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

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Dynamic behavior of hydration water in Calcium-Silicate-Hydrate gel: a QENS investigation

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Abstract

The translational dynamics of hydration water confined in calcium-silicate-hydrate (C-S-H) gel was studied by quasi-elastic neutron scattering spectroscopy (QENS) in the temperature range from 280K to 230K. The stretch exponent β, the self-diffusion constant D, the average translational relaxation time $<\tau>$, 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 relaxing cage model (RCM). 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 decreases with increasing water contents, while α increases. The values of D and $<\tau>$ 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 Stoke-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 recent accepted model is Jennings's colloidal model (CM-II) [3]. This one is a hybrid model and able to explain several experimental evidences such as those derived from scattering measurements [1] and sorption isotherms 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 disk-like object, whose thickness is around 4 nm. Moreover, the globule presents a layered internal structure which defines interlamellar spaces and in very small cavities (intraglobular pores, IGP) 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 (SGP), 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) being a smaller amount present in the intralayer small cavities (i.e. IGP) proper of the internal structure of the globules [5]. Although 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 dynamic 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 quasi-elastic 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 water/weight 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 25K to 300K. 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 230K, 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 to 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 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 to water confined in IGP and water interacting with the C-S-H surface, while process 3 to water that behaves in a cooperative way). But the investigation only focus 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 CSH gel with different water content, both low and high? Could the dynamic crossover in the confined water be observed also in the pure CSH gel at the temperature near 230K for all water contents? To answer these questions, we have done a further study about the dynamics of hydration water confined in pure CSH gel by a QENS experiment.

The present investigation aims to study the temperature dependence of 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 QENS technique using the BASIS spectrometer [14] at ORNL. The investigated temperature range is from 280K to 230K. 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 230K 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 230K.

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 280K to 230K. BASIS is well suited for probing diffusive and relaxation motions for confined and hydration water [16, 17]. In the quasi-elastic 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 2.08meV. The initial neutron energy is determined from the neutrons time-of-flight. The silicon analyzer crystals cover about 2.0 sr (16% of 4π). Neutron 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 as 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 29 mm inner diameter in order to fully intercept the neutron beam, which is about

30×30 mm^2 at the sample position. The resolution function was measured at 3.5K 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 σ_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 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.

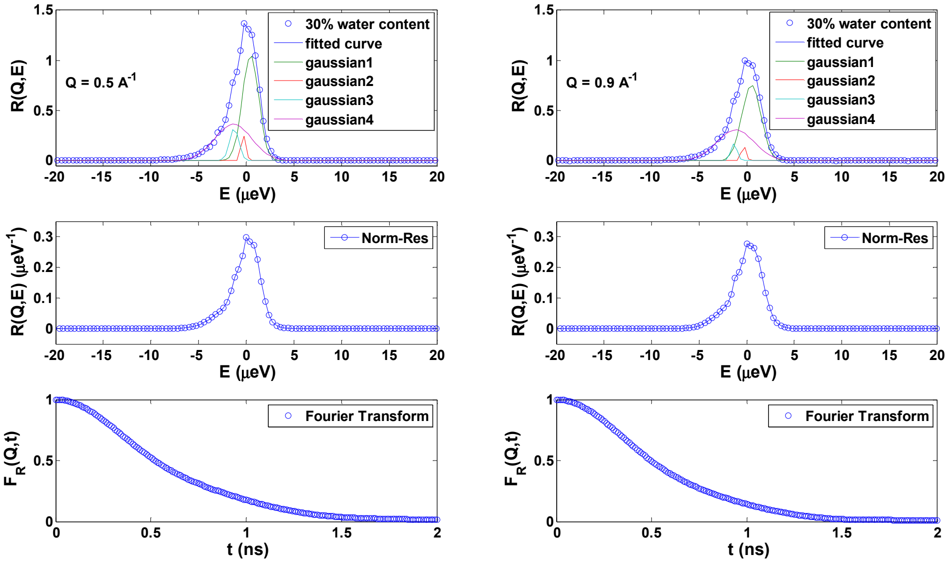

Figure 1. The upper panel shows the measured resolution function (open circles on the top graph) fitted by four Gaussian functions. The middle panel shows the corresponding normalized measured and fitted resolution function. The third panel shows the Fourier transform $F_R(Q,t)$ of the normalized resolution function. The resolution functions shown are for C-S-H gel with water level 30% at Q=0.5 and 0.9 $Å^{-1}$.

