The structure of liquid aluminosilicate under Al$_2$O$_3$ content: simplesxes and shell core analyses

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Abstract. The structural properties of $x$Al$_2$O$_3$($1-x$)SiO$_2$ liquid systems have beeninvestigated by molecular dynamics simulation in a wide range of compositions, $x = 0.05 - 0.7$ at 3000 K. The structure of liquid aluminosilicate system is clarified by analyzing the simplex and shell-core. The simulated results showed that the liquid consists of a large quantity of void-simplex, O-simplex, T-simplex, and SC-particle. Our simulation reveals that the densification of the liquid is due to the fact that the number of large simplesxes and the radii of simplesxes and SC-particles decrease. Besides, results also indicated that the distribution of cations in the liquid is not uniform.

1. Introduction
Silica and its mixtures with various oxides such as Al$_2$O$_3$, MgO, PbO, CaO, Na$_2$O, etc. play an important role in many geochemical and geophysical phenomena, and they are the main components of the Earth’s mantle [1-5]. Therefore, the structure of the alumino-silicate and other oxides is studied by many experimental techniques [6-19]. By using different experimental techniques such as NMR spectroscopy, Raman spectroscopy and X-ray diffraction, the existence of 4-, 5- and 6-fold Al-coordination units in aluminosilicate systems have been revealed, in which the 4-fold Al-coordination unit is main at low Al$_2$O$_3$ content. There is a gradual growth in the fraction of these high-coordinated species as the Al$_2$O$_3$ content increases and these results are consistent with the chemical shift of AlKα and the density data. Particularly, the structural clusters such as TO$_4$ (T=Al or Si) tetrahedral tri-clusters are observed in the aluminosilicate system which may act as the mullite nuclei during
crystallization. Moreover, as the Al2O3 content rises, the intermediate-range order of the glass structure rapidly breaks down and the SiO4 units are considerably distorted.

So far, the structural characteristics of the aluminosilicate system have not been fairly studied yet. Therefore, in order to understand physical properties of the aluminosilicate system, many studies are performed by molecular dynamics simulation for this system [20-23]. The results showed that the aluminosilicate system consists mainly of [4]O and [6]O having an important role in the diffusion of atomic species and the significant fraction of [4]O with increasing Al2O3 content from 33.33 to 66.67 mol %. These results also confirmed the existence of mixture four-five, and six-fold coordinated Al atoms and the increase of these high coordinated Al species as the Al2O3 content increases, which have been found by experimental studies [24-26]. By using molecular dynamics simulations and structural visualization techniques, the distribution of TO4 units (T is Si, Al) with T is the network former or network modifier element has been clarified [27-32]. These structural units connect each other through common oxygen atoms to form the continuous random network in the three-dimensional space.

Although the liquid aluminosilicate system has been studied for a long time, the structure of aluminosilicate system is not fully understood. In this work the structure of the liquid xAl2O3 (1-x)SiO2 at ambient pressure and 3000 K is studied via analysis the distribution of simplex and shell-core as Al2O3 content varies from 0.05 to 0.7.

2. Calculation method

Molecular dynamics (MD) simulation is done for xAl2O3 (1-x)SiO2 system in a wide range of compositions, x = 0.05–0.7, which consists of 5000 atoms of each sample at a temperature of 3000K and ambient pressure. The model is employed the Born-Mayer potential as following:

\[ \phi_{ij} = \frac{q_i q_j}{r_{ij}} + A_{ij} \exp(-B_{ij} r_{ij}) \]

These potential parameters can find in Ref. 33. In this paper, the motion equation is approximately solved by a numerical method using the Verlet algorithm with the MD step of 1.0 fs. The initial configuration of different samples, as noted in Table 1, is generated by randomly placing atoms in a cubic simulation box. In order to eliminate the effect of remembering the initial configuration, the samples are then heated up to 6000 K and are relaxed to reach the equilibrium after 50 ps. Next, the samples are cooled down to the temperature of 3000 K with a rate of about 10^12 K/s, followed by a relaxation period for over 10^7 MD steps in the NPT ensemble until reaching the equilibrium.

