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# Viscosity of Aqueous $CO_2$ Solutions Measured by Dynamic Light Scattering

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The viscosity of water containing dissolved $CO_2$ at pressures up to 5 MPa was measured by a new method based on dynamic light scattering. The accuracy of this method was checked by measuring the temperature dependence of the viscosity of pure water at pressures up to 5 MPa and comparing the results to values in the literature that were determined by other methods. The discrepancy of the measured viscosity of water from the literature was ±6%, which roughly equals the composite uncertainty of this method. The effect of dissolved $CO_2$ on the viscosity was determined as functions of time, temperature, and pressure. The results indicated that the viscosity of the solution increased with increasing $CO_2$ concentration. High viscosities were observed under conditions in which $CO_2$ hydrates can form.

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## 1. Introduction

The increasing level of greenhouse gases, particularly $CO_2$, is now a serious global concern. To reduce $CO_2$ emissions into the atmosphere, sequestration of $CO_2$ in ocean depths has been proposed as a promising method. However, to assess the environmental impacts of the $CO_2$ sequestration method, we should understand how $CO_2$ solutions behave after liquid $CO_2$ is injected into seawater. The viscosity of the solution is one such property that should be known, but its measurement has so far been limited to low $CO_2$ concentrations.

At high $CO_2$ concentrations, especially at lower temperatures, the solution is known to form $CO_2$ clathrate-hydrates, hereafter referred to as $CO_2$ hydrates. $CO_2$ hydrates are icelike inclusion compounds formed from $CO_2$ and water at the water−$CO_2$ interface at temperatures $T$ below 283.4 K and at pressures $P$ above 4.45 MPa.[1] Such conditions exist deep in the sea; in fact, such hydrates are known to form naturally on the seabed.[2] The formation of $CO_2$ hydrates can therefore significantly influence $CO_2$ sequestration processes. This crystalline compound forms from $CO_2$ solutions after an induction period, that is, a period of time in which no crystalline solid is observed even if the $P-T$ condition is suitable for hydrate formation. During the induction period, which was reported to vary from several minutes to over several weeks, the structure of the solution likely changes. For example, during this period the water molecules in the solution could construct a hydrogen-bonded cagelike structure around the dissolved $CO_2$ molecules, which would raise the viscosity of the solution.

Some experimental efforts have been made to determine the structure of the $CO_2$ solution. Uchida et al.[3] studied the intramolecular vibrations of $CO_2$ molecules in solution by using Raman spectroscopy. They found that the intramolecular vibrations of solute molecules were affected by the solvent molecules close to the solute molecules. For the C−O stretching mode of the $CO_2$ molecules, the difference in the spectra between the solution and the hydrate structure indicated that vibrations of the $CO_2$ molecules in water were affected by the water molecules within about 0.25 nm of each $CO_2$ molecule, a distance that is slightly smaller than the pore space in the hydrate lattice. In contrast, similar measurements were done for $CH_4$ (another hydrate former), which indicated that the local environments of the $CH_4$ molecules in water are nearly the same as those in the hydrate structure.[4] Kumagai and Yokoyama[5] and Oyama et al.[6] measured the viscosity of $CO_2$ solutions via a falling capillary viscometer and a strain-gauge-type viscometer, respectively. They found that the viscosity of the $CO_2$ solutions increased prior to the hydrate formation. Their results indicate that structuring of the solution raised the viscosity of the solution. Such macroscopic measures of viscosity should be complemented with microscopic measures to understand how $CO_2$ solutions become structured prior to hydrate formation. Also, although previous techniques have determined the viscosity coefficients for bulk properties of the solution, they require other physical properties, such as density and mole fraction of $CO_2$, to obtain the viscosity coefficient; thus, they are difficult to apply to dynamic conditions.

We applied the dynamic light scattering (DLS) method to observe such dynamic conditions because it is a powerful tool that probes the microscopic structure as a function of time during the induction period and thus can complement the bulk viscosity measurements. The present paper aims to evaluate the DLS method for viscosity measurements of $CO_2$ solutions and to determine how the viscosity depends on time, temperature, and pressure.

