Molecular dynamics simulation of the Soret effect in a CaSiO₃ glass melt

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The mole fraction distributions of CaO and SiO₂ under a temperature gradient in a CaSiO₃ melt were calculated by molecular dynamics. The temperatures at the cold and hot ends of the gradient were 1800 and 2200 K, respectively, in the simulation. We used two sets of potential parameters proposed by Matsui et al. and Seo et al. The simulation results obtained with the two potentials indicate that the mole fraction of CaO in the cold region was higher than that in the hot region, and the mole fraction of SiO₂ in the hot region was higher than that in the cold region. This is qualitatively consistent with previous experimental results obtained by laser local heating inside a CaSiO₃ glass\[Shimizu et al., Optics letters, 36, 2161–2163(2010)].

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Temperature gradient-driven diffusion in multi-component systems is called the Soret effect. Although it has been over 100 years since this phenomenon was discovered, the mechanism of the Soret effect is still controversial. There have been several approaches to predict Soret coefficients, which indicate whether the components diffuse toward the hot or cold regions and the separation degree of the components between the hot and cold regions.

The Soret effect in multicomponent silicate melts that contain more than two oxides has been investigated in the field of earth sciences, and two important conclusions were drawn. First, the SiO₂ component concentrates in the hot region. Second, heavier isotopes concentrate in the cold region more than lighter isotopes. Our group has investigated the Soret effect in two-component silicate glass melts with a laser irradiation method. In a CaSiO₃ glass melt, Ca concentrated in the cold region, and Si and O concentrated in the hot region, suggesting that CaO concentrated in the cold region and SiO₂ concentrated in the hot region. However, the factors determining the diffusion direction and separation degree have not been fully elucidated. It is also important to elucidate these factors for the glass industry because the spatial homogeneity of glass composition is important for optical and thermally stable glass. In industrial glass-melting tanks and glass-melting crucibles, temperature gradients induce the Soret effect because optical absorption at the small focal spot causes a steep temperature gradient. Determining the main factors involved in the Soret effect may enable us to avoid or predict the inhomogeneity in glass tanks and damage caused by laser irradiation.

Molecular dynamics (MD) is a powerful tool for investigating the factors that dominate the Soret effect. Reith et al. revealed that heavier, smaller, and more-strongly bonded Lennard-Jones atoms concentrate in the cold region. MD enables us to change only one atomic parameter, such as weight, size, or bond strength, which is an advantage over experimental methods. MD has been used to investigate and predict the structural, elastic, thermodynamic, and kinetic properties of molten silicate, and thus is a critical tool for investigating the Soret effect in silicate melts. The Soret effect of Mg isotopes in MgSiO₃ has been investigated. However, the Soret effect of each oxide in silicate melts has not been investigated by MD simulation.

In this study, we calculated the Soret coefficients of CaO and SiO₂ in a CaSiO₃ glass melt by MD simulation. We constructed an MD program. The leapfrog Verlet algorithm with the Ewald method was used. The number of ions and time step were fixed as 1000 and 1.5 fs, respectively. The initial positions of the ions were random and the initial velocities of all ions were zero. The two-body interionic potential functions proposed by Matsui et al. and Seo et al. were used to calculate the interactions between ions in the MD simulation. The potential contains the Coulomb interaction term, short-range repulsion term, and dispersion force term as

\[V(r) = \frac{1}{4\pi\varepsilon_0} \sum_{i} \sum_{j} \left( \frac{z_i z_j e^2}{r} + f(b_i + b_j) \exp \left( \frac{a_i + a_j - r}{b_i + b_j} \right) - \frac{c_i e^2}{r^6} \right)\]

where \(z\) is the effective ionic charge, \(\varepsilon_0\) is the dielectric constant of a vacuum, \(a, b,\) and \(c\) are the characteristic parameters of each ion, \(f\) is the standard force, and \(r\) is the distance between the ion pair (i and j). Table 1 shows the potential parameters of ions in the CaSiO₃ melt.