The structural heterogeneity of liquid silicate not only is described by structural units but also relates to the fluctuation of free volume as well as the composition chemistry. The information about the porosity of materials is provided through the simplex analysis. So, the simplex and shell-core methods are suitable to describe the structural heterogeneity of liquid silicate because the cutoff radius is removed in this case. The structural properties of liquid aluminosilicate are analyzed by the simplex and shell core (SC) methods [34, 35]. Here, the simplex is a sphere which passes through the centers of four arbitrary atoms. The void-simplex with the radius of RVS is the simplex with no atoms in-side and is characterized by number abc where a, b and c are the number of oxygen, silicon and aluminum atoms, respectively, that the simplex passes through (a + b + c = 4). Void-simplex is computed by the method used in Ref. [34]. The simplexes and SC-particle are schematically illustrated in Fig. 1 and Fig.2. Accordingly, each atom is considered as a sphere in which the radius of atoms Al, Si, and O are taken as 1.23, 1.43 and 0.73 Å respectively. To calculate void-simplexes, we perform as follow: firstly, we determine all sets consisting of four atoms from the system and have to make sure that the distance between any two atoms is not large than 9.5 Å. Thereby, we have obtained a set of void-simplex with different radii. In the next step, we remove void-simplexes locating almost inside or intersecting volume with other void-simplexes over 50 %.

The simplexes that contain only oxygen or cation atoms inside are called O-simplex or T-simplex. The O-simplex and T-simplex with the radius of ROS and RCS, respectively, are also characterized by the number a,b,c; (i=1 for O-simplex and 2 for T-simplex) where a, b, and c; are the number of oxygen,
silicon and aluminum atoms, respectively, that these simplexes pass through. The SC-particle consists of the core and the shell, in which the core and the shell contain only cation and oxygen, respectively. The $R_c$, $D_S$ and $[s, c]$ are the radius of the core, the length of the shell and the number of oxygen atoms in the shell and the number of cations in the core, respectively. To calculate O-simplex, T-simplex as well as SC-particle, we use methods presented in [35, 36]. O-simplex and T-simplex are computed as follows. Firstly, we determine all O-simplexes and T-simplexes in the system. The obtained O-simplexes and T-simplexes have different the radius and number of atoms inside. In the next step, a part of O-simplexes and T-simplexes is removed. Namely, the first simplex is removed if the list of atoms is a subset of the second simplex. If the list of atoms of two simplexes is the same then the simplex having the larger radius is removed. For SC-particle we calculate as follows: firstly, we determine all simplexes passing four oxygen atoms and containing only cation atoms inside. Then, we choose the sphere which has the largest radius, the center is center of simplex and the area between surfaces of those spheres contains only oxygen atoms. Finally, we obtain SC-particles containing only oxygen atoms in the shell and cation atoms in the core.

![Simplexes](image1)

**Figure 1.** The description of simplexes: (a) the void-simplex; (b) the oxygen-simplex; (c) the cation-simplex (T-simplex). The orange and blue sphere are oxygen and cation atoms, respectively. (The color in these figures, the reader is found to the online of this paper.)

![Simplexes](image2)

**Figure 2.** The description of simplexes: SC-particle. The blue and green sphere are cation and oxygen atoms, respectively. (The color in these figures, the reader is found to the online of this paper.)

| Sample | Mole fraction % | Al$_2$O$_3$ SiO$_2$ | Si | Al | O | Total |
|--------|----------------|---------------------|----|----|---|-------|
| M1     | 5              | 95                  | 1615| 170| 3485| 5270  |
| M2     | 20             | 80                  | 1200| 600| 3300| 5100  |
| M3     | 25             | 75                  | 1125| 750| 3375| 5250  |
| M4     | 33             | 67                  | 930 | 930| 3255| 5115  |
| M5     | 40             | 60                  | 840 | 1120| 3360| 5320  |
| M6     | 50             | 50                  | 650 | 1300| 3250| 5200  |
| M7     | 60             | 40                  | 480 | 1440| 3120| 5040  |
| M8     | 70             | 30                  | 360 | 1680| 3240| 5280  |

