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# Finite-size effect at both high and low temperatures in molecular dynamics calculations of the self-diffusion coefficient and viscosity of liquid silica

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## Abstract

Molecular dynamics simulations are performed at 6543 and 3310 K to investigate how the viscosity and self-diffusion coefficient scale with system size in the liquid BKS silica system. We find that at high temperature the finite-size effect on shear viscosity is negligible. However, the size effect on the diffusion coefficient still exists and scales linearly with  $1/N^{1/3}$ , where  $N$  is the total number of particles in the system. At low temperature, the size effect on the viscosity becomes stronger than that on diffusion, and the logarithm of the viscosity and the diffusion coefficient scale linearly with  $1/N$ . These results are consistent with previous theoretical developments, and demonstrate that the finite-size effect should be considered in both high- and low-temperature molecular dynamics simulations of liquid silica.

## 1. Introduction

To understand the dynamics of a liquid, the viscosity and self-diffusion coefficient of the liquid are frequently calculated using molecular dynamics simulations and compared with other thermodynamic and dynamic properties. For example, the self-diffusion coefficient is often combined with the configurational entropy to test the validity of the Adam–Gibbs equation and to establish a relation between the kinetics and the thermodynamics of a supercooled liquid [1–3]. Decoupling of diffusion from that predicted by the Stokes–Einstein equation is used as an indication of the dynamic heterogeneity in a supercooled liquid [4, 5]. More recently, the diffusion coefficient has been connected with the structural order parameters to understand the anomalous behaviours of water and silica [6, 7]. For all these investigations, an accurate calculation of the viscosity and self-diffusion coefficient is essential.

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One of the factors that influence the accuracy of the calculation is the dependence of the calculated properties on the size of the system. The finite-size effect exists in many different systems with its magnitude dependent on temperature, density, and the type of the system. For example, a very strong size effect is found in a high-density soft-core mixture with the  $\alpha$  structural relaxation times being different by almost 20-fold for two systems containing 108 and 10 000 particles, respectively [8]. For comparison, in a Lennard-Jones system, the size effect on the relaxation time occurs only for systems with  $N \leq 40$  [9], where  $N$  is the total number of particles in the simulated system. The ratio of the diffusion coefficient of the system with  $N = 40$  to that with  $N = 1000$  is  $\sim 0.1$  [10], showing thus a very strong size effect. However, for the same system but with  $N > 60$ , the size effect on diffusion remains within  $\sim 20\%$  at different temperatures [10].

To understand better the mechanisms for causing these size effects and then to be able to predict their magnitudes, we need to know further how the viscosity and diffusion coefficient scale with the system size. The scaling of self-diffusion coefficient with system size has a long history of investigations [11–13]. As demonstrated by the most recent study [13], a very useful equation is derived from these investigations:

$$D_{\text{PBC}} = D_0 - \frac{k_{\text{B}} T \xi}{6\pi \eta L}, \quad (1)$$

where  $D_{\text{PBC}}$  is the diffusion coefficient calculated in a periodic boundary system,  $D_0$  that of an infinite system,  $k_{\text{B}}$  the Boltzmann constant,  $T$  the temperature,  $\eta$  the shear viscosity,  $L$  is the edge length of the simulation box, and  $\xi$  is about 2.837 297. The equation is very successful when applied to liquid water at room conditions and Lennard-Jones and hard-sphere systems [13]. For these systems, the size effect on viscosity is negligible at the temperature and density of the investigation. However, it is also found that the diffusion coefficient of liquid butane near its boiling point scales linearly with  $1/N$  [14], and that at a lower density and a higher temperature it scales with  $1/N^{1/3}$  [15]. These results indicate that another form of the scaling function is still possible.