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 280K to 230K. The measured neutron intensity of each sample was obtained by subtracting the measured data of the empty cell holder for descripting the contribution only from the hydration water confined in CSH gel.

Synthetic C−S−H was prepared by hydrating pure tricalcium silicate (C_3S) in an excess of decarbonated water. A chemically pure batch of C_3S was obtained from CTG-Italcementi (Bergamo, Italy) as a gift. The specific surface area of C_3S as detected by N_2 sorption isotherms (BET) resulted 0.65 m^2/g while the mean radius was 4.66 µm. C−S−H was prepared by mixing 4 g of C_3S with 1150 g of distilled water. The excess of water in respect of C_3S was essential to minimize the Portlandite ($Ca(OH)_2$) co-precipitation. The resulting C_3S/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 polydisperse C−S−H phase, which is usually refereed as C−S−H (I) [18]. The dispersion was then filtered under a N_2 atmosphere to avoid carbonation and the obtained solid was dried in an oven at 60 °C for about three hours. The resulting gel was dried to the desired water content using a vacuum oven operating under a N_2 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. Thermo gravimetric analysis (TGA) was performed to check the final water, Portlandite and calcium carbonate contents. $Ca(OH)_2$ and $CaCO_3$ resulted in the range 5−10% in respect of 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 to 1000 °C under a constant flux of pure N_2 (100 mL/min). 10%, 17%, and 30% water contents (wt%=grams of water /grams of dry cement) were achieved at the end of the drying process. Table I gives a list for all the samples of C-S-H gel with different water content studied in the present work.

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

Sample name	Measured temperature （K）
10% water content	280，270，260，250，240，230
17% water content	280， 270， 260， 250， 240
30% water content	280，270，260，250，240，230

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 MD 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 equation (2) represents an elastic neutron scattering (ENS) and the second part quasi-elastic neutron scattering (QENS).

$$\begin{aligned} I(Q,E) &= \left[p(Q)\delta(E) + \left(1 - p(Q)\right) \cdot \mathcal{F}\{F_S(Q,t)\}\right] \otimes R(Q,E) \\ &= S_H(Q,E) \otimes R(Q,E) \end{aligned} \tag{2}$$

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 confined diffusion. The factor can then be expressed as $p(Q) = \exp\left[-(\alpha Q)^2/3\right]$, in the case of a spherical confinement geometry of radius α. This is sometimes called the elastic 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 $\mathcal{F}\{\mathrm{F_S(Q,t)}\}$ 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 3.5K 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 $Q \leq 1.1$ Å^{-1}, the rotational contribution becomes negligible. The SISF can be obtained by

$$F_S(Q,t) \approx F_T(Q,t) = F^v(Q,t)\exp\left(-\left(t/\tau_T(Q)\right)^{\beta}\right) \tag{3}$$

Where $\tau_T(Q) = \tau_0(aQ)^{-\gamma}$, a=0.5Å [19], $F^v(Q,t)$ 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 $F^v(Q,t)$ tends to it's asymptotic value $F^v(Q,t) = \exp(-Q^2a^2/3) \doteq 1$. The stretch exponent $\beta \leq 1$ takes into account the slow relaxation processes associated with the deviation from a simple exponential decay (β=1) which is found in the event of Debye-like relaxation process, and $\tau_\mathrm{T}(\mathrm{Q})$ is the Q-dependent relaxation time.

According to RCM, the four temperature dependent parameters $p(Q)$, β, τ_0, and γ 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 $\langle\tau\rangle = \left(\frac{\tau_0}{\beta}\right)\Gamma\left(\frac{1}{\beta}\right)$, the parameter α can be fitted using equation $p(Q) = \exp\left[-(\alpha Q)^2/3\right]$. Moreover, in the case of an exponential relaxation as a result of a continuous diffusion, one has $\mathrm{F_S(Q,t)} = \exp\left(-\mathrm{DQ^2t}\right)$, where D is the self-diffusion coefficient. In RCM, both the exponents β and γ in equation (3) are Q dependent [21], and in the hydrodynamic limit (Q→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:

$$\because \exp\left(-\left(\frac{t}{\tau_T(Q)}\right)^{\beta}\right) \overset{Q\to 0,\beta\to 1,\gamma\to 2}{=} \exp\left(-\left(\frac{t}{\tau_0}\right)(aQ)^2\right) = \exp\left(-DQ^2 t\right)$$

$$\therefore D = \frac{a^2}{\tau_0} = \frac{2.5}{\tau_0(ns)} \qquad (10^{-8}\, cm^2/s) \tag{4}$$

