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Dynamic behavior of hydration water in calcium-silicate-hydrate gel: A quasielastic neutron scattering spectroscopy investigation

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The translational dynamics of hydration water confined in calcium-silicate-hydrate (C-S-H) gel was studied by quasielastic neutron scattering spectroscopy in the temperature range from 280 to 230 K. The stretch exponent β, the self-diffusion constant D, the average translational relaxation time ⟨τ⟩, and the temperature dependence of confinement radius α extracted from the elastic fraction of immobile water molecules p(Q) were obtained from the analyses of the low-Q spectra according to the relaxing cage model. Measurements were made using C-S-H of three different water contents, 10%, 17%, and 30%. Among the three samples of C-S-H gel with different water contents, the values of β decrease with increasing water contents, while α increases. The values of D and ⟨τ⟩ are insensitive to temperature for the two lower water contents, as opposed to the 30% case where a slight variation is observed. The trend for violation of the Stokes-Einstein relation is only visible in the case of 30% water content.

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I. INTRODUCTION

Hydration water is an important component in a cement paste, regulating the transport phenomena, thus influencing the mechanical properties as well as the durability of the hardened matrix. Calcium-silicate-hydrate (C-S-H) is the main binding phase in Ordinary Portland Cement. Its presence is critical to the strength development and the durability of a cement paste. By mixing water and a cement powder to form a cement paste which eventually evolves into a solid concrete, the pore system evolves during the cement setting process from a percolated macroporous to a percolated microporous structure [1]. C-S-H is a gel-like material. There are several existing C-S-H structure models [2,3]. The recently accepted model is Jennings’s colloidal model (CM-II) [3]. This is a hybrid model and it is able to explain several experimental evidences such as those derived from scattering measurements [1] and sorption isotherm experiments [3]. By this model, C-S-H is formed by nanoscale objects (also called “globules”), which clusterize in fractal objects with an associated pore system. The basic globule is a disklike object, whose thickness is around 4 nm. Moreover, the globule presents a layered internal structure which defines interlamellar spaces, in very small cavities [intraglobular pores (IGPs)] with dimensions smaller than 1 nm. The packing of these globules produces a porous fractal structure. The pores are usually identified as the small gel pores (SGPs), with dimensions of 1–3 nm, and the large gel pores (LGP), 3–12 nm in size [3,4]. In a C-S-H system, water molecules are mainly confined in the inter-globular space (i.e., SGP and LGP), there being a smaller amount present in the intralayer small cavities (i.e., IGPs) proper of the internal structure of the globules [5]. Although the nanostructure of C-S-H has been intensively studied in real cement pastes and in synthetic C-S-H [3–7], many questions remain concerning the dynamics of water confined in C-S-H gel at different water contents.

Recently, many time-resolved studies on the dynamics of hydration water in curing cement pastes have been made by incoherent quasielastic neutron scattering (QENS) experiments [8–12]. A QENS experiment generally measures the dynamics of hydrogen atoms in the investigated sample. Thus, QENS results can provide information about long-time diffusive motions (on the time scale of 1 ns) of water molecules confined in cement pastes. In a previous study [12], we have investigated the hydration kinetics and transport properties of aged cement pastes at w/c = 0.31 (i.e., weight of water divided by weight of dry cement) at room temperature with and without superplasticizer (SP) by QENS. Moreover, we performed elastic scans for all samples in the temperature range from 25 to 300 K. From these measurements, the mean squared displacement (MSD) of hydrogen atoms associated to the hydration water on a time scale of 1 ns was extracted as a function of temperature. In all cases, MSD evidenced the presence of a discontinuity occurring at a temperature of about 230 K, which can be related to a dynamic crossover phenomenon. The experimental results indicate that the diffusive dynamics of hydration water confined in cement paste is related to the curing time and that the presence of SP retards the hydration process and slightly decreases the dynamic crossover temperature.

Main hydration products in a cement paste are C-S-H gel and calcium hydroxide crystals (i.e., Portlandite), which both contain hydrogen atoms participating in the QENS spectrum. For this reason, the investigation of a pure synthetic C-S-H gel could provide information coming solely from hydration water in the C-S-H phase avoiding any contamination from the Portlandite phase. Water dynamics confined in C-S-H (I) have been recently investigated by broadband dielectric spectroscopy (BDS) [13] as function of temperature and water content. Three different dielectric processes have been

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disclosed where the fastest is related to some intrinsic dipolar
group in the C-S-H gel while the other two processes are
directly related to the water molecules confined in the gel
(i.e., process 2 is related to water confined in IGP and water
interacting with the C-S-H surface, while process 3 is related to
water that behaves in a cooperative way). But the investigation
only focused on the low temperature (range of 100–250 K) and
low water contents (from 6 to 15 wt %). What is the behavior of
the diffusive dynamics of hydration water confined in C-S-H
gel with different water content, both low and high? Could the
dynamic crossover in the confined water be observed also in
the pure C-S-H gel at a temperature near 230 K for all water
contents? To answer these questions, we have done a further
study about the dynamics of hydration water confined in pure
C-S-H gel by a QENS experiment.