The purpose of the present study is to investigate the finite-size effect in the liquid silica system. Previous studies show that at 3760 K the difference of the relaxation time between the systems with  $N = 336$  and 8016 reaches a factor of 1.5. At 1600 K, it increases up to  $\sim 4.0$  [16]. More recently, the  $\alpha$  relaxation time of liquid silica containing 99 particles is found to be about three times larger than that having 1000 particles [17]. However, from these investigations, we do not know how the self-diffusion coefficient and viscosity scale with system size and whether temperature has an effect on the form of the scaling function. These issues are tackled in the present study. In the next section, technical details of our molecular dynamics simulations are given. In the third section, simulation results, including the self-diffusion coefficient, the viscosity, the magnitude of the size effect, and the functions describing the scaling of diffusion coefficient and viscosity with system size are presented. In the fourth section, results from molecular dynamics calculations and a theoretical derivation are compared. Finally, the conclusions are drawn in section 5.

## 2. Simulation details

BKS silica [18] is selected for our simulations. The original model is modified according to [19] to overcome its inflection at short ion–ion distances, resulting in a final potential model of the form

$$U(r) = -\frac{A}{r^6} + B \exp(-Cr) + D \left[ \left( \frac{E}{r} \right)^{30} - \left( \frac{E}{r} \right)^6 \right], \quad (2)$$

where  $r$  is the ion–ion separation distance. The parameters  $A$ ,  $B$ , and  $C$  are taken from the original publication [18], and in our case we use  $D_{\text{O-O}} = 1.979\,28 \times 10^{-10}$  eV,  $D_{\text{Si-O}} = 1.398\,56 \times 10^{-9}$  eV,  $E_{\text{O-O}} = 3.411\,653$  Å, and  $E_{\text{Si-O}} = 2.288\,769$  Å.

Molecular dynamics simulations are performed in the  $NVE$  ensemble using the program MOLLY [20]. The choice of the ensemble is appropriate for our study as it avoids the artificial dynamics [16]. Two series of simulations are performed; one is at the temperature of 6543 K, and another at 3310 K. The temperatures of the simulations in the same series are fixed using our method [21]. The number of particles in the simulated system varies from 2400 to 30 for the high-temperature runs, and from 2400 to 180 for the low-temperature runs. To achieve better statistics, simulations are repeated at each system size. For the smallest system ( $N = 30$ ), the number of repeated simulations reaches 31. For the largest systems ( $N = 2400$ ), the minimum number of repeated simulations is five.

To investigate specifically the size effect and prevent the influence from other factors, the simulation parameters are kept identical. Thus the density is always at  $2.37$  g cm $^{-3}$ , the real space cut-off distance is  $10$  Å, the reciprocal space cut-off distance is  $2.3026$  Å $^{-1}$ , the  $\alpha$  parameter for the Ewald sum is  $0.3393$ , and the timestep is  $0.2$  fs at high temperature and  $1$  fs at low temperature. The real space cut-off distance is larger than previously used values [3, 22], so the shifted force method is not employed. With these parameters, the internal energy remains constant within  $10^{-5}$ . The equilibration time is  $0.2$  ns for the high-temperature simulations and  $4$  ns for the low-temperature ones. The latter is about  $20$  times larger than that for the stress auto-correlation function to reach zero ( $\sim 200$  ps).

Stress data accumulated at every step for a period of  $1$  ns at high temperature and  $14$  ns at low temperature are used to calculate the shear and longitudinal viscosity using the Green–Kubo equation [23]. Particle configurations are dumped every  $0.3$  ps during the accumulation time. More than  $10\,000$  configurations are used to calculate the self-diffusion coefficient using the Einstein equation [23].

### 3. Results

At 6543 K, the studied system sizes are  $N = 2400, 300, 90,$  and  $30$  (table 1). As shown in figure 1(A), the lines for shear and longitudinal viscosity are flat, indicating a negligible size effect. However, the self-diffusion coefficients decrease notably with decreasing system size (figure 1(B)). Thus at high temperature, the size effect on diffusion is not negligible and is larger than that on viscosity.