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 RCM equation (2) by inverse Fourier transforming both sides:

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

$$= \mathrm{F_H(Q,t)}\cdot \mathrm{F_R(Q,t)} \tag{5}$$

where $I(Q,E)$ is the measured neutron spectral intensity, and the $R(Q,E)$ is the Q-dependent energy resolution function. From equation (5), we can directly fit the measured neutron intensity in time domain, $\mathcal{F}^{-1}\{I(Q,E)\}$, by multiplying the RCM intermediate scattering function, $F_H(Q,t)$, to the resolution function in time domain $F_R(Q,t) = \mathcal{F}^{-1}\{R(Q,E)\}$.

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 230K for two distinct Q values, 0.5 Å^{-1} 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 dash 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 μeV 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 non-linear least squares method for each Q and each sample, respectively.

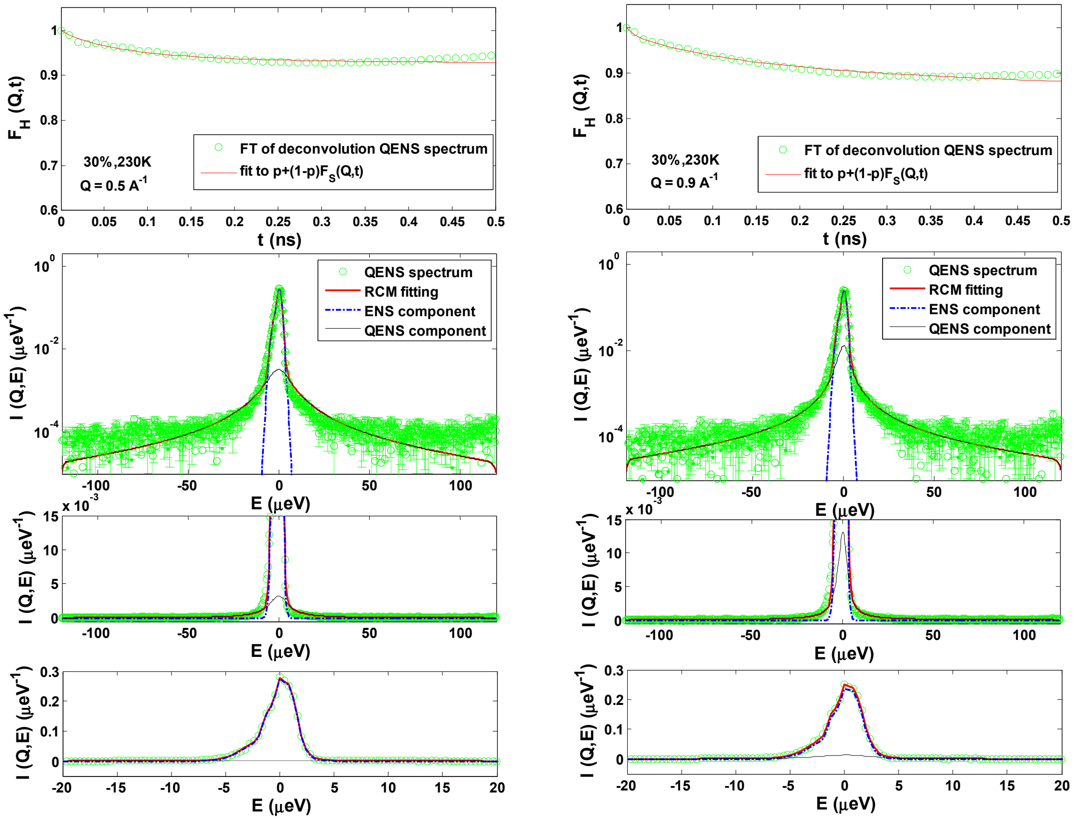

Figure2. Examples of the RCM model fitting of measured QENS spectra for the C-S-H gel with 30% water content at temperature 230K. Left-hand side panels refer to the neutron spectrum at Q = 0.5 Å^{-1}, and right-hand side panels refer to Q = 0.9 Å^{-1}. In each panel the top figure is the Fourier Transform $F_H(Q,t)$ of the deconvoluted measured neutron spectrum namely, the Fourier transform of the incoherent dynamic structure factor $S_H(Q,E)$ of typical hydrogen atom. The lower three figures are respectively the fitted curves of the measured neutron spectrum represented with different scales.

Figure 3 gives the extracted four parameters: stretch exponent, β, the self-diffusion coefficient, D, the average translational relaxation time, <τ>, and the temperature dependence of confinement radius α in $p(Q) = \exp\left[-(\alpha Q)^2/3\right]$ for all the investigated cases. From Figure 3(a), we can see that β 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 is more uniform (the value of β is larger). This is because water molecules confined