## 2. Experimental Methods

The light from a laser was focused at the center of the sample cell containing the sample solution. The light scattered by the sample was collected by an objective lens at an angle of 90° to the incident light. The wavelength of the scattered light was modulated by the Brownian motion of the sample material and then went through two pinholes. The light from these pinholes interfered and created

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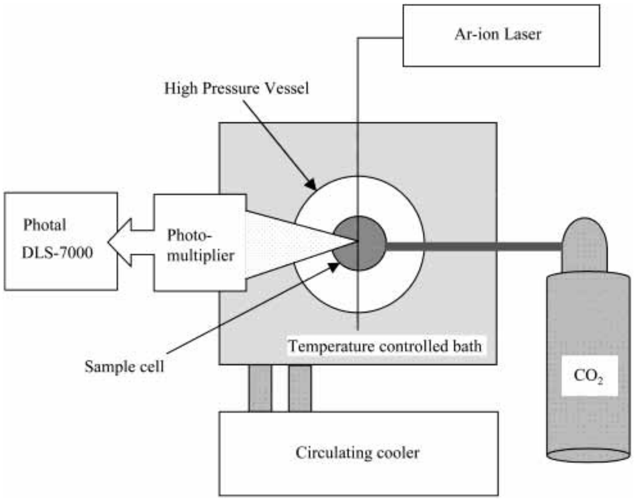

**Figure 1.** Block diagram of the experimental system.

a self-beat. The time-dependent intensity of the light scattering was measured with a photomultiplier system. Then the autocorrelation function $q_e(\tau)$ of the temporal fluctuations in the photocurrent output was computed and the translational diffusion coefficient of the solution $D_\tau$ was determined using its measured value from a dynamic light scattering spectrophotometer:[7]

$$q_e(\tau) = \exp(-D_\tau q^2 \tau) \tag{1}$$

where $\tau$ is the relative time and $q$ is the scattering vector. The particle diameter $d$, the viscosity $\eta_0$, and the diffusion coefficient $D_\tau$ are related by the Stokes−Einstein law:

$$d = kT/3\pi\eta_0 D_\tau \tag{2}$$

where $k$ is Boltzmann's constant. A very small quantity of polystyrene latex particles with a uniform size of 204 nm (Seradyn Uniform Microparticles) was mixed with distilled water, added to the solution as local probes, and then mixed with the $CO_2$ solution in the sample cell. This latex solution is appropriate for eq 2 because the latex particles all have nearly the same size and the latex solution was very dilute. Thus, we assume no particle size variation. This particle size has the best performance for the light scattering in our experimental setup. The existence of the small amount of latex particles is assumed not to affect the structure formation in the solution based on the experimental aspects reported by Servio et al.[8]

The diffusion coefficient in the mixed solution was measured using the apparatus in Figure 1 and then calculated using the Marquardt approximation method. To ensure the quality of the results, the data used for further analysis were selected only if the residual errors for the approximations were less than 0.5%. The diffusion coefficient derived from this experiment can be approximated to that of the latex particles because the intensity of the light scattered by the latex particles is much stronger than that of the light scattered by the $CO_2$ solution. To determine the viscosity, the known particle diameter and the measured diffusion coefficient of the latex particles in the solution were substituted in eq 2.

The incident light source was an Ar-ion laser ($\lambda = 488$ nm), and the laser power was approximately 20 mW. The laser light went into the high-pressure vessel (Otsuka Electronics, sample cell volume of about 0.6 $cm^3$) through one of six silica-glass windows. The pressure vessel was designed to contain pressures of up to 5 MPa. Temperature was measured with a type T thermocouple, pressure was measured with a pressure transducer (Kyowa, type PHS-100KA), and both were recorded on a Chino type AL3000 recorder. The 90° scattered light intensity was measured with a dynamic light scattering spectrophotometer (Photal, type DLS-7000) and analyzed by the computer system. The temperature of the sample was controlled by circulating oil around the pressure vessel with a Haake type F8-C40 circulating cooler. The temperature was controlled to within ±0.2 K.