To initialize the system, the simulation started in a homogeneous temperature distribution at 3500 K for Matsui potential and at 4000 K for Seo potential, and then the system was cooled to 2000 K. A typical configuration of ions after cooling is shown in Fig. 1(a). After the initialization, the cubic simulation box was

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divided into eight slices with equal interval along the x direction [Fig. 1(b)] and a different temperature was set in each slice. Each slice contained about 125 ions on average. To obtain a one-dimensional temperature gradient along the x direction, the temperature in the first slice was kept at 1800 K, and that of the fifth slice was kept at 2200 K with a velocity rescaling method (14) at every step. We refer to the 1800 K slice as the cold slice and to the 2200 K slice as the hot slice. The total momentums in the two slices were preserved during each velocity rescaling process. This temperature control produced an almost linear temperature gradient between the cold and hot slices. A CaSiO3 melt was used because of the availability of experimental data such as bond length, (17) density, (18) tracer diffusion coefficient, (19–21) and diffusion direction caused by the Soret effect. (9)

To check the structural and diffusion properties of the melts in our simulation and to determine the time necessary for the simulation system to reach the steady state under the temperature gradient, the MD simulations were performed at 2000 K under no temperature gradient. For the Matsui potential, we performed simulations at 9, 503, and 1019 MPa, because in the laser irradiation experiment inside CaSiO3 glass, (9) the pressure may be higher than several hundred megapascals owing to the increase in temperature over the melting point in the closed space. The calculated pair distribution functions are shown in Fig. 2(a), and the results are summarized in Table 2(a). The peak positions are in good agreement with those determined by X-ray diffraction of temperature and density measurements.

### Table 1. Interionic potential parameters used for this MD simulation.

| Potential | Ion | \( a / 10^{-10} \) m | \( b / 10^{-10} \) m | \( c / 10^{-30} \) m\(^3\) (kJ mol\(^{-1}\)) | \( z \) |
|-----------|-----|----------------------|----------------------|--------------------------------|-------|
| Matsui et al. | O | -0.945 | 1.8215 | 0.13800 | 90.61 | 0.0 |
| | Si | 1.890 | 0.7204 | 0.02300 | 49.30 | 0.0 |
| | Ca | 0.945 | 1.1720 | 0.04000 | 45.00 | 0.0 |
| Seo et al. | O | -1.266 | 2.0474 | 0.17566 | 143.94 | 0.0 |
| | Si | 2.400 | 0.8688 | 0.03285 | 47.41 | 0.0 |
| | Ca | 1.400 | 1.1500 | 0.05600 | 0.00 | 0.0 |

### Table 2. (a) Distance between the ions (\( d \)) calculated from the main peak position in the pair distribution function in the MD simulation. (b) Density (\( \rho \)) and Self-diffusion coefficient (\( D_s \))

#### (a)

| Potential | Pressure/MPa | \( d_{Ca-O} \)/nm | \( d_{Ca-O} \)/nm | \( d_{Ca-Si} \)/nm | \( d_{Si-O} \)/nm |
|-----------|--------------|-------------------|-------------------|-------------------|-------------------|
| Matsui et al. | 9 | 0.162 | 0.240 | 0.264 | 0.314 |
| | 503 | 0.162 | 0.238 | 0.264 | 0.318 |
| | 1019 | 0.162 | 0.236 | 0.264 | 0.314 |
| Seo et al. | 5 | 0.162 | 0.234 | 0.266 | 0.318 |
| Experiment \( ^{21} \) | | 0.162 | 0.236 | 0.266 | 0.321 |

\(^{21}\) Interatomic distance measured with X-ray diffraction at 1973 K in 50CaO–50SiO\(_2\). (5)