The present investigation aims to study the temperature
dependence of the dynamic behavior of the water confined in a
synthetic C-S-H (I) gel as a function of different water contents
(i.e., 10%, 17%, and 30%) by the QENS technique using the
BASIS spectrometer [14] at Oak Ridge National Laboratory
(ORNL). The investigated temperature range is from 280
to 230 K. From the data analysis, we obtain the diffusion
coefficients describing the transport phenomena in the C-S-H
phase at different water levels. Moreover, the presented results
seem to indicate the breakdown of the Stokes-Einstein relation
at 230 K only for the 30% water level case. The breakdown
of the Stokes-Einstein relation in supercooled water [15] is
the physical equivalent of a dynamic crossover temperature at
around 230 K.

II. QENS EXPERIMENTS

The high-resolution backscattering spectrometer BASIS
[14] at ORNL Spallation Neutron Source (SNS) was used
to measure the C-S-H gel with different water levels from
280 to 230 K. BASIS is well suited for probing diffusive and
relaxation motions for confined and hydration water [16,17].
In the quasielastic regime of operation BASIS can be used to
probe dynamic processes on the pico- to nanosecond time scale. BASIS is an inverse geometry time-of-flight
backscattering spectrometer. The detected near-backscattering
neutrons are reflected from Si(111) analyzer crystals to select
the final neutron energy of 2.08 meV. The initial neutron energy
is determined from the neutrons’ time of flight. The silicon
analyzer crystals cover about 2.0 sr (16% of 4π). Neutrons
are scattered by a sample illuminated by a polychromatic
neutron beam, the bandwidth of which is defined by a set
of neutron choppers. The dynamic range of the experiment
can be adjusted. In our QENS experiment, the dynamic range
of neutron energy is chosen from −120 to +120 μeV.

In the QENS experiment, the samples of C-S-H gel with
different water content were loaded uniformly between folded
aluminum foils. Each foil with a sample was wrapped around
the inner surface of a hollow aluminum cylindrical sample
holder. The holder is 5 cm high and has a 29-mm inner
diameter in order to fully intercept the neutron beam, which
is about 30 × 30 mm2 at the sample position. The resolution
function was measured at 3.5 K for each sample. In order to
have analytical function representing the measured resolution
function used in data analysis while minimizing the errors
introduced during the convolution operation, the resolution
function \( R(Q, E) \) is first fitted by a sum of four Gaussian functions:

\[
R(Q, E) = \sum_{i=1}^{4} \frac{A_i}{\sqrt{2\pi} \sigma_i} \exp \left[ -\frac{(E - E_i)^2}{2\sigma_i^2} \right], \tag{1}
\]

where \( A_i \) is the amplitude, \( E_i \) is the peak position, and \( \sigma_i \) is
the width of each Gaussian component; \( Q \) is the magnitude
of the wave vector transfer of scattered neutrons; \( E \) is the neutron
energy transfer. In this way the smooth resolution function can
be obtained from the experimental resolution function for each
C-S-H gel at each \( Q \).

Figure 1 shows the fitted resolution function with a sum
of four Gaussians for C-S-H gel with 30% water content at
\( Q = 0.5 \) and 0.9 Å⁻¹, the corresponding normalized measured
and fitted resolution functions, and the Fourier transform of
the normalized fitted resolution function. We can see that the
four Gaussians are enough to fit the shape of the experimental
resolution function at each \( Q \). In this way, the fitted resolution
function can be used in the data analysis of QENS spectra
to extract parameters describing the translational dynamics of
water confined in the synthetic C-S-H (I) gels.

In our QENS experiment, we measured three samples of C-S-H
gel with water content of 10%, 17%, and 30%, and an
empty cell holder. The measured temperature ranges from 280
to 230 K. The measured neutron intensity of each sample was
obtained by subtracting the measured data of the empty cell
holder for describing the contribution only from the hydration
water confined in C-S-H gel.