The systematic errors for estimating the viscosity of the $CO_2$ solution are mainly from the variation of the temperature control and from the change of the refractive index of water due to the $CO_2$ dissolution, which is the parameter $q$ in eq 1. The maximum refractive index change of water due to the $CO_2$ dissolution is the difference between the refractive indices of pure water and of $CO_2$-saturated water. This index change was estimated using the Lorentz−Lorentz equation to be approximately 0.9% for the temperatures in the present study. The systematic error for the estimate of $\eta$ is about 2.3% in the present experimental setup.

The sample was prepared from distilled and deionized pure water (resistivity, about 18 MΩ·cm) and filtered through a 0.2-$\mu$m filter three times prior to use. A small amount of uniformly sized polystyrene latex particles (less than $5 \times 10^{-6}$ by mass fraction) was then added to the water. Approximately 0.5 $cm^3$ of the sample solution was introduced into the high-pressure vessel, and the temperature of the system was controlled at room temperature. Then the initial water viscosity was measured prior to each experimental run to check the reliability of the particle distribution. The pressure of the system was controlled using the regulator on the $CO_2$ bomb. To evaluate the viscosity measurement via the DLS method, we measured the temperature dependence of the viscosity coefficient of pure water at pressures up to 5 MPa. We used the liquid pumping system (FLOW type 301) to control the water pressure.

We did the following measurements: (1) viscosity of pure water to evaluate the DLS method, (2) viscosity change of the $CO_2$ solution after pressurization of $CO_2$, and (3) viscosity of the $CO_2$-saturated solution at various temperatures and pressures. We used $CO_2$ from Hokkaido Air-Water (purity, approximately 0.99).

## 3. Results and Discussion

***3.1. Viscosity of Pure Water.*** The viscosity of pure water at pressures up to 5 MPa was measured at temperatures between (276.2 and 285.2) K to evaluate this method for viscosity measurements. The temperature was first set at 285.2 K and then reduced in 2 K steps for measuring the temperature dependence of the viscosity. Each measurement was done 1 h after reaching the set temperature and repeated three times to evaluate measurement deviations. The maximum deviation of the obtained values from the average value was at most ±5%.

The temperature dependence of the viscosity of pure water $\eta_w^{exp}$ under atmospheric pressure is shown in Figure 2 and listed in Table 1. The dotted line indicates the viscosity of pure water $\eta_w^{lit}$ obtained from the literature.[9] This figure shows that the average value of each experimental data point agrees well with data in the literature (i.e., the absolute deviation $AD = |(\eta_w^{exp} - \eta_w^{lit})/\eta_w^{lit}|$ is within ±4.3%). The composite uncertainty of this system for the viscosity calculation is estimated to be at most 6%, which is larger than the deviation of the average value from the

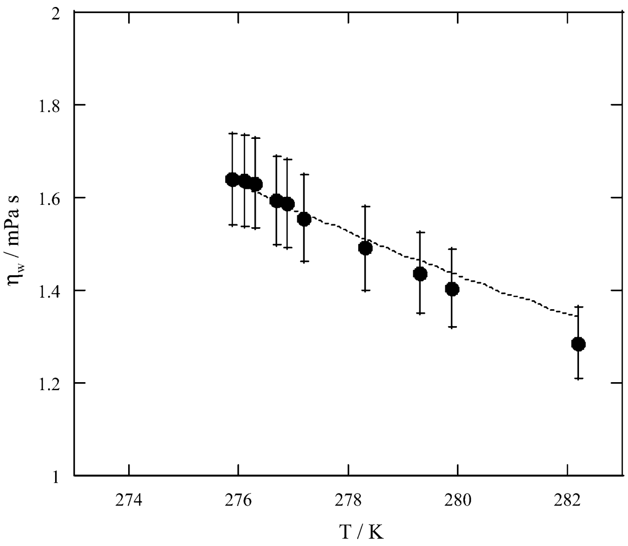

**Figure 2.** Temperature *T* dependence of the viscosity of pure water $\eta_w$. The experimental uncertainty, ±6%, is shown by the error bars. The dotted line is data from the literature (ref 9).