**Table 1.** The fraction of Al$_2$O$_3$ and SiO$_2$ content and atomic numbers of each sample at 3000 K.
3. Results and discussions

Figure 3 displays the distribution of the radius of void-simplex in the molten aluminosilicate system. The position of main peaks of the radius distribution shifts from 2.21 to 1.99 Å, their height rises slightly as the Al₂O₃ composition increases. This means the radius of the void-simplexes reduces as the Al₂O₃ content grows. This can lead to the change of the diffusion of atoms in the liquid. At x=0.05, we can see that the model has a number of very large V-simplexes having a radius larger than 3 Å about 3.62%. These V-simplexes have a volume fourteen and nine times bigger than the volume of Al and Si atoms, respectively. So, these V-simplexes have large enough volume to can freely receive fourteen and nine additional Al and Si atoms and participate in the diffusion process. This means that these V-simplexes cannot is considered vacancies as was found in crystalline materials. These V-simplexes are attributed as a microscopic cavity and the fraction of theirs decreases as Al₂O₃ content increases. So, the existence of very large void-simplexes (with the radius bigger than the atomic radius) indicated that the structure of material is loose. This is discussed in [34] for SiO₂ and [37] for Al₂O₃. However, our results show that the model exists V-simplexes with the volume much larger in comparison with results in [34] and [37].

Table 2 shows the characteristics of void-simplexes for different contents of Al₂O₃. According to this table, the void-simplexes are dominant 031, 040 and 130 at the low-Al₂O₃ content, whereas with high-Al₂O₃ content most of those are 031, 040, 130 and 220 types.

![Figure 3. The radius distribution of void-simplexes.](image)

As the Al₂O₃ content increases, the N₉ of 031 and 040 types of the void-simplexes, which pass through four cation atoms, reduces significantly (about 80.6% of 031 type and 51.9% of 040 type), whereas the N₉ of 130 and 220 types grows considerably as Al₂O₃ content rises (about 86.6% of 130 type and 98.7% of 220 type). The space region inside TOₓ unit is occupied by the 310 void-simplexes, while the 211, 220, 130, 031 and 040 void-simplexes occupies the space region between two and more two adjacent TOₓ units. Thus, the increase in the number of the high-coordinated TOₓ and the decline of the T-O-T angle are attributed by the reduction in a radius of these void-simplexes. Therefore, we can conclude that the densification of the liquid is due to the radius and the number of large void-simplex decrease.
Table 2. The properties of void-simplex: N\textsubscript{VS} is the mean number of void-simplex per atom.

| abc | \(x5\) | \(x20\) | \(x25\) | \(x33\) | \(x40\) | \(x50\) | \(x60\) | \(x70\) |
|-----|------|------|------|------|------|------|------|------|
| N\textsubscript{VS} | N\textsubscript{VS} | N\textsubscript{VS} | N\textsubscript{VS} | N\textsubscript{VS} | N\textsubscript{VS} | N\textsubscript{VS} | N\textsubscript{VS} | N\textsubscript{VS} |
| 022 | 0.1450 | 0.0612 | 0.0495 | 0.0418 | 0.0282 | 0.0237 | 0.0077 | 0.0089 |
| 031 | 3.4471 | 2.4839 | 2.1610 | 1.8344 | 1.5534 | 1.2438 | 0.9688 | 0.6684 |
| 040 | 2.5078 | 2.1339 | 2.0314 | 1.8377 | 1.7284 | 1.5506 | 1.3772 | 1.2038 |
| 012 | - | 0.0004 | 0.0004 | - | 0.0004 | - | - | - |
| 021 | 0.4989 | 1.6737 | 2.0328 | 2.4364 | 2.7376 | 3.0806 | 3.4050 | 3.7273 |
| 011 | 0.0165 | 0.2259 | 0.3293 | 0.4976 | 0.6211 | 0.8179 | 1.0683 | 1.2814 |
| 000 | 0.0020 | 0.0011 | 0.0031 | 0.0041 | 0.0065 | 0.0111 | 0.0110 | 0.0000 |