#### (b)

| Potential | Pressure/MPa | \( \rho / g \) cm\(^{-3}\) | \( D_s \)/cm\(^2\) s\(^{-1}\) | \( D_{Si-O} \)/cm\(^2\) s\(^{-1}\) | \( D_{Ca-O} \)/cm\(^2\) s\(^{-1}\) |
|-----------|--------------|-----------------|-----------------|-----------------|-----------------|
| Matsui et al. | 9 | 2.500 | \( 1.3 \times 10^{-5} \) | \( 9.1 \times 10^{-6} \) | \( 3.6 \times 10^{-6} \) |
| | 503 | 2.580 | \( 1.3 \times 10^{-5} \) | \( 1.0 \times 10^{-5} \) | \( 2.8 \times 10^{-5} \) |
| | 1019 | 2.646 | \( 1.1 \times 10^{-5} \) | \( 8.6 \times 10^{-6} \) | \( 1.9 \times 10^{-5} \) |
| Seo et al. | 5 | 2.452 | \( 4.0 \times 10^{-6} \) | \( 1.9 \times 10^{-6} \) | \( 1.0 \times 10^{-5} \) |
| Experiment \( ^{22} \) | | 2.61 \( ^{23} \) | \( 1.2 \times 10^{-6} \) | \( 1.1 \times 10^{-6} \) | \( 5.4 \times 10^{-6} \) |

\(^{22}\) Density measured with Archimedes method at 1973 K in 50CaO–50SiO\(_2\). (18)

\(^{23}\) Density measured with Archimedes method at 1973 K in 50CaO–50SiO\(_2\). (18)

\(^{24}\) Tracer-diffusion coefficient of O\(^{16}\) measured at 1873 K in 50CaO–50SiO\(_2\). (18)

\(^{25}\) Tracer-diffusion coefficient of Si\(^{18}\) measured at 1873 K in 48CaO–52SiO\(_2\). (20)

\(^{26}\) Tracer-diffusion coefficient of Ca\(^{40}\) measured at 1873 K in 49CaO–51SiO\(_2\). (21)
CaSiO$_3$ melts. The average coordination number of O around Si was four, which was determined from the first plateau of the Si–O curve in the running coordination number [Fig. 2(b)]. This corresponds to the formation of SiO$_4$ tetrahedra. By using Einstein’s equation, we calculated the self-diffusion coefficients of three kinds of ions from the mean square displacements [Fig. 2(c)], and the results are summarized in Table 2(b). The self-diffusion coefficients were compared with the experimentally obtained tracer diffusion coefficients, because a self-diffusion coefficient is generally close to, but not exactly the same as the tracer diffusion coefficient. The self-diffusion coefficients obtained by MD simulations were larger than the tracer-diffusion coefficient for the three kinds of ions, which was because of the potential parameters used in this study and the temperature difference between the MD simulation (2000 K) and the experiment (1873 K). Especially for the Matsui potential, the curve of the potential function used in this MD may be gentler than that of the actual potential, because the bulk modulus of 18 GPa at 1900 K calculated by MD is lower than that of the experimental data of 23 GPa at 1900 K, as shown in Ref. 15. The diffusion coefficients for Si, O, and Ca are in the order $D_{\text{Si}} < D_{\text{O}} < D_{\text{Ca}}$, in this MD simulation, which is consistent with the experimental results. By using $D_{\text{Si}}$, which is the smallest of the three coefficients, we calculated the time necessary for the system to reach the steady state, according to

$$t_{\text{steady}} = \frac{(x_{\text{hot}} - x_{\text{cold}})^2}{\pi D_{\text{Si}}},$$

where $x_{\text{hot}}$ and $x_{\text{cold}}$ are the central positions of the hot and cold slices, respectively, in the simulation box in Fig. 1(b).

The times to reach the steady state for the Matsui potential are 542, 469, and 553 ps for 9, 503, and 1019 MPa, respectively. The time to reach the steady state for the Seo potential is 2692 ps for 5 MPa. The sampling of mole-fraction distribution started at 1800 and 2745 ps for the Matsui potential and the Seo potential, respectively, after the system was exposed to the temperature gradient. The mole fraction in each slice in the simulation box was sampled in every step during 3600 ps for Matsui potential and during 5490 ps for Seo potential.