We find that  $D$ ,  $\ln(D)$ , and  $1/D$  all scale non-linearly with  $1/N$ , but linearly with  $1/N^{1/3}$ . The fitted functions for the upper and lower lines in figure 1(B) are

$$\begin{aligned} D_{\text{O}} &= -\frac{239(29)}{N^{1/3}} + 227(5) \\ D_{\text{Si}} &= -\frac{243(30)}{N^{1/3}} + 186(4), \end{aligned} \quad (3)$$

where the numbers in the parentheses give the uncertainties of the corresponding parameters estimated using the chi-square fitting [24]. If we use an average shear viscosity value of  $0.26 \times 10^{-2}$  Pa s (table 1), Equation (1) predicts a value of  $217$  for the slopes of equation (3). The value is slightly smaller than the actual fitting parameters, but is still within the statistical uncertainty of the fitting parameters. The consistency thus extends the validity of equation (1) to high-temperature liquid silica system. We note that equation (1) predicts that size effect will become increasingly larger with the increase of temperature and density. Together with the versatile applicability of equation (1) to many different types of systems [13], we see that the

**Table 1.** Simulation results. (Note: for symbols used in the table, the first column is the number of particles in the simulated system.  $T$  and  $P$  denote the temperature and the pressure;  $\eta_S$  and  $\eta_L$  are the shear and longitudinal viscosity;  $D_{Si}$  and  $D_O$  are the self-diffusion coefficients of silicon and oxygen. The same symbols are used in the text. The last column is the number of simulations repeated at each system size. The numbers after  $\pm$  are  $2\sigma$  errors with 95% confidence.)

$N$	$T$ (K)	$P$ (Pa)	$\eta_S$ ( $10^{-2}$ Pa s)	$\eta_L$ ( $10^{-2}$ Pa s)	$D_{Si}$ ( $10^{-10}$ m <sup>2</sup> s <sup>-1</sup> )	$D_O$ ( $10^{-10}$ m <sup>2</sup> s <sup>-1</sup> )	Runs
2400	6543 $\pm$ 1	-1531 $\pm$ 4	0.247 $\pm$ 0.007	0.665 $\pm$ 0.019	168 $\pm$ 3	210 $\pm$ 4	5
300	6544 $\pm$ 2	-1594 $\pm$ 5	0.249 $\pm$ 0.004	0.655 $\pm$ 0.014	145 $\pm$ 6	189 $\pm$ 6	10
90	6542 $\pm$ 1	-2066 $\pm$ 8	0.270 $\pm$ 0.004	0.736 $\pm$ 0.014	131 $\pm$ 6	171 $\pm$ 7	20
30	6542 $\pm$ 3	-3036 $\pm$ 13	0.274 $\pm$ 0.003	0.624 $\pm$ 0.010	110 $\pm$ 9	152 $\pm$ 7	31
2400	3310 $\pm$ 1	-1724 $\pm$ 9	84 $\pm$ 7	243 $\pm$ 29	0.44 $\pm$ 0.02	0.74 $\pm$ 0.01	7
600	3310 $\pm$ 3	-1700 $\pm$ 21	103 $\pm$ 7	305 $\pm$ 25	0.39 $\pm$ 0.02	0.67 $\pm$ 0.03	13
300	3310 $\pm$ 2	-1591 $\pm$ 22	129 $\pm$ 7	353 $\pm$ 26	0.33 $\pm$ 0.02	0.58 $\pm$ 0.02	24
180	3311 $\pm$ 6	-1596 $\pm$ 45	186 $\pm$ 22	547 $\pm$ 72	0.27 $\pm$ 0.02	0.48 $\pm$ 0.03	18

finite-size effect should always be considered when the diffusion coefficient is being calculated and especially at high temperatures.