Table 3 presents the number of O-simplex at different \(\text{Al}_2\text{O}_3\) contents. From this table, we can see that the O-simplex comprises from 2 to 14 oxygen atoms. However, O-simplex type consisting of from 3 to 7 oxygen atoms is dominant, in which the Nos has the value about 91.49 and 93.79% at low- and high-\(\text{Al}_2\text{O}_3\) content, respectively. This means the spatial distribution of oxygen atoms becomes more homogeneous at high- \(\text{Al}_2\text{O}_3\) content. On the other hand, the liquid exists a significant number of large O-simplexes which comprises more than 6 oxygen atoms and a radius bigger than 3 Å (see in Figure 4), in which the fraction of these O-simplexes decreases as \(\text{Al}_2\text{O}_3\) content increases. These large O-simplexes are an interstitial site for oxygen atoms. For O-simplexes containing more than 6 oxygen atoms, the fraction of these O-simplexes very strongly reduces about 25.64% (see Table 3) and this is about 5% for O-simplexes with the radius bigger than 3 Å. This means the densification is due to the disappearance of the large O-simplexes. As mentioned above, the model exists O-simplexes having the number of oxygen atoms bigger than 6 and are considered as the interstitial site for oxygen atoms. If these simplexes are located nearby then, they can form negative-charged clusters of different sizes. The diffusion of atoms in these regions slower than the ones in the remaining regions. Therefore, these regions play a role as a trap to keep atoms for a long time [38].

![Figure 4](image_url)

**Figure 4.** The dependence on \(\text{Al}_2\text{O}_3\) content of the radius of O-simplex.

Figure 4 indicates the radius distribution of O-simplexes at the different \(\text{Al}_2\text{O}_3\) content. We can see that the graph shifts to a smaller radius upon increasing \(\text{Al}_2\text{O}_3\) content, in which the position of the main peaks of the radius distribution is almost unchanged as \(\text{Al}_2\text{O}_3\) content increases from 5 to 70%.
Figure 4 shows that the radius of the O-simplex decreases with increasing of Al\textsubscript{2}O\textsubscript{3} content. So, the densification of liquids is caused by decreasing of the radius of O-simplex.

Table 3. The properties of O-simplexes: Nos is the number of O-simplex types.

| a,b,c | x5 | x20 | x25 | x33 | x40 | x50 | x60 | x70 |
|-------|----|-----|-----|-----|-----|-----|-----|-----|
| Nos   | Nos| Nos | Nos | Nos | Nos | Nos | Nos | Nos |
| 200   | 8  | 31  | 44  | 74  | 118 | 193 | 265 | 413 |
| 300   | 652| 895 | 1052| 1244| 1510| 1646| 1874| 2380|
| 400   | 2439| 2554| 2578| 2626| 2731| 2693| 2800| 2766|
| 500   | 2965| 2748| 2780| 2547| 2582| 2292| 2020| 1883|
| 600   | 2121| 1709| 1685| 1551| 1507| 193 | 982 | 874 |
| 700   | 1177| 926 | 813 | 629 | 488 | 1217| 982 | 874 |
| 800   | 543 | 362 | 302 | 270 | 195 | 115 | 99  | 99  |
| 900   | 229 | 105 | 105 | 77  | 43  | 37  | 25  | 25  |
| 1000  | 60  | 43  | 30  | 42  | 21  | 14  | 7   | 6   |
| 1100  | 18  | 15  | 7   | 8   | 6   | 3   | 1   | 1   |
| 1200  | 14  | 5   | 3   | 1   | 1   | 1   | 1   | 1   |
| 1300  | 3   | -   | -   | -   | -   | -   | -   | -   |
| 1400  | 2   | -   | -   | -   | -   | -   | -   | -   |

Table 4. The properties of T-simplexes: N\textsubscript{CS} is the number of T-simplex.