Generally, in a binary non-electrolyte system under one-dimensional temperature gradient, the mass flux is expressed for species 1 as the following equation:

$$J_{1,s} = -\rho \left[ D_{\text{M}} \left( \frac{\partial n_1}{\partial x} \right) + n_1(1-n_1)D_{\text{T}} \left( \frac{\partial T}{\partial x} \right) \right],$$

where $x$ is the position, $n_1$ is the mole fraction of the species 1, $\rho$ is the mass density, $T$ is the temperature and $D_{\text{M}}$ and $D_{\text{T}}$ are the mutual diffusion coefficient and the thermal diffusion coefficient, respectively. In the steady state, the mass flux $J_{1,s} = 0$, and we obtained the Soret coefficient as following equation:

$$\sigma_{\text{soret,1}} = \frac{D_{\text{T}}}{D_{\text{M}}} = -\frac{1}{n_1(1-n_1)} \frac{\partial n_1}{\partial T}. \quad (4)$$

The positive value of the Soret coefficient indicates that the species have a higher mole fraction in the cold region than in the hot region. The Soret coefficients meet the relation of $\sigma_{\text{soret,1}} = -\sigma_{\text{soret,2}}$. In this paper, we set CaO as the species 1 and SiO$_2$ as the species 2. In addition, we assume that the Soret coefficient does not depend on temperature, therefore, fitted the data plots with a line.

Because electric neutrality was preserved in each slice, namely the mole fraction of O was roughly double the mole fraction of Si plus the mole fraction of Ca, we discuss the distribution of the mole fractions of CaO and SiO$_2$. The mole-fraction distributions of CaO and SiO$_2$ with Matsui potential under 9 MPa are shown in Fig. 3. The sixteen sets of the MD program were run to obtain sufficient statistics. Soret coefficients were obtained from the gradient of the fitting line and Eq. (4). Although the dispersion of the mole fraction is large and data plots are frequently not on the fitting line, the gradients of fitting lines indicate that, averagely, CaO has a higher mole fraction in the cold region, and SiO$_2$ has a higher mole fraction in the hot region in the all results of the Fig. 3. The dispersions of the values may be attributed to the distribution of the short-range and medium-range structures in each slice. In the cold region, the melt structure in each slice may vary since a slice in the simulation box has only 125 ions averagely.

The results of all runs are shown in Table 3. The “Initial ion positions” in the Table 3 means the set of ion positions at the moment when the simulation started. The different number represents the different set of ion positions. For a set of initial ion positions, the mole fraction was sampled in every step during 3600 ps for Matsui potential and during 5490 ps for Seo potential.

![Fig. 3. Mole fraction distributions (Run No. 1–16 in Table 3). The numbers near the plots in the Run No. 1 corresponds to the slice numbers in Fig. 1(b). Solid lines are the fitted straight lines for determining the mole-fraction gradient.](image-url)
Table 3. Soret coefficients for various potential, initial ion positions, temperature control, and pressure. The values in brackets in the average row are the double standard errors.