**Table 1. Temperature *T* Dependence of the Viscosity $\eta_w$ of Pure Water[a]**

| $T$ | $\eta_w^{exp}$ | $\eta_w^{lit}$ | AD | $T$ | $\eta_w^{exp}$ | $\eta_w^{lit}$ | AD |
|---|---|---|---|---|---|---|---|
| K | mPa·s | mPa·s | % | K | mPa·s | mPa·s | % |
| 282.2 | 1.286 | 1.344 | 4.3 | 276.9 | 1.588 | 1.581 | 0.4 |
| 279.9 | 1.405 | 1.440 | 2.4 | 276.7 | 1.594 | 1.591 | 0.1 |
| 279.3 | 1.437 | 1.466 | 2.0 | 276.3 | 1.631 | 1.612 | 1.2 |
| 278.3 | 1.491 | 1.513 | 1.4 | 276.1 | 1.637 | 1.623 | 0.9 |
| 277.2 | 1.555 | 1.566 | 0.7 | 275.9 | 1.640 | 1.633 | 0.4 |

[a] $\eta_w^{exp}$, measured viscosity of pure water; $\eta_w^{lit}$, calculated viscosity of pure water (ref 9); AD, absolute deviation between $\eta_w^{exp}$ and $\eta_w^{lit}$.

**Table 2. Temperature *T* Dependence of the Viscosity of $CO_2$ Solutions $\eta$ at Pressures *P* of about 0.7 MPa[a]**

| $T$ | $P$ | $\eta$ | $10^3C$ | $\eta_w^{exp}$ | $\eta_w^{lit}$ | AD |
|---|---|---|---|---|---|---|
| K | MPa | mPa·s | mole fraction | mPa·s | mPa·s | % |
| 282.1 | 0.729 | 1.267 | 6.05 | 1.319 | 1.347 | 2.1 |
| 281.8 | 0.769 | 1.225 | 6.15 | 1.329 | 1.359 | 2.2 |
| 281.0 | 0.71 | 1.367 | 6.41 | 1.357 | 1.392 | 2.5 |
| 279.3 | 0.71 | 1.440 | 6.95 | 1.428 | 1.465 | 2.6 |
| 279.1 | 0.71 | 1.438 | 7.02 | 1.442 | 1.475 | 2.2 |
| 277.5 | 0.69 | 1.570 | 7.54 | 1.558 | 1.550 | 0.5 |
| 277.1 | 0.69 | 1.604 | 7.66 | 1.589 | 1.570 | 1.2 |
| 276.7 | 0.69 | 1.605 | 7.79 | 1.613 | 1.590 | 1.4 |
| 276.4 | 0.69 | 1.647 | 7.89 | 1.622 | 1.606 | 1.0 |
| 276.0 | 0.69 | 1.694 | 8.02 | 1.652 | 1.630 | 1.4 |
| 275.9 | 0.69 | 1.705 | 8.05 | 1.660 | 1.632 | 1.7 |
| 275.6 | 0.69 | 1.739 | 8.15 | 1.683 | 1.649 | 2.1 |
| 275.3 | 0.67 | 1.771 | 8.24 | 1.705 | 1.665 | 2.4 |
| 275.1 | 0.67 | 1.807 | 8.31 | 1.725 | 1.676 | 2.9 |
| 275.0 | 0.67 | 1.794 | 8.34 | 1.737 | 1.676 | 3.7 |
| 274.7 | 0.67 | 1.818 | 8.44 | 1.774 | 1.699 | 4.4 |
| 274.5 | 0.67 | 1.839 | 8.50 | 1.808 | 1.710 | 5.7 |
| 274.3 | 0.67 | 1.851 | 8.57 | 1.827 | 1.722 | 6.1 |
| 273.9 | 0.67 | 1.855 | 8.69 | 1.833 | 1.745 | 5.0 |

[a] $\eta$, obtained viscosity of $CO_2$ solution; *C*, estimated $CO_2$ concentration (mole fraction) in pure water (ref 10); $\eta_w^{exp}$, obtained viscosity of pure water; $\eta_w^{lit}$, calculated viscosity of pure water (ref 9); AD, absolute deviation between $\eta_w^{exp}$ and $\eta_w^{lit}$.

literature value. We also compared the obtained results at 0.7 and 5 MPa to the literature values. The values are listed in Tables 2 and 3. These results also support the evaluation described here. Therefore, we conclude that the DLS method proposed in this study can determine the viscosity of the liquid sample with a maximum uncertainty of 6%.