| a,b,c | x5 | x20 | x25 | x33 | x40 | x50 | x60 | x70 |
|-------|----|-----|-----|-----|-----|-----|-----|-----|
| Ncs   | Ncs| Ncs | Ncs | Ncs | Ncs | Ncs | Ncs | Ncs |
| 001   | 1237| 845 | 736 | 587 | 478 | 343 | 258 | 176 |
| 002   | 126 | 43  | 46  | 38  | 24  | 20  | 9   | 8   |
| 010   | 38  | 99  | 97  | 89  | 108 | 83  | 91  | 86  |
| 011   | 171 | 314 | 346 | 319 | 376 | 335 | 252 | 219 |
| 012   | 1   | 1   | -   | -   | 1   | -   | -   | -   |
| 020   | 25  | 285 | 428 | 655 | 801 | 1076| 1313| 1672|
| 021   | -   | 2   | 5   | -   | -   | 5   | 9   | 3   |
| 030   | -   | 6   | 4   | 7   | 12  | 17  | 34  | 29  |

Table 4 displays the characteristics of T-simplex in all range contents of Al\textsubscript{2}O\textsubscript{3}. As can be seen in this Table, T-simplexes contain from 1 to 3 cation atoms. Here the N\textsubscript{CS} of 001 T-simplex type containing one Al atom has a value equal to 1237 at low Al\textsubscript{2}O\textsubscript{3} content and this value very strongly goes down to 176 at high Al\textsubscript{2}O\textsubscript{3} content. This tendency is similar for 002 T-simplex. Meanwhile, the value of N\textsubscript{CS} of 020 T-simplex, which contains only two Si atoms, dramatically increases from 25 to 1672 upon increasing Al\textsubscript{2}O\textsubscript{3} content. On the other hand, the liquid has a number of large T-simplexes with the radius bigger than 3.0 Å (see Figure 5). The fraction of these T-simplexes decreases as Al\textsubscript{2}O\textsubscript{3} content increases (almost equal to zero at 70% Al\textsubscript{2}O\textsubscript{3} content). We can see that the model exists a significant number of T-simplexes with a number of cation atoms bigger than 3. As mentioned above, they are the interstitial sites for cation atoms. As was discussed in Ref. [38], these simplexes tend to connect with each other to form positive-charged clusters in the model. The existence of these regions plays a role as diffusion channels for Al atom in comparison to Si and O atoms. This means that the distribution of cation atoms in the model is heterogeneous.
The radius distribution of T-simplexes shows in Figure 5, in which the position of main peaks shifts to the left with increasing Al\textsubscript{2}O\textsubscript{3} content and the height of main peaks rises from 17.2 to 19.8% as Al\textsubscript{2}O\textsubscript{3} content increases from 5 to 70%. This means the radius of simplex reduces upon Al\textsubscript{2}O\textsubscript{3} content increases. Thus, the densification of the liquid is due to the radius and a number of large T-simplexes decrease.

Table 5 presents the properties of SC-particle in low-and high-Al\textsubscript{2}O\textsubscript{3} content. We can see that N\textsubscript{SC} of [4,1], [5,1] and [6,1] SC-particles types are dominant about 80% at low-Al\textsubscript{2}O\textsubscript{3} content. However, in high-Al\textsubscript{2}O\textsubscript{3} content the N\textsubscript{SC} of these types significantly decreases about 38.6%, while the N\textsubscript{SC} of [4,2], [5,2] and [6,2] types substantially increase about 44.1%. In addition, at low-Al\textsubscript{2}O\textsubscript{3} content the N\textsubscript{SC} of [s,1] type monotonously goes down as the number of oxygen atoms in the shell increases and the N\textsubscript{SC} of [s,3] type is almost equal to zero. At high-Al\textsubscript{2}O\textsubscript{3} content the N\textsubscript{SC} of [s,1] and [s,2] types also gradually reduce with the increasing number of oxygen atoms in the shell. Simultaneously, in the liquid exists a number of [s,3] type. Thus, at high-Al\textsubscript{2}O\textsubscript{3} content a significant number of the large SC-particles comprising 2 and 3 cations in the core (about 60.2%) were found in the liquid. This means the distribution of cation atoms in the liquid is not uniform as Al\textsubscript{2}O\textsubscript{3} content increases. This can lead to the formation of the positive charge cluster.