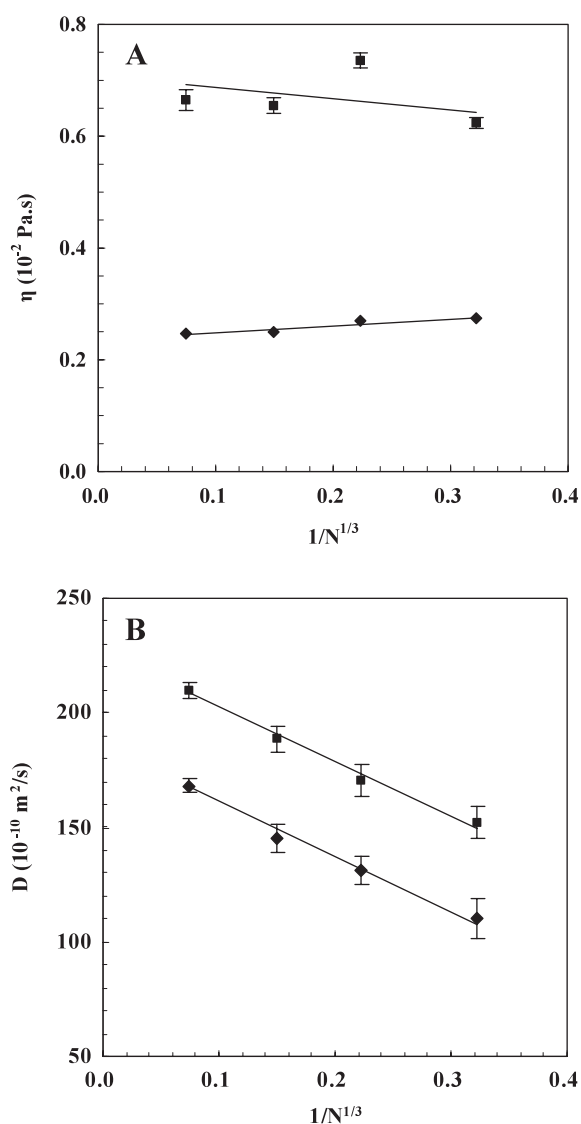
To give an idea of the magnitude of the size effect at high temperature, the intercepts in equation (3) are used to represent the properties of the bulk system. The magnitude of the size effect, defined as  $(D_N - D_b)/D_b$  with  $D_b$  the diffusion coefficient of the bulk system and  $D_N$  that of a system containing  $N$  particles, is about  $-10\%$  at  $N = 2400$  and about  $-20\%$  at  $N = 300$ . This stresses again that the size effect should be considered even in high-temperature simulations.

Now we look at the results from the low-temperature simulations. At 3310 K, the studied system sizes are  $N = 2400, 600, 300,$  and  $180$  (table 1). From  $N = 2400$  to  $300$ , the relative change is  $54\%$  for the shear viscosity and  $45\%$  for the longitudinal viscosity, and is  $-26\%$  for  $D_{Si}$  and  $-22\%$  for  $D_O$ . Thus at low temperature the size effects on viscosity and on diffusion coefficient both exist and also are stronger than those at high temperature. In addition, the size effect on viscosity becomes stronger than that on diffusion, in contrast with the high-temperature case.

We find that at low temperature,  $\ln(\eta)$  and  $\ln(D)$  scales linearly with  $1/N$  (figure 2). This is different from the high-temperature case, and implies that the mechanism for causing the size effect is different in the two cases. In other words, there exists more than one mechanism for causing the size effects we see in molecular dynamics simulations. Furthermore, we find that equation (1) becomes invalid at low temperature. The mechanism involved in the equation may still be operating at low temperature, but has a much less dominant role. By putting temperature and viscosity values at the two temperatures into equation (1), we can see that the slope of equation (1) is reduced by almost 1000-fold from high to low temperature.