**Table 3. Temperature *T* Dependence of the Viscosity of $CO_2$ Solutions $\eta$ at Pressures *P* of about 5 MPa[a]**

| $T$ | $P$ | $\eta$ | $10^2C$ | $\eta_w^{exp}$ | $\eta_w^{lit}$ | AD |
|---|---|---|---|---|---|---|
| K | MPa | mPa·s | mole fraction | mPa·s | mPa·s | % |
| 282.2 | 4.87 | 1.576 | 2.77 | 1.335 | 1.339 | 0.3 |
| 282.1 | 4.92 | 1.539 | 2.77 | 1.338 | 1.343 | 0.3 |
| 281.0 | 4.91 | 1.728 | 2.80 | 1.375 | 1.387 | 0.9 |
| 279.6 | 4.91 | 1.848 | 2.83 | 1.425 | 1.447 | 1.5 |
| 278.6 | 4.89 | 1.948 | 2.85 | 1.459 | 1.492 | 2.2 |
| 277.7 | 4.85 | 2.078 | 2.87 | 1.459 | 1.534 | 4.9 |
| 276.5 | 4.81 | 2.171 | 2.90 | 1.634 | 1.594 | 2.5 |
| 276.3 | 4.79 | 2.179 | 2.91 | 1.652 | 1.604 | 3.0 |
| 276.1 | 4.78 | 2.225 | 2.91 | 1.670 | 1.614 | 3.4 |
| 274.6 | 4.61 | 2.951 | 2.94 | 1.804 | 1.696 | 6.4 |
| 274.2 | 4.63 | 2.827 | 2.95 | 1.802 | 1.719 | 4.8 |

[a] $\eta$, obtained viscosity of $CO_2$ solution; *C*, estimated $CO_2$ concentration (mole fraction) in pure water (ref 10); $\eta_w^{exp}$, obtained viscosity of pure water; $\eta_w^{lit}$, calculated viscosity of pure water (ref 9); AD, absolute deviation between $\eta_w^{exp}$ and $\eta_w^{lit}$.

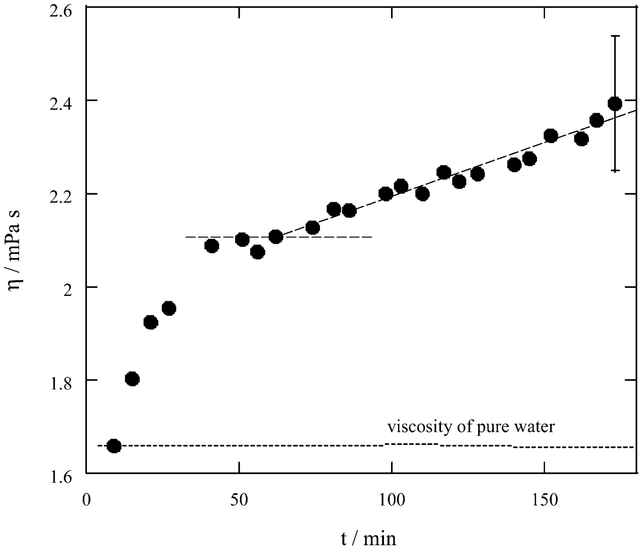

**Figure 3.** Variation of the viscosity $\eta$ at 275 K with time *t* after $CO_2$ pressurization to 4 MPa. The dotted line is the viscosity of pure water at 4 MPa and 275 K (ref 9). The experimental uncertainty of ±6% is shown on the final data point.