The data in Table 5 also shows that the core radius of SC-particle types monotonously increases with the number of oxygen atoms in the shell at both low- and high-Al\textsubscript{2}O\textsubscript{3} content. Meanwhile, the radius of the core of the [s,1] type slightly decreases with increasing Al\textsubscript{2}O\textsubscript{3} content. The length of the shell of [4,1] type slightly reduces as Al\textsubscript{2}O\textsubscript{3} content increases, in which the length of shell of [4,1] type strongly decreases about 33.8%. However, the length of the shell of [s,2] type significantly reduces from 18.3 to 36.1% as Al\textsubscript{2}O\textsubscript{3} content increases.

The average atomic density of SC-particle can be estimated by $\rho_{\text{SC}} = (s+c)/(4\pi(Rc+Ds)^{3}/3)$ [33]. We can see that at low-Al\textsubscript{2}O\textsubscript{3} content the $\rho_{\text{SC}}$ of [s,1] and [s,2] types have a value bigger than 0.06 except [9,1] and [8,3] types. At high Al\textsubscript{2}O\textsubscript{3} content this value of [s,1] and [s,2] types significantly increase, while the $\rho_{\text{SC}}$ of [4,1] and [4,2] types have the strongest rise about 26.9% and 39%, respectively. Therefore, as Al\textsubscript{2}O\textsubscript{3} content goes up the average atomic density of SC-particle increases. Thus, the increase of the average atomic density is related to the densification in the liquid.
Table 5. The characteristics of SC-particles at 0.05 and 0.70 Al$_2$O$_3$ content, where the number of SC-particles are $N_{SC}$, the mean radius of core and length of the shell are $R_c$, $D_s$ respectively; the mean atomic density of SC-particles is $\rho_{SC}$.

| [s,c] | $N_{SC}$ | $<R_c>\,(\text{Å})$ | $<D_s>\,(\text{Å})$ | $<\rho_{SC}>\,(\text{Å}^{-3})$ | $N_{SC}$ | $<R_c>\,(\text{Å})$ | $<D_s>\,(\text{Å})$ | $<\rho_{SC}>\,(\text{Å}^{-3})$ |
|-------|---------|----------------|----------------|----------------|---------|----------------|----------------|----------------|
| [4,1] | 692     | 2.01           | 0.65           | 0.063          | 435     | 2.03           | 0.43           | 0.080          |
| [5,1] | 473     | 2.26           | 0.47           | 0.070          | 176     | 2.21           | 0.45           | 0.076          |
| [6,1] | 121     | 2.49           | 0.45           | 0.066          | 46      | 2.47           | 0.42           | 0.069          |
| [7,1] | 47      | 2.68           | 0.43           | 0.063          | 16      | 2.61           | 0.43           | 0.068          |
| [8,1] | 15      | 2.84           | 0.46           | 0.100          | 3       | 2.79           | 0.43           | 0.064          |
| [9,1] | 3       | 2.95           | 0.43           | 0.019          | -       | -              | -              | -              |
| [4,2] | 15      | 2.12           | 0.72           | 0.063          | 362     | 2.07           | 0.46           | 0.088          |
| [5,2] | 51      | 2.19           | 0.74           | 0.066          | 352     | 2.19           | 0.55           | 0.081          |
| [6,2] | 90      | 2.25           | 0.80           | 0.067          | 200     | 2.32           | 0.62           | 0.075          |
| [7,2] | 68      | 2.37           | 0.84           | 0.065          | 69      | 2.48           | 0.58           | 0.075          |
| [8,2] | 22      | 2.64           | 0.76           | 0.061          | 17      | 2.57           | 0.61           | 0.074          |
| [9,2] | 4       | 2.64           | 0.87           | 0.061          | 4       | 2.67           | 0.71           | 0.068          |
| [10,2]| 3       | 2.68           | 0.79           | 0.069          | -       | -              | -              | -              |
| [5,3] | -       | -              | -              | -              | 3       | 2.03           | 0.65           | 0.112          |
| [6,3] | -       | -              | -              | -              | 7       | 2.48           | 0.74           | 0.064          |
| [7,3] | -       | -              | -              | -              | 5       | 2.67           | 0.51           | 0.074          |
| [8,3] | 1       | 2.74           | 0.95           | 0.057          | 3       | 2.38           | 1.00           | 0.062          |
| [9,3] | -       | -              | -              | -              | -       | -              | -              | -              |
| [10,3]| -       | -              | -              | -              | 1       | 2.62           | 0.51           | 0.078          |

Figure 6. The dependence of number SC-particle on number of atoms in the core and shell.