Synthetic C-S-H was prepared by hydrating pure tricalcium
silicate (C₃S) in an excess of decarbonated water. A chemically
pure batch of C₃S was obtained from CTG-Italcementi
(Bergamo, Italy) as a gift. The resulting specific surface area
of the C₃S as detected by N₂ sorption isotherms (BET) was
0.65 m²/g while the mean radius was 4.66 μm. C-S-H was
prepared by mixing 4 g of C₃S with 1150 g of distilled
water. The excess of water in respect to C₃S was essential
to minimize the Portlandite [Ca(OH)₂] coprecipitation. The
resulting C₃S-water dispersions were continuously stirred for
at least 40 days in a plastic bottle, which was sealed to avoid any
carbonation. The synthesis was conducted at 25 ± 2°C. This
synthesis leads to a quite polydisperser C-S-H phase, which
is usually referred to as C-S-H (I) [18]. The dispersion was
then filtered under a N₂ atmosphere to avoid carbonation and
the obtained solid was dried in an oven at 60°C for about 3 h.
The resulting gel was dried to the desired water content using a
vacuum oven operating under a N₂ atmosphere at temperatures
below 100°C. Energy-dispersive x-ray spectroscopy (EDX,
X-sight Oxford microprobe) evidenced an average Ca-Si ratio
of 1.5 ± 0.3 confirming the expected inhomogeneity of the
sample. Thermogravimetric analysis (TGA) was performed
to check the final water, Portlandite, and calcium carbonate
contents. Ca(OH)₂ and CaCO₃ resulted in the range 5%–10%
in respect to the total mass of the samples. TGA experiments
were carried out with a SDT Q600 apparatus (TA Instruments)
heating the sample in alumina pans at 10°C/min from 25°C to
1000°C under a constant flux of pure N₂ (100 mL/min). Water
contents of 10%, 17%, and 30% (weight percent = grams of
water divided by grams of dry cement) were achieved at the

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an elastic neutron scattering (ENS) and the second part formula (2), which can be decomposed into a sum of two parts. The first part in the right-hand side of Eq.(2) represents the translational part and the rotational part. As to the spectra with $Q \leq 1.1 \text{ Å}^{-1}$, the rotational contribution becomes negligible. The SISF can be obtained by

$$I(Q,E) = (p(Q)\delta(E) + (1-p(Q))\mathcal{F}(F_{S}(Q,t))) \otimes R(Q,E)$$

where $Q$ is the magnitude of the wave vector transfer of scattered neutrons, $E$ is the neutron energy transfer, and $t$ is the time. The elastic fraction $p(Q)$ can arise from the incoherent structure factor (EISF) [20]. It can also arise due to the contribution from hydrogen atoms that cannot migrate over a dimension more than $2\pi/Q$ within the experimental observation window (i.e., the fraction of water that is structurally bound to the C-S-H phase). In this case it is called the Debye-Waller factor [21]. $F_{S}(Q,t)$ is the self-intermediate scattering function (SISF) of QENS part, and $\mathcal{F}(F_{S}(Q,t))$ is the Fourier transform of SISF. $R(Q,E)$ is the $Q$-dependent energy resolution function. It is obtained by fitting formula (1) to the measured neutron spectra intensity data obtained from a low-temperature run at 3.5 K with each sample (at which temperature all the relaxation processes are frozen).

According to the RCM model, the SISF is a product of the translational part and the rotational part. As to the spectra with $Q \leq 1.1 \text{ Å}^{-1}$, the rotational contribution becomes negligible. The SISF can be obtained by

$$F_{S}(Q,t) \approx F_{t}(Q,t) = F_{t}^{\nu}(Q,t) \exp\{-t/\tau(Q)\}$$

### III. THE MODEL ANALYSIS OF QENS SPECTRA

QENS experiment can provide information about whether there is a dynamic transition temperature in confined water. The measured neutron spectral intensity distribution is proportional to the incoherent dynamic structure factor $S_{H}(Q,E)$ of hydrogen atoms in water molecules confined in C-S-H gel. We analyzed the measured neutron spectra by the relaxing cage model (RCM), which has been tested extensively by means of molecular dynamic simulation and QENS experiments [12,19]. The measured neutron intensity $I(Q,E)$ as a function of energy transfer $E$ at each $Q$ is analyzed with the following formula (2), which can be decomposed into a sum of two parts. The first part in the right-hand side of Eq. (2) represents an elastic neutron scattering (ENS) and the second part quasieelastic neutron scattering (QENS).