***3.2. Viscosity Variation during Dissolution of $CO_2$.*** At a constant temperature of approximately 275 K, the viscosity of water was measured under a $CO_2$ pressure of about 4 MPa. Figure 3 shows how the viscosity increased with time after pressurization. Initially, the water has negligible $CO_2$ and our measured viscosity in this case agrees with that for pure water.[9] The viscosity of the $CO_2$ solution then increased gradually with time and temporarily reached a constant value at about 50 min, as marked by a horizontal dashed line. As discussed later, this value coincides with that expected from the viscosity dependence on temperature and pressure. The constant value is approximately 25% greater than $\eta_w^{lit}$, the value for pure water marked with a dotted line. This phenomenon indicates that the viscosity of the solution increases as more $CO_2$ dissolves into water. Under the experimental temperature and pressure conditions, the water sample of approximately 0.5 $cm^3$ is apparently saturated with $CO_2$ within 1 h.

A second dashed line in Figure 3 shows a linearly increasing viscosity after 60 min. The analysis of each measurement indicated that the distribution of particle size obtained from the DLS data did not change even during this second (linearly) increasing stage, so the viscosity change was not due to coagulation of the latex particles.

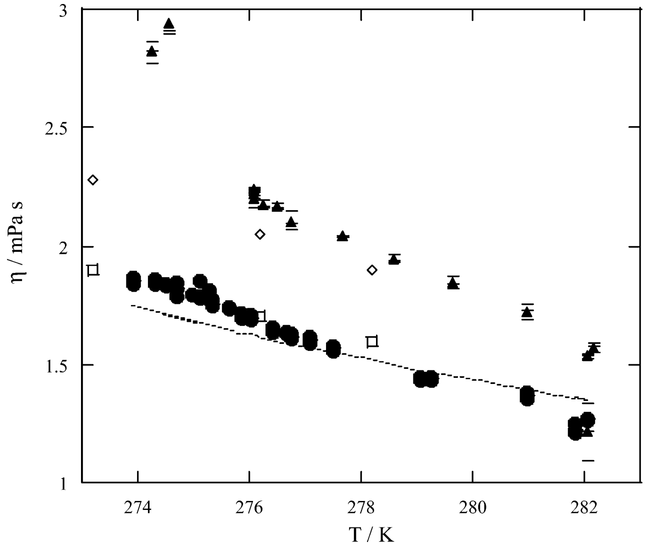

**Figure 4.** Temperature *T* dependence of the viscosity of $CO_2$ solutions $\eta$ at two pressures: ●, for pressure $P = 0.7$ MPa; ▲, for $P = 5$ MPa. Each data point is the average of three repeated measurements shown by the flat bars. The dotted line is the value for pure water $\eta_w$ (ref 9) at $P = 0.7$ MPa. □ and ◇, estimated from the high-pressure experiments (ref 5) by assuming the mole fraction of $CO_2$ to be saturated.

Because $CO_2$ hydrate is stable at this temperature and pressure, the increase of the viscosity after the $CO_2$ saturation could be related to a changing structure in the solution. Because there was no evidence to indicate hydrate formation, such as a temperature increase from the enthalpy of the phase change, the linear viscosity increase is considered to be a property of the induction period. An increase of the viscosity of $CO_2$ solution during the induction period was also reported by Oyama et al.,[6] which was measured macroscopically by a rotating viscometer. Therefore, we conclude that the viscosity increases with time during the induction period with increasing $CO_2$ supersaturation until a bulk hydrate crystal is formed in the system.

***3.3. Viscosity of $CO_2$ Solutions at Various Temperatures and Pressures.*** The viscosity of $CO_2$ solutions was measured at 0.7 and 5 MPa for temperatures ranging from about 283 to 274 K. To measure the steady-state value, each measurement was done 1 h after the temperature change. At the lower pressure, $CO_2$ hydrate could not form at the experimental temperature and the $CO_2$ phase was vapor; at the higher pressure, $CO_2$ hydrate is stable and the $CO_2$ phase was liquid.