The distribution number of SC-particle depending on the number of cation atom in the core and in the shell is indicated in Fig. 6. For SC-particle consisting of one atom in the core, the number of SC-particles has a maximum of 1351 at x=0.5 Al$_2$O$_3$ content. This value reduces significantly with increasing Al$_2$O$_3$ content. Meanwhile, the SC-particle type comprising 4 oxygen atoms in the shell, the
maximum of number of SC-particles is 707 at x=0.5 Al₂O₃ content and this increases as Al₂O₃ content increases.

\[ N_{SCC} \]

Figure 7. The number of SC-clusters as a function of Al₂O₃ content, \( N_{SC} \) is the number of SC-particles.

Figure 7 shows the dependence of the number of SC-cluster on the Al₂O₃ content. The number of SC-cluster with \( N_{SC} = 1 \) has the largest value about 88.4% of the total \( N_{SCC} \) of SC-cluster types at low-Al₂O₃ content and this \( N_{SCC} \) significantly decreases (about 50%) upon increasing Al₂O₃ content. Meanwhile, the \( N_{SCC} \) of SC-cluster containing 2 and 3 SC-particles slightly increases with Al₂O₃ content, the SC-cluster type comprising 4 SC-particles is not significant. In addition, the \( N_{SCC} \) sharply reduces as \( N_{SC} \) increases. Namely, the SC-cluster type containing 1 SC-particle inside has the value of \( N_{SCC} \) equal to 1351, for SC-cluster types which contain 2 and 3 SC-particles, the values are 131 and 34, respectively, at low-Al₂O₃ content. Meanwhile, at high-Al₂O₃ content, the \( N_{SCC} \) of these SC-particle types is 676, 171 and 56, respectively. On the other hand, in the liquid a number of large SC-particle types which have more than 6 cations inside with the largest SC-cluster has 39 cations (see Figure 8). This means, the distribution of atoms is not uniform with Al₂O₃ content and this cause leads to the formation of cation-rich regions in the liquid.

Figure 8. The number of SC-clusters distribution as a function of number of cations.
4. Conclusion
In this work, the structure of the liquid alumino-silicate system has been investigated through void-simplexes. The results show that the structure of liquids consists of a large number of 031, 040, 130 and 220 void-simplexes. The loose property of the structure is due to the existence of the large void-simplexes. In addition, a large number of O-simplexes and T-simplexes are also found in the liquid.

The densification in the liquid alumino-silicate is caused by factors namely the decrease in the number of 022, 031 and 040 types, the decrease in the radius of the large void and 220 void simplexes. In this work, the structure of the liquid is also realized by shrinking of the radius of the core of SC-particle.