in the C-S-H gel with lower water level mainly resides inside the globules, namely in the interlamellar spaces and in the IGP [3,4]. With increasing water level, hydration water molecules confined in the C-S-H gel tends to explore also the pores greater than 1 nm (i.e. SGP and LGP). So the space explored by the confined water molecules is less uniform and the value of β decreases. From Figure 3(b) and (c) we can see that D and <τ> retain almost the same values within error bars in the 10% and 17% water content cases. On the contrary D decreases and <τ> increases while decreasing temperature for the C-S-H gel at 30% water level. This indicates that parameters D and <τ> are sensitive to temperature only in the 30% water level case. From Figure 3(d) we can see that the parameter α increases with increasing water content, which is correspondent 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 Figure 3(d) we can also see that in all cases α 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 $p(Q) = \exp\left[-Q^2\langle r^2\rangle/3\right]$ [21], where $\langle r^2\rangle$ is the mean square displacement, MSD, of the confined water molecules (i.e. MSD is actually α^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 α. Furthermore, we can confirm that the value of α 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 α is about 0.7-1.2Å 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 α^2 is about 1.3 Å^2 at 280K, which is the same quantity, within the error bars, as the MSD calculated by molecular simulation on a C-S-H model system [22].

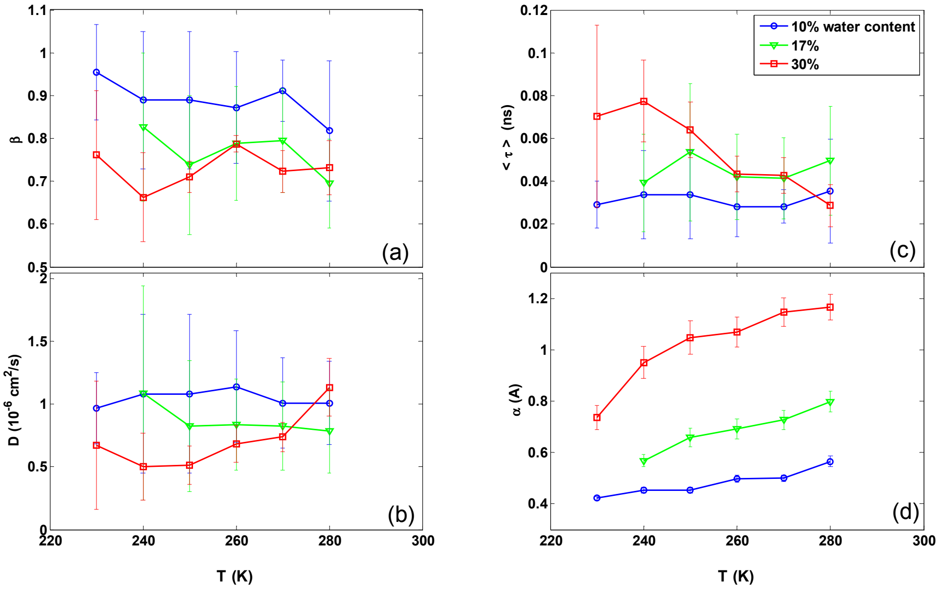

Figure 3. Temperature dependence of Values of the four extracted parameters for hydration water confined in C-S-H gel using RCM: (a) the stretch exponent β; (b) the self-diffusion coefficient D; (c) the average translational relaxation time <τ>; and (d) the parameter α defined by $p(Q) = \exp\left[-(\alpha Q)^2/3\right]$. Water content of C-S-H gels is respectively 10% (shown in blue circle), 17% (green triangle) and 30% (red square).