The temperature dependence of the viscosity of $CO_2$ solutions $\eta$ is shown in Figure 4 and listed in Tables 2 and 3. These tables also include the estimated concentration of $CO_2$ in the solution *C*. In the present study, *C* is assumed to equal the solubility of $CO_2$ in liquid water.[5] As described previously, $\eta_w^{exp}$, $\eta_w^{lit}$, and AD are also shown in Tables 2 and 3 to estimate the uncertainty of $\eta$. The viscosity of the $CO_2$ solution is larger than $\eta_w^{lit}$ at all temperatures except temperatures just below room temperature. The difference of the viscosity between the experimental data $\eta$ and $\eta_w^{lit}$ is larger than the experimental uncertainty at lower temperatures and at higher pressures. The reason for this difference could be related to the higher solubility of $CO_2$ in water under these conditions. On the basis of the fact that the viscosity increased as $CO_2$ dissolved, as shown in the previous section, we conclude that the viscosity of the solution is controlled by the concentration of $CO_2$ in the solution.

To compare our experimental data with previous literature data, we estimated the viscosity of $CO_2$ solutions at our conditions from the data of Kumagai and Yokoyama.[5] They used a drop-needle viscometer to measure the viscosity of $CO_2$ solutions at 30 MPa with various mole fractions of $CO_2$. Because they had no data for the lower pressures that we used in this study, we estimated the viscosity from their data by assuming the $CO_2$ mole fractions in the saturated solution. This assumption is based on the observation shown in the previous section, that is, the sample was $CO_2$-saturated in 1 h. The good agreement of our data with those interpreted from the literature[5] (open squares in Figure 4) indicates that the viscosity increase during $CO_2$ dissolution is due to the increasing $CO_2$ concentration in the solution.

On the basis of this conclusion, the difference of viscosity between higher and lower pressure experiments can be explained. When we extend the viscosity line of Kumagai and Yokoyama[5] to the $CO_2$ concentrations for saturated conditions at 5 MPa under various temperatures,[10] the estimated viscosities at temperatures of (273.2, 276.2, and 278.2) K are approximately (2.30, 2.05, and 1.90) mPa·s, respectively. Because we did not measure the mole fraction directly, we assumed that the mole fractions of $CO_2$ in our measurements were saturated. The estimated values are shown as open diamonds in Figure 4, which qualitatively agree with our experimental data. At temperatures below about 278 K, however, the measured viscosities are larger than the expected values. Also, the viscosities measured at approximately 274 K (about 2.9 mPa·s) are 30% larger than those estimated from the literature (about 2.2 mPa·s). These higher viscosities could also be due to clathrate-like structures forming during the relatively long exposure times to the $CO_2$ phase compared to the higher temperature measurements in the same run.

As shown in Figure 3, the viscosity of the solution increases linearly with time after reaching its saturation condition. This saturated value agrees well with that expected from Figure 4. Therefore, it may be interpreted that the viscosity of the $CO_2$ solution at 5 MPa for temperatures below 278 K coincides with those estimated from Kumagai and Yokoyama[5] when the water is saturated with $CO_2$. However, after that, the viscosity will continuously increase with time, possibly due to a changing structure of the solution. This process could be related to hydrate formation processes. For example, Oyama et al.[6] reported that the viscosity of $CO_2$ solution greatly increased prior to hydrate formation. We suggest that the relatively large quantity of water molecules tends to construct a hydrogen-bonded precursor to the hydrate structure. Therefore, we argue that the viscosity increase prior to hydrate formation is due to a changing structure of the $CO_2$ solution.

## 4. Conclusions

The dynamic light scattering method was used to measure the viscosity of $CO_2$ solutions at high pressures and low temperatures. The measured viscosity agreed well with data in the literature, which shows the validity of this new method. After the water was pressurized by $CO_2$, the viscosity (1) increased with time until reaching a nearly steady-state value that (2) was higher at lower temperatures and (3) was higher at higher pressures. All three trends can be explained by a viscosity that increases with increasing $CO_2$ concentration. A large increase in viscosity occurred under hydrate-forming conditions in which the solution structure was likely becoming more like the clathrate structure. Thus, we argue that the viscosity is sensitive to the structure of the solution.

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