The straight lines in figure 2 have the following functional forms:

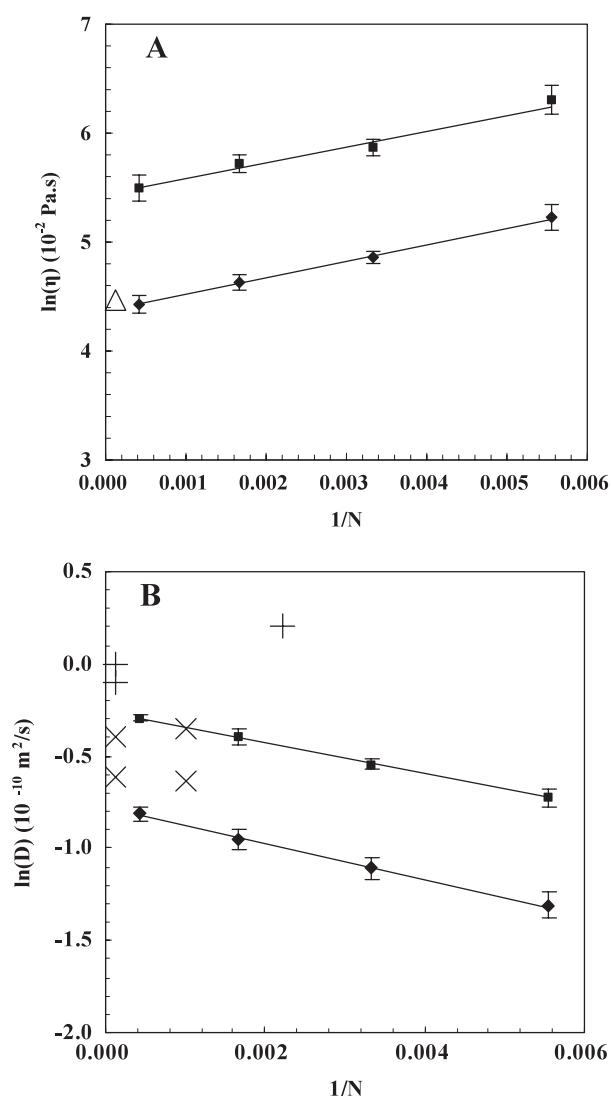
$$\begin{aligned}
 \ln(\eta_S) &= \frac{151(25)}{N} + 4.37(0.07) \\
 \ln(\eta_L) &= \frac{144(31)}{N} + 5.44(0.09) \\
 \ln(D_{Si}) &= -\frac{97(14)}{N} - 0.78(0.04) \\
 \ln(D_O) &= -\frac{84(8)}{N} - 0.26(0.02).
 \end{aligned} \tag{4}$$



**Figure 1.** (A) Variation of  $\eta$  with  $1/N^{1/3}$  at 6543 K. Filled squares denote the longitudinal viscosity, and filled diamonds the shear viscosity. (B) Variation of  $D$  with  $1/N^{1/3}$  at 6543 K. Filled squares represent  $D_O$ , and filled diamonds  $D_{Si}$ . In both figures, the error bars on data points mark  $\pm 2\sigma$  errors with 95% confidence. The error bars for  $\eta_S$  are smaller than the size of the symbols. The straight lines are the least chi-square fits [24] of the data.

These relations permit a comparison of results from investigations at the same state point but using different system sizes. In figure 2(A), the shear viscosity of a large system ( $N = 8016$ ) [22] is consistent with our results. However, the diffusion data (figure 2(B)) show large scatterings among different studies. Our data, extrapolated to  $N = 8016$  using equation (4), is different from [22] by  $\sim 15\%$ . However, the largest discrepancy reaches a factor of  $\sim 2$  when our data are compared with those interpolated from [7].