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 η the viscosity of the fluid. Since η is proportional to the relaxation time $<\tau>$ according to Maxwell's visco-elastic 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 Stokes-Einstein relation. In a paper by Chen et. al. [15], it has been shown experimentally the equivalence of the violation of 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 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 Stoke-Einstein relation between the water confined in C-S-H (I) gel and the water confined in MCM-41 [15]. From Figure 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 (260-280K), while increases noticeably as the temperature approaches the 230K from above. On Figure 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 225K, 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 approaching 225K for both cases. This breakdown of the Stokes-Einstein relation in confined water is physically equivalent to having a dynamic crossover temperature at around 230K. A further experiment on the same sample in a broader temperature range is in forecast in order to clearly define the presence of the dynamic crossover in C-S-H (I) gels.

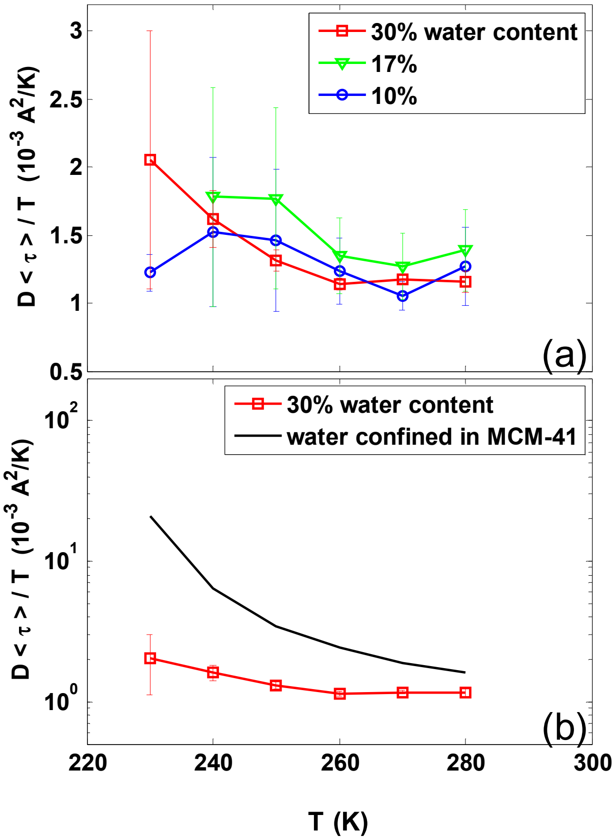

Figure 4. Comparison of the degree of violation of the Stoke-Einstein relation between water confined in C-S-H gel and in MCM-41 [15]. The upper panel (a) shows the value of product D<τ>/T for C-S-H gels with three different water contents. The lower panel (b) shows the comparison between water confined in C-S-H gel with 30% water content and water confined in MCM-41.

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 280K to 230K by QENS technique. The data were analyzed according to RCM model. We show that the measured low-Q ($Q \leq 1.1$ Å^{-1}) QENS spectra agree very well with the RCM model. By the RCM model fitting, four parameters, which are the stretch exponent β, the self-diffusion constant D, the average translational relaxation time <τ>, and the temperature dependence of the confined radius α (approximately the same as MSD) given by the relation $p(Q) = exp[-(\alpha Q)^2/3]$, have been extracted. The obtained results allowed to detail the transport phenomena in a synthetic C-S-H (I) phases, which is important to describe 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 β 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 <τ> 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 α 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 α 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 of α^2 in the temperature ranges 280K to 230K 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<τ>/T for C-S-H gel with 30% water content is constant within the error bars at higher temperatures 260-280K, while it seems to increase as temperature approaches 230K from above. This might indicate that the Stokes-Einstein relation is violated as temperature approaches 230K 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 230K in the case of supercooled water [15].

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