| Run Number | Potential | Initial ion positions | Temperature Pressures /MPa | $\sigma_{\text{CaO}}/10^{-4}$K$^{-1}$ | $\sigma_{\text{SiO}_2}/10^{-4}$K$^{-1}$ |
|------------|-----------|----------------------|-----------------------------|-----------------------------------|-----------------------------------|
| 1          | Matsui    | normal               | 26.7                        | 2.49                              | -2.49                             |
| 2          | Matsui    | reverse              | 16.7                        | 1.50                              | -1.50                             |
| 3          | Matsui    | normal               | 19.5                        | 1.78                              | -1.78                             |
| 4          | Matsui    | reverse              | 23.1                        | 0.73                              | -0.73                             |
| 5          | Matsui    | normal               | 16.6                        | 4.15                              | -4.15                             |
| 6          | Matsui    | reverse              | 16.6                        | 2.67                              | -2.67                             |
| 7          | Matsui    | normal               | 24.6                        | 0.84                              | -0.84                             |
| 8          | Matsui    | reverse              | 22.2                        | 2.18                              | -2.18                             |
| 9          | Matsui    | normal               | 21.7                        | 1.99                              | -1.99                             |
| 10         | Matsui    | reverse              | 23.7                        | 1.16                              | -1.16                             |
| 11         | Matsui    | normal               | 23.1                        | 3.50                              | -3.50                             |
| 12         | Matsui    | reverse              | 20.9                        | 3.25                              | -3.25                             |
| 13         | Matsui    | normal               | 21.1                        | 2.83                              | -2.83                             |
| 14         | Matsui    | reverse              | 18.5                        | 1.16                              | -1.16                             |
| 15         | Matsui    | normal               | 21.9                        | 3.57                              | -3.57                             |
| 16         | Matsui    | reverse              | 24.2                        | 2.71                              | -2.71                             |
| Average    | (No.1–16) |                      | 21.3                        | 2.28                              | -2.28                             |

| Run Number | Potential | Initial ion positions | Temperature Pressures /MPa | $\sigma_{\text{CaO}}/10^{-4}$K$^{-1}$ | $\sigma_{\text{SiO}_2}/10^{-4}$K$^{-1}$ |
|------------|-----------|----------------------|-----------------------------|-----------------------------------|-----------------------------------|
| 17         | Matsui    | normal               | 518.9                       | 2.68                              | -2.68                             |
| 18         | Matsui    | reverse              | 524.5                       | 1.05                              | -1.05                             |
| 19         | Matsui    | normal               | 524.7                       | 1.09                              | -1.09                             |
| 20         | Matsui    | reverse              | 522.8                       | 1.56                              | -1.56                             |
| 21         | Matsui    | normal               | 526.6                       | 1.51                              | -1.51                             |
| 22         | Matsui    | reverse              | 528.7                       | 1.14                              | -1.14                             |
| 23         | Matsui    | normal               | 522.2                       | 1.86                              | -1.86                             |
| 24         | Matsui    | reverse              | 524.2                       | 1.61                              | -1.61                             |
| 25         | Matsui    | normal               | 526.0                       | 0.69                              | -0.69                             |
| 26         | Matsui    | reverse              | 530.8                       | 1.78                              | -1.78                             |
| 27         | Matsui    | normal               | 521.7                       | 1.88                              | -1.88                             |
| 28         | Matsui    | reverse              | 525.7                       | 0.88                              | -0.88                             |
| 29         | Matsui    | normal               | 525.0                       | 1.84                              | -1.84                             |
| 30         | Matsui    | reverse              | 522.1                       | 1.78                              | -1.78                             |
| 31         | Matsui    | normal               | 519.7                       | 2.67                              | -2.67                             |
| 32         | Matsui    | reverse              | 519.6                       | 3.13                              | -3.13                             |
| Average    | (No.17–32)|                      | 524.0                       | 1.70                              | -1.70                             |