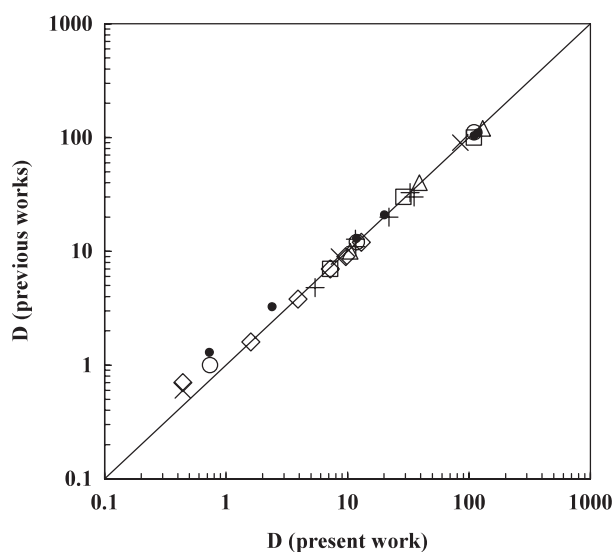
To investigate whether the discrepancy extends to other state points, we calculate the diffusion coefficients at many different state points and compare them with those from previous



**Figure 2.** (A) Linear relations between  $\ln(\eta)$  and  $1/N$  at 3310 K. The upper line is for the longitudinal viscosity and the lower line for the shear viscosity. The open triangle is interpolated from figure 10 of [22], which uses 8016 ions in the simulations. (B) Linear relations between  $\ln(D)$  and  $1/N$  at 3310 K. The upper line is for  $D_O$  and the lower line for  $D_{Si}$ . The crosses are for  $D_{Si}$  interpolated from figure 9 of [3], figure 8 of [22], figure 1 of [25], and figure 3 of [26]. The pluses are for  $D_O$  taken from figure 8 of [22] and figure 1 of [25], figure 4 of [7]. Again, the error bars represent  $\pm 2\sigma$  errors with 95% confidence.

investigations (figure 3). Obviously, they all agree very well in the high-diffusion-coefficient range, but in the low-diffusion-coefficient range scatterings appear among all investigations. The discrepancy may arise from the difference in the technical details of these simulations, such as the equilibration and accumulation times, the number of runs repeated at each point, etc, and re-emphasizes the observation that the diffusion coefficient of liquid silica is difficult to calculate in the low-temperature and low-density region [7].

Similarly to the high-temperature case, the intercepts in equation (4) are used to estimate the size effect at different  $N$ . It is clear from table 2 that the size effect reduces rapidly with the



**Figure 3.** Comparison of self-diffusion coefficients obtained in the present study with those from previous investigations. For  $D_{Si}$ , the open diamonds are interpolated from figure 9 of [3], the open squares from figure 11 of [3], the crosses from figure 8 of [22], the pluses from figure 1 of [27]. For  $D_O$ , the open triangles from figure 11 of [3], the open circles from figure 8 of [22], and the solid dots from figure 4 of [7]. The thin straight line has unit slope. All self-diffusion coefficients have the unit of  $10^{-10} \text{ m}^2 \text{ s}^{-1}$ .

**Table 2.** Magnitudes of size effect at different system sizes and temperatures. (Note: the magnitude of the size effect is defined as  $(X_N - X_b)/X_b$ , where  $X$  represents the viscosity or the self-diffusion coefficient, subscript  $N$  denotes the system containing  $N$  particles, and subscript  $b$  denotes the bulk system.  $X_b$  is taken directly from the intercepts of equations (3) and (4). Symbols used in table 2 are the same as those in table 1. The numbers in the parentheses are for the size effect at 6543 K; the rest are for that at 3310 K.)

$N$	For $\eta_S$ (%)	For $\eta_L$ (%)	For $D_{Si}$ (%)	For $D_O$ (%)
2400	6	6	-3(-10)	-4(-8)
600	30	32	-16	-13
300	63	53	-28(-22)	-25(-17)
180	136	138	-41	-37

increase of system size. To make the size effect on diffusion less than 10% the system needs to contain more than 1500 particles. For comparison, more than 2100 particles are needed to make the size effect on viscosity less than 10%. In table 2, the magnitudes of the size effect at high temperature are also given. This shows clearly again that the size effect should be considered at both high and low temperatures if the diffusion coefficient is being calculated.