$$I(Q,E) = (p(Q)\delta(E) + (1-p(Q))\mathcal{F}(F_{S}(Q,t))) \otimes R(Q,E)$$

### TABLE I. List of the measured C-S-H gel samples at different water contents and temperatures.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Measured temperature (K)</th>
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<tr>
<td>10% water content</td>
<td>280, 270, 260, 250, 240, 230</td>
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<tr>
<td>17% water content</td>
<td>280, 270, 260, 250, 240</td>
</tr>
<tr>
<td>30% water content</td>
<td>280, 270, 260, 250, 240, 230</td>
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where $\tau_T(Q) = \tau_0 (aQ)^{-\gamma}$, $a = 0.5$ Å [19]; $F^x(Q,t)$ is the short-time vibrational motion of the water molecules in the cage. In the longer time scale where the stretch exponential relaxation term becomes substantial, the $F^x(Q,t)$ tends to its asymptotic value $F^x(Q,t) = \exp(-Q^2 a^2/3) \approx 1$. The stretch exponent $\beta < 1$ takes into account the slow relaxation processes associated with the deviation from a simple exponential decay ($\beta = 1$) which is found in the event of a Debye-like relaxation process, and $\tau_T(Q)$ is the $Q$-dependent relaxation time.

According to RCM, the four temperature-dependent parameters $p(Q)$, $\beta$, $\tau_0$, and $\gamma$ can be obtained by fitting the measured spectral intensity data simultaneously at five $Q$ values. Then the averaged translational relaxation time can be calculated according to $(\tau) = (\frac{2}{\beta}) \Gamma(\frac{2}{\beta})$, and the parameter $\alpha$ can be fitted using equation $p(Q) = \exp[-(\alpha Q^2)/3]$. Moreover, in the case of an exponential relaxation as a result of a continuous diffusion, one has $F_s(Q,t) = \exp(-D Q^2 t)$, where $D$ is the self-diffusion coefficient. In RCM, both the exponents $\beta$ and $\gamma$ in Eq. (3) are $Q$ dependent [21], and in the hydrodynamic limit ($Q \to 0$ limit), $\beta \to 1$, $\gamma \to 2$. For this case, the self-diffusion coefficient $D$ for a water molecule within the mobile fraction can be identified by the following equations:

$$
\exp\left\{\left[-\frac{t}{\tau_T(Q)}\right]^{\beta}\right\} \quad Q \to 0, \beta \to 1, \gamma \to 2
\quad \exp\left[\left(-\frac{t}{\tau_0}\right)^{(aQ)^2}\right] = \exp(-D Q^2 t),
$$

$$
D = \frac{\alpha^2}{\tau_0} = \frac{2.5}{\tau_0}(10^{-8} \text{cm}^2/\text{s}),
$$

In order to avoid the complication of taking the Fourier transform of SISF and its convolution with the resolution function in the energy domain, we transform Eq. (2) by inverse Fourier transforming both sides:

$$
\mathcal{F}^{-1}[I(Q,E)] = [p(Q) + [1 - p(Q)] \times \exp\left(-[t/(\tau_0(0.5 Q^{-\gamma}))]^\beta\right)]\mathcal{F}^{-1}[R(Q,E)] = F_H(Q,t)F_R(Q,t),
$$

FIG. 2. (Color online) Examples of the RCM model fitting of measured QENS spectra for the C-S-H gel with 30% water content at temperature 230 K. The left-hand side panels refer to the neutron spectrum at $Q = 0.5$ Å$^{-1}$, and the right-hand side panels refer to $Q = 0.9$ Å$^{-1}$. In each panel the top figure is the Fourier transform $F_H(Q,t)$ of the deconvoluted measured neutron spectrum, namely, the Fourier transform of the incoherent dynamic structure factor $S_H(Q,E)$ of a typical hydrogen atom. The lower three panels are, respectively, the fitted curves of the measured neutron spectrum represented with different scales.
where $I(Q, E)$ is the measured neutron spectral intensity, and $R(Q, E)$ is the $Q$-dependent energy resolution function. From Eq. (5), we can directly fit the measured neutron intensity in time domain, $F^{-1}[I(Q, E)]$, by multiplying the RCM intermediate scattering function, $F_H(Q, t)$, to the resolution function in time domain $F_R(Q, t) = F^{-1}[R(Q, E)]$.