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References
[1] Kim J R, Lee Y S, Min D J, Jung S M and Yi S H 2004 ISIJ Int 44(8) 1291-1297.
[2] Wilding M C and Benmore C J 2006 Rev. Mineral. Geochem 63(1) 275-311.
[3] Murdoch J B, Stebbins J F and Carmichael I S 1985 Am. Mineral 70(3-4) 332-343.
[4] Wang Y, Sakamaki T, Skinner L B, Jing Z, Yu T, Kono Y, et al 2014 Nat. Commun 5(1) 1-10.
[5] Zeidler A, Salmon P S and Skinner L B 2014 Proc. Natl. Acad. Sci., India 111(28) 10045-10048.
[6] Risbud S H, Kirkpatrick R J, Tagliavare A P and Montez B 1987 J. Am. Ceram. Soc 70(1) C-10.
[7] Dedson P and Dupree R 1991 J. Phys. Chem 95 4483-4489.
[8] Meinhold R H, Slade R C T and Davies T W 1993 Appl. Magn. Reson 4(1-2) 141-155.
[9] Schmücker M and Schneider H 1996 Ber. Bunsenges. Phys. Chem 100(9) 1550-1553.
[10] Schmücker M, MacKenzie K J D, Schneider H and Meinhold R 1997 J. Non-Cryst. Solids 217(1) 99-105.
[11] Schmücker M, Schneider H, MacKenzie K J D and Okuno M 1999 J. Eur. Ceram. Soc 19(1) 99103.
[12] Xue X and Kanzaki 1999 M, J. Phys. Chem. B 103(49) 10816-10830.
[13] Poe B T, McMillan P F, Cote B, Massiot D and Coutures J P 1992 J. Phys. Chem 96(21) 8220-8224.
[14] Poe B T, McMillan P F, Angell C A and Sato R K 1992 Chem. Geol 96(3-4) 333-349.
[15] Allwardt J R, Stubbins J F, Schmidt B C, Frost D J, Withers A C and Hirschmann M M 2005 Am. Mineral 90(7) 1218-1222.
[16] Lee S K 2004 J. Phys. Chem. B 108(19) 5889-5900.
[17] Morikawa H, MIWA S I, Miyake M, Marumo F and Sata T 1982 J. Am. Ceram. Soc 65(2) 78-81.
[18] McMillan P and Piriou B 1982 J. Non-Cryst. Solids 53(3) 279-298.
[19] Okuno M, Zotov N, Schmücker M and Schneider H 2005 J. Non-Cryst. Solids 351(12-13) 1032-1038.
[20] Van Hoang V 2007 Phys. Lett. A 368(6) 499-503.
[21] Van Hoang V 2007 Phys. Rev. B 75(17) 174202.
[22] Takei T, Kameshima Y, Yasumori A and Okada K 2000 J. Mater. Res 15(1) 186-193.
[23] Winkler A, Horbach J, Kob W and Binder K 2004 J. Chem. Phys 120(1) 384-393.
[24] Wilding M C, Benmore C J and Weber J K R 2010 J. Phys. Chem. B 114(17) 5742-5746.
[25] Aksay I A, Pask J A and Davis R F 1979 J. Am. Ceram. Soc 62(7-8) 332-336.
[26] Schmücker M and Schneider H 2002 J. Non-Cryst. Solids 311(2) 211-215.
[27] Pfleiderer P, Horbach J and Binder K 2006 Chem. Geol 229(1-3) 186-197.
[28] Binder K, Horbach J, Winkler A and Kob W 2005 Ceram. Int 31(5) 713-717.
[29] Hong N V, Yen N V, Lan M T and Hung P K 2014 Can. J. Phys 92(12) 1573-1580.
[30] Zheng K, Zhang Z, Yang F and Sridhar S 2012 *ISIJ Int* **52**(3) 342-349.
[31] Lan M T, Iitaka T and Hong N V 2018 *Int. J. Mod. Phys. B* **32**(24) 1850271.
[32] Yen N V, Lan M T, Vinh L T and Hong N V 2017 *Mod. Phys. Lett. B* **31**(05) 1750036.
[33] Van Hoang V 2007 *Phys. B* **400**(1-2) 278-286.
[34] Vinh L T, Hung P K, Hong N V and Tu T T 2009 *J. Non-Cryst. Solids* **355**(22-23) 1215-1220.
[35] Hung P K, Vinh L T, Van T B, Hong N V and Yen N V 2017 *J. Non-Cryst. Solids* **462** 1-9.
[36] Nguyen T H T, Nguyen Y V, Pham H K and Nguyen H V 2017 *Int. J. Mod. Phys. B* **31**(15) 1750127.
[37] Hoang V V and Oh S K 2005 *Physica B: Condensed Matter* **364**(1-4) 225-232.
[38] Hung P K, Noritake F, Yen N V and San L T 2016 *J. Non-Cryst. Solids* **452** 14-22.