The above results demonstrate that temperature has a great influence on the functions describing the scaling of dynamic properties with system size.

#### 4. Discussion

The mechanism for causing the size effect can be attributed to the momentum conservation term at normal and higher temperatures [13]. At lower temperatures, several mechanisms



have been proposed to explain the size effect in molecular dynamics simulations, such as the disturbances that leave and re-enter the simulation box [28, 16], the existence of the dynamical heterogeneity [8], and the limited number of inherent structures in finite systems [9]. Recently, by combining the potential energy landscape formalism and the Adam–Gibbs equation, a formula of the following form is proposed [29]:

$$\ln(\eta/\eta_\infty) = \frac{\varepsilon}{T\alpha Nk_B(1 - \frac{T_K^2}{T^2})}, \quad (5)$$

where  $\eta$  is the viscosity,  $\eta_\infty$  the viscosity at infinite temperature,  $T_K$  the Kauzmann temperature,  $\varepsilon$  the parameter in the Adam–Gibbs equation,  $N$  the number of particles, and  $\alpha$  determines how the number of inherent structures scales with the number of particles in a system. We find the formula is very consistent with observations made in the present and previous size-effect studies in the following aspects.

- (1) The linear relation between  $\ln(\eta)$  and  $1/N$  we find in the present study in the low-temperature simulations is consistent with equation (5), thus supporting the proposal that the size effect is closely related to the limited number of inherent structures in finite systems [9].
- (2) In the low-temperature range, equation (5) predicts that a reduction of temperature increases the slope of  $\ln(\eta) \sim 1/N$  and results in a larger size effect. This is again consistent with previous observations [8, 16], where a much larger size effect is found at low temperatures. It will be interesting to see whether the increase of size effect follows closely the prediction of equation (5). This may in turn provide a check for the validity of equation (5) or other alternative models developed in [29].
- (3) Because a strong system has a small  $\alpha$  and a fragile system a large  $\alpha$  [29], equation (5) predicts that a strong system will have a larger size effect and a fragile system a smaller one. The observation made in [16] between low- and high-density silica is consistent with this prediction.

The above discussion indicates that the combination of the potential energy landscape formalism and the Adam–Gibbs equation has the potential to explain the phenomena observed in size-effect studies at low temperatures.

For practical applications, it is expected from equations (1) and (5) and the present results that the size effect on the diffusion coefficient will increase towards both high and low temperatures. However, the size effect on viscosity will only increase towards low temperature.

## 5. Conclusions

Molecular dynamics simulations are performed at a high and a low temperature to investigate how the self-diffusion coefficient and viscosity scale with system size and whether temperature has an effect on the scaling functions. We found the following in the liquid silica system.

- (1) For diffusion coefficients, the size effect exists at both high and low temperatures. Consequently, the size effect should be considered in both high- and low-temperature molecular dynamics simulations. More importantly, scaling functions have different forms in the two cases. At high temperature, the diffusion coefficient scales linearly with  $1/N^{1/3}$ . At low temperature, the logarithm of the diffusion coefficient scales linearly with  $1/N$ . The different scaling functions imply that the mechanisms for causing the size effect are different at the two temperatures. In other words, there exists more than one mechanism for creating the size effect we see in molecular dynamics simulations.

- (2) For viscosity, the size effect at high temperature is negligible, but becomes very strong at low temperature. Similar to the size effect on diffusion, the logarithm of viscosity scales linearly with  $1/N$  at low temperature.
- (3) The high-temperature results support the validity of equation (1) in liquid silica system at very high temperatures, but at low temperatures the equation becomes inappropriate for predicting the size effect. The low-temperature results are more consistent with a description combining the potential energy landscape formalism and the Adam–Gibbs equation.

Further investigations are clearly required. For example, parameters in equation (5) need to be further quantified to make it have a predictive power similar to equation (1). Ultimately, a combination of equation (1) and (5) may become very useful in the quantification of the size effect.

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