Figure 2 displays the measured data (green open circle) and the fitted curve (solid red line) using the RCM model for C-S-H (I) gel with 30% water content at 230 K for two distinct $Q$ values, 0.5 and 0.9 Å$^{-1}$. The top panels in each of the two columns show $F_H(Q, t)$, which is the Fourier transform of $S_H(Q, E)$. The lower three panels show the corrected and normalized measured neutron spectra, which are plotted on the log and linear scales to show the level of agreement between the experimental data and the model fitting. We can see that the Fourier transform of the deconvoluted measured neutron spectrum and the measured neutron intensity fitted by RCM are very good. The normalized neutron spectra can be decomposed into a sum of elastic neutron scattering (ENS) line (the dashed blue line) and QENS line (the solid thin black line). We can also see that the neutron spectrum becomes broader at the temperature 230 K as the $Q$ value increases.

IV. RESULTS AND DISCUSSION

The above-mentioned time domain data fitting process is used to analyze all the measured QENS spectra of the C-S-H (I) gel with 10%, 17%, and 30% water contents. For the data analysis, we used the measured neutron spectral intensity in the dynamic energy range from $-120$ to $120$ μeV, at five $Q$ values: 0.3, 0.5, 0.7, 0.9, and 1.1 Å$^{-1}$. A measured QENS spectrum is first corrected for the sample holder contribution. Before fitting by RCM, the measured neutron intensity and the fitted resolution function should be aligned, centered, and area normalized to unity. Then all the sample data are fitted by a nonlinear least squares method for each $Q$ and each sample, respectively.

Figure 3 gives the extracted four parameters: stretch exponent $\beta$, the self-diffusion coefficient $D$, the average translational relaxation time $\tau$, and the temperature dependence of confinement radius $\alpha$ in $p(Q) = \exp[-(\alpha Q)^2/3]$ for all the investigated cases. From Fig. 3(a), we can see that $\beta$ decreases from about 0.9 to about 0.75 by increasing the water content from 10% to 30%. This indicates that the space occupied by water molecules confined in the C-S-H gel with lower water level mainly resides inside the globules, namely SGP and LGP. So the space explored by the confined water molecules is less uniform and the value of $\beta$ decreases. From Figs. 3(b) and 3(c) we can see that $D$ and $\tau$ retain almost the same values within error bars in the 10% and 17% water content cases. On the contrary $D$ decreases while decreasing temperature for the C-S-H gel at 30% water level. This indicates that parameters $D$ and $\tau$ are sensitive to temperature only in the 30% water level case. From Fig. 3(d) we can see that the parameter $\alpha$ increases with increasing water level.
content, which corresponds to a decrease of the immobile fraction, \( p(Q) \). With increasing water content, the confined water is not only present inside the globules, but also in the pores between the C-S-H globules, where greater cavities are accessible. This results in a decrease of the immobile water fraction. So the elastic fraction \( p(Q) \) also decreases. From Fig. 3(d) we can also see that in all cases \( \alpha \) decreases when the temperature diminishes. When a water molecule is confined in a space in equilibrium with a thermal bath at temperature \( T \), the \( p(Q) \) is a Debye-Waller factor and has the functional form \( p(Q) = \exp(-Q^2/\langle r^2 \rangle /3) \) [21], where \( r^2 \) is the mean square displacement, MSD, of the confined water molecules (i.e., MSD is actually \( \alpha^2 \)). With decreasing temperature, the MSD decreases as a result of the decreasing thermal motions of the water molecules confined in the C-S-H gel; so does the value of \( \alpha \). Furthermore, we can confirm that the value of \( \alpha \) agrees with data reported in the literature [12,22] both from neutron scattering experiments on cement pastes and molecular simulations on mineral equivalents. The magnitude of \( \alpha \) is about 0.7–1.2 Å for C-S-H gel with 30% water content, well in agreement with the square root of the MSD measured for a cement paste with no additive by our previous QENS experiment at the same temperatures [12]. In our case, we found that \( \alpha^2 \) is about 1.3 Å\(^2\) at 280 K, which is the same quantity, within the error bars, as the MSD calculated by molecular simulation on a C-S-H model system [22].

