of 4 kPa/cm in water saturated core, which is called the threshold pressure gradient (TPG) as defined in Figure 4. The TPG increases with lower permeability and smaller pores and throats\(^{23,24}\). Investigation of the effect of TPG on gas flow in the tight sandstone gas reservoir found that the biggest pores and throats of cores start to contribute to gas flow at TPG, similar to the physical phenomenon of entry pressure, and more pores and throats contribute to the flow forming a concave curve by increasing the pressure gradient\(^{23}\). After the concave curve, the increasing ratio of airflow rate against the pressure gradient became constant, and the extrapolation line intersects the horizontal axis (flow rate \( \Delta p \)) at a pressure gradient of 5.8 kPa/cm, which is called as the pseudo threshold pressure gradient (PTPG) as shown in Figure 19\(^{22}\).

After the pressure gradient exceeds the PTPG, the number of the pores and throats become stable in the core, so the curve becomes a straight line. In the case of the core including formed in-situ gel, air flow starts approximately from 11 kPa/cm of TPG that is almost 2.6 times of that of the solution. These results show that in-situ gel formed in sandstone pores has effective blocking performance compared with water.

Figure 19 also shows the effect of in-situ S-C-gel on permeability change in the Berea sandstone core. The pressure gradient is proportional to the gas permeability after breaking air flow. Therefore, the gas permeability of the core filled with in-situ S-C-gel is almost 1/80 and 1/10 of dry and water-filled cores, respectively, as shown in Table 2.
These findings of the threshold pressure gradient and permeability show that in-situ S-C-gel can reduce reservoir permeability by the blocking performance.

4. Conclusions

The properties of in-situ gel, expected to be a cost-effective and environmentally safe blocking material, such as gelation time, gel strength and gel stability, were characterized and evaluated using various sodium metasilicate (Na2SiO3·9H2O, S-MS) concentrations, CO2 gas pressures, temperatures, salinity or NaCl wt%, and divalent ion (Ca²⁺) concentrations. Results can be summarized as follows:

a) Gel formed from S-MS and CO2 gas is sodium carbonate gel (S-C-gel) based on Raman spectroscopy and SEM-EDS results.

b) Gelation time was decreased with higher S-MS concentration and CO2 gas pressure, and by higher temperature. Gel strength and stability were also increased with higher S-MS concentration and CO2 gas pressure.

c) Gel strength and stability were affected by temperature, salinity (NaCl wt%) and divalent ion (Ca²⁺) concentration. 10 wt% S-MS concentration and CO2 gas pressure (> 4 MPa) generated the most stable gel with high gel strength (> “E”) for temperature (< 55 °C) and presence of salt/divalent ion (< 4.91 mmol/L).

d) Presence of high salt/divalent ion concentration reduced gel strength and gel stability, so preflush of reservoir water is required for applying this method.

e) Threshold pressure gradient and permeability of the core saturated by the S-C-gel was 2.6 times higher and about 1/10 lower, respectively, compared with the core saturated by water. These core testing results indicated that in-situ S-C-gel formation showed good performance as a blocking agent.

f) In-situ S-C-gel formation can be a potential blocking agent to stop short-cut paths to improve oil recovery and also CO2 sequestration, and suggests a new process of the EOR and CO2 sequestration for oil reservoirs.

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要　旨
CO₂固定化を考慮したメタケイ酸ナトリウム水溶液とCO₂圧入による炭酸ナトリウムゲル原位置生成による
貯留層ブロッキングの評価

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不均一あるいはき裂卓越型の貯留層における水攻法適用後の
チャネリングフローやCO₂が貯留されている帯水層からの漏
えい液の防止は，採油増進回収およびCO₂貯留プロジェクト
の課題となっている。本論文では，メタケイ酸ナトリウム（S-
MS）水溶液と溶解CO₂の反応によるゲル化特性を評価した。
ラマン分光およびSEM-EDSの測定から，生成ゲルは主として
Na₂CO₃ゲル（S-C-ゲル）であり，そのゲル化時間，強度およ
び安定性を温度，S-MS濃度，NaCl濃度，Ca²⁺濃度およびCO₂
圧力について調べた。ゲル化時間は温度と圧力により約1～24
時間の範囲で変化し，ゲル強度はS-MS濃度およびCO₂圧力が
高いほど増加した。内部でゲルを生成させた砂岩コアは，水飽
和の場合に比較して限界圧力勾配（Threshold Pressure Gradient）
が2.6倍に増加し，ガス透過率は約1/10に減少することが明ら
かになった。また，原位置S-C-ゲルの生成は安定な地下貯留
層におけるCO₂固定化方法としても期待できる。