| Run Number | Potential | Initial ion positions | Temperature Pressures /MPa | $\sigma_{\text{CaO}}/10^{-4}$K$^{-1}$ | $\sigma_{\text{SiO}_2}/10^{-4}$K$^{-1}$ |
|------------|-----------|----------------------|-----------------------------|-----------------------------------|-----------------------------------|
| 33         | Matsui    | normal               | 1023.4                      | 1.98                              | -1.98                             |
| 34         | Matsui    | reverse              | 1021.7                      | 1.89                              | -1.89                             |
| 35         | Matsui    | normal               | 1020.2                      | 3.04                              | -3.04                             |
| 36         | Matsui    | reverse              | 1027.1                      | 1.22                              | -1.22                             |
| 37         | Matsui    | normal               | 1022.8                      | 0.80                              | -0.80                             |
| 38         | Matsui    | reverse              | 1018.1                      | 0.44                              | -0.44                             |
| 39         | Matsui    | normal               | 1019.0                      | 0.76                              | -0.76                             |
| 40         | Matsui    | reverse              | 1023.1                      | 2.36                              | -2.36                             |
| 41         | Matsui    | normal               | 1020.6                      | 0.54                              | -0.54                             |
| 42         | Matsui    | reverse              | 1027.2                      | 2.59                              | -2.59                             |
| 43         | Matsui    | normal               | 1024.0                      | 1.93                              | -1.93                             |
| 44         | Matsui    | reverse              | 1019.7                      | 3.22                              | -3.22                             |
| 45         | Matsui    | normal               | 1024.5                      | 1.17                              | -1.17                             |
| 46         | Matsui    | reverse              | 1022.2                      | 0.13                              | -0.13                             |
| 47         | Matsui    | normal               | 1021.9                      | 1.22                              | -1.22                             |
| 48         | Matsui    | reverse              | 1025.1                      | 0.73                              | -0.73                             |
| Average    | (No.33–48)|                      | 1022.5                      | 1.50                              | -1.50                             |

| Run Number | Potential | Initial ion positions | Temperature Pressures /MPa | $\sigma_{\text{CaO}}/10^{-4}$K$^{-1}$ | $\sigma_{\text{SiO}_2}/10^{-4}$K$^{-1}$ |
|------------|-----------|----------------------|-----------------------------|-----------------------------------|-----------------------------------|
| 49         | Seo       | normal               | -28.4                       | 4.34                              | -4.34                             |
| 50         | Seo       | reverse              | -27.6                       | 4.63                              | -4.63                             |
| 51         | Seo       | normal               | -15.1                       | 1.73                              | -1.73                             |
| 52         | Seo       | reverse              | -12.8                       | 0.89                              | -0.89                             |
| 53         | Seo       | normal               | -20.4                       | 3.18                              | -3.18                             |
| 54         | Seo       | reverse              | -14.7                       | 0.59                              | -0.59                             |

Continued.

Fig. 4. Summary of the Soret coefficients obtained by the MD. The error bars indicate the double standard errors.

positions, we simulated with two types of temperature distributions. “Normal” temperature distribution means the conditions where the hot slice is the fifth slice and the cold slice is the first slice [see Fig. 1(b)]. “Reverse” temperature distribution means that the hot slice is the first slice and the cold slice is the fifth slice. In all runs, temperature in each slice is almost linearly changed from hot slice (~2200 K) to cold slice (~1800 K), as shown in the run number 1 in the Fig. 3. The signs of the Soret coefficients of CaO are positive, and those of SiO$_2$ are negative, except for the Run No. 61. The positive signs of CaO indicate that the CaO is concentrated in the cold region. The negative signs of the SiO$_2$ indicate that the SiO$_2$ is concentrated in the hot region.

Figure 4 shows the summary of the results in this research. As shown with the error bar, because of the dispersity of the obtained data, it is difficult to determine whether the Soret coefficient is pressure-dependent, although the average value increases gradually with the pressure. The result that CaO has a higher mole fraction in the cold region and SiO$_2$ has a higher mole fraction in the hot region is qualitatively consistent with the experimental results of ion migration under the temperature distribution during laser irradiation.\(^1\) The difference of the Soret coefficients is small between the Matsui potential and the Seo potential, which is contrast with the large difference in the diffusion coefficients.

In future work, we intend to elucidate the contribution of the short- and medium-range structure of the CaSiO$_3$ melt to the Soret effect. This study indicates that we will be able to discuss the Soret effect in silicate melts in terms of the weight of the ions,
shape of interionic potential function, and the free energy of the system.

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