A fluid under normal condition can be described by the hydrodynamic theory. Consequently, a particle of radius \( R \) suspended in the fluid satisfies the well-known Stokes-Einstein relation \( D = k_B T/6\pi \eta R \), where \( D \) is the self-diffusion constant of the particle and \( \eta \) the viscosity of the fluid. Since \( \eta \) is proportional to the relaxation time (\( \tau \)) according to Maxwell’s viscoelastic theory, the product \( D(\tau)/T \) should be a constant independent of the temperature \( T \). In a supercooled liquid, due to the occurrence of the dynamic heterogeneity the above-mentioned product can vary with temperature. This is called the violation of the Stokes-Einstein relation. Chen et al. [15] have shown experimentally the equivalence of the violation of the Stokes-Einstein relation with the dynamic crossover phenomenon in the case of supercooled water confined in MCM-41. At higher temperatures, the product \( D(\tau)/T \) is indeed a constant in the confined water. However, when the confined water approaches the crossover temperature from above, the product \( D(\tau)/T \) starts to deviate from the constant value (increasing very rapidly). We now show that this violation of the Stokes-Einstein relation can also be observed in the case of water confined in the C-S-H gel with 30% water content. However, the violation is not present in the case of the two lower water contents.

Figure 4 shows the comparison of the degree of violation of the Stokes-Einstein relation between the water confined in C-S-H (I) gel and the water confined in MCM-41 [15]. From Fig. 4(a) we can see that the quantity \( D(\tau)/T \) is sensitive to temperature only for C-S-H gel with 30% water level. In this case, the quantity \( D(\tau)/T \) is almost constant at the higher temperature (260–280 K), while it increases noticeably as the temperature approaches the 230 K from above. In Fig. 4(b), the value of \( D(\tau)/T \) for water confined in MCM-41 is calculated according to the experimental data of the \( D \) and \( \tau \) fitted by the empirical Vogel-Fulcher-Tamman (VFT) law, which are respectively, \( 1/D = 2.4 \exp[332/(T - 187)] \) (10\(^7\) s/m\(^2\)) and \( \tau = 0.077 \exp[313/(T - 200)] \) (ps). Figure 4(b) shows that the quantity for water confined in C-S-H gel at 30% water content and water confined in MCM-41 have the same increasing trend as the temperature approaches the crossover temperature of 225 K, although the increase is slower for water confined in C-S-H gel. This might indicate that the Stokes-Einstein relation is violated as temperature approaches 225 K for both cases. This breakdown of the Stokes-Einstein relation in confined water is physically equivalent to having a dynamic crossover temperature at around 230 K. A further experiment on the same sample in a broader temperature range is planned in order to clearly define the presence of the dynamic crossover in C-S-H (I) gels.

V. CONCLUSION

In this paper, we study dynamics of water confined in C-S-H (I) gel with contents varying from 10% to 17% and 30% and in the temperature range from 280 to 230 K by the QENS technique. The data were analyzed according to the RCM model. We show that the measured low-\( Q \) (\( Q \ll 1.1 \) Å\(^{-1}\)) QENS spectra agree very well with the RCM
model. By the RCM model fitting, four parameters, which are the stretch exponent $\beta$, the self-diffusion constant $D$, the average translational relaxation time $\langle \tau \rangle$, and the temperature dependence of the confined radius $\alpha$ (approximately the same as MSD) given by the relation $p(Q) = \exp[-(\alpha Q)^2/3]$ have been extracted. The obtained results allowed us to detail the transport phenomena in a synthetic C-S-H (I) phases, which is important in describing the porous structure of C-S-H gel containing different pores occupied by water molecules in the C-S-H gel samples with different water content. In particular, the parameter $\beta$ decreases with the increasing water content, which means that the uniformity of the pores occupied by the water molecules confined in the C-S-H gel decreases with increasing water content. Among the three different water levels, $D$ and $\langle \tau \rangle$ are sensitive to the measured temperature only for the C-S-H gel at the highest water content (i.e., 30%). The temperature dependence of $\alpha$ increases with the increasing water content showing that the water molecules confined in C-S-H gel exist not only in the finest porosity, IGP, but also in SGP and LGP. The parameter $\alpha$ decreases with the decreasing temperature in all cases as a result of the decrease of the MSD of water molecules confined in C-S-H gel. The magnitude $\alpha^2$ in the temperature ranges 280 to 230 K for C-S-H gel with 30% water content has the same value of the MDS measured by a previous QENS experiment on a curing cement paste.

Furthermore, we show the product $D(\tau)/\tau$ for C-S-H gel with 30% water content is constant within the error bars at higher temperatures 260–280 K, while it seems to increase as the temperature approaches 230 K from above. This might indicate that the Stokes-Einstein relation is violated as temperature approaches 230 K for this specific sample. This breakdown of the Stokes-Einstein relation has been shown to be physically equivalent to having a dynamic crossover temperature at around 230 K in the case of supercooled water [15].

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