Molecular Dynamics Study of Structural Properties of Refining Slag with Various CaO/Al₂O₃ Ratios

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Abstract: SiO₂-Al₂O₃-CaO-MgO is the main type of refining slag in a ladle furnace. Here the effects of the CaO/Al₂O₃ mass ratio on the structural properties of the refining slag system are studied by molecular dynamics simulations. The pair distribution function, coordination number, microstructure unit and diffusion capacity were analyzed. An increase in the CaO/Al₂O₃ ratio did not change the bond lengths of these units. However, an increase of the CaO/Al₂O₃ ratio caused more charge compensators to be introduced into the refining slag system, which led to conversion of free oxygen and non-bridging oxygen to tricluster oxygen and bridging oxygen in the system. Moreover, this augmented the content of relatively stable 4- and 5-coordinate Al. As the micro-structure of the system became more complex, the overall diffusion capacity of the refining slag became poorer. In the micro-structure, the diffusion capabilities of different atoms decreased in the following order: Mg > Ca > O > Al > Si.

Keywords: molecular dynamics; microstructure; refining slag; CaO/Al₂O₃ ratio; transport properties

1. Introduction

Developments in industry and civil engineering have placed higher requirements on steel quality [1,2]. In metallurgical fields, considerable attention has been paid to the physical and chemical properties of ladle furnace (LF) refining slag [3–6]. At present, refining processes use a series of mature refining technologies, such as vacuum degassing, breathable brick blowing argon, and synthetic refining slag. Among these, a good refining slag system can give better overall refining effects [7–10].

The composition of synthetic refining slags directly affects smelting products. Refining slags are mainly composed of several oxides, including CaO, SiO₂, MgO, Al₂O₃, and FeO. The composition of refining slags must be adjusted. For example, by adjusting the contents of CaO and SiO₂ in the refining slag, the basicity of the refining slag can be accurately controlled. Yu et al. studied the desulfurization ability of refining slag with medium basicity. They found that SiO₂-CaO-MgO-Al₂O₃ system slag with a basicity of approximately 3.5–5.0 and Al₂O₃ content of approximately 20 wt% has strong desulfurization ability, which meets the requirements of High Strength Low Alloys (HSLA) steel [10]. In addition, Al₂O₃ in the refining slag can effectively adjust the melting point of the CaO-SiO₂-Al₂O₃ ternary slag system, such that the refining slag is in a low melting point region [11].

There are two main ways for the refining slag to adsorb inclusions in steel. One is by non-metallic inclusions in the steel dissolving into slag at the interface of the slag and liquid steel. The non-metallic inclusion in the steel dissolve into the refining slag after contacting the interface of steel and slag or slag droplets, and are absorbed by the refining slag. The adsorption effect of the refining slag on the inclusions mainly occurs at the interface of steel and slag, so the refining slag with good fluidity has a better wetting effect on the inclusions at the interface. It can quickly blend into refining slag, and accelerate the adsorption and removal of inclusions. The other is because the main component of refining slag is oxide melt, and the main component of inclusions in steel is also oxides, so the inclusions in
steel are easily adsorbed by the refining slag [12–14]. The adsorption process is as follows: (1) Inclusions move from the molten steel to the slag interface; (2) inclusions pass through the slag interface; (3) inclusions dissolve into the slag. The process of inclusion removal mainly occurs at the interface of liquid steel and slag [15,16]. Valdez et al. [17] studied the ability of different slag systems to adsorb non-metallic inclusions in steel, and found that good fluidity can improve the adsorption speed of inclusions by refining slags. Ja-Yon Choi et al. [18] studied the dissolution rate of Al$_2$O$_3$ inclusions in molten CaO-SiO$_2$-Al$_2$O$_3$ slag systems. Their experimental results showed that the viscosity of the slag has a great influence on the dissolution rate of Al$_2$O$_3$ inclusions. As the viscosity of the slag increases, the dissolution rate of Al$_2$O$_3$ inclusions decreases. Many studies have found that good fluidity promotes adsorption and dissolution of inclusions in the slag layer. As the amount of slag adsorption of inclusions increases, the viscosity of the slag increases and the ability of the slag layer to adsorb inclusions gradually decreases.

However, most studies have focused on the macroscopic properties of refining slag, including viscosity and melting temperature [18,19]. Changes in microstructure are directly related to variation of the macroscopic properties [20,21]. High temperatures in the smelting process make it difficult to detect the structure and properties of the slag. Some studies have proved that molecular dynamics can be applied to slag research [21–26]. Therefore, this paper uses molecular dynamics simulation to study the influence of the CaO/Al$_2$O$_3$ ratio on the structure and properties of refining slag at a microscopic scale. This paper compares and analyzes factors such as the pair distribution function (PFD), coordination number (CN), microstructural units, and atomic diffusion ability. These results confirm that an increase of the CaO/Al$_2$O$_3$ ratio will cause the microstructure of the refining slag to become more complex, thereby affecting the diffusion ability of different atoms. This result provides guidance for a deeper understanding of the microstructure of the refining slag and support for the removal of inclusions in the refining process.

2. Simulation Method

All simulation process used the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS 29 Mar 2019) [27] with the timetep of 0.001 ps. Successful molecular dynamics simulations require careful choice of the potential function and parameters. The two-body potential function Born-Mayer-Huggins (BMH) [28] has been used in the simulation and confirmed to correctly simulate the structure and properties of molten slag. Its specific function is as follows:

$$E = \frac{q_i q_j}{r_{ij}} + A_{ij} \exp\left(\frac{\sigma - r_{ij}}{\rho}\right) - \frac{C_{ij}}{r_{ij}^6}$$  

(1)

where, $q_i$ and $q_j$ are the selected charges for different atoms and $r_{ij}$ represents the distance between atoms $i$ and $j$. The parameters $\rho$ and $\sigma$ describe different pairs of atoms, which are distance units. The energy parameters $A_{ij}$ and $C_{ij}$ describes the repulsive and van der Waals forces of attraction, respectively. The specific parameters [29] are shown in Table 1.

The main component of the refining slag system studied in this paper is SiO$_2$-Al$_2$O$_3$-CaO-MgO. The samples were divided into five groups with various CaO/Al$_2$O$_3$ ratio as shown in Table 2. Almost 10,000 atoms were created in every simulation model and they were placed in a cubic box with periodic boundary conditions. All the simulation processes were performed in the NVT ensemble, meaning that the particle numbers (N), sample volume (V), and temperature (T) were constant. To ensure an amorphous state for the system, the simulation was started at 4000 K from a random configuration. Then cooled to 1800 K, and maintained for 600 ps to achieve equilibrium. Finally, the simulation was run for 500 ps to collect simulation data, including the atomic coordinate, pair distribution function, coordination number, microstructure unit, bond angle, and mean displacement.
Table 1. Specific parameters for Born-Mayer-Huggins (BMH) potential [29].

| Pair   | \( A_{ij} \) | \( \rho \) | \( \sigma \) | \( C_{ij} \) |
|--------|--------------|---------|---------|----------|
| Si-Si  | \( 3.198 \times 10^{-12} \) | 0.046   | 1.44    | 2430.5   |
| Si-Ca  | \( 4.379 \times 10^{-12} \) | 0.063   | 1.89    | 2218.5   |
| Si-Mg  | \( 4.379 \times 10^{-12} \) | 0.063   | 1.61    | 1432.2   |
| Si-Al  | \( 3.962 \times 10^{-12} \) | 0.057   | 1.51    | 1815.2   |
| Si-O   | \( 1.119 \times 10^{-11} \) | 0.161   | 2.54    | 4467.1   |
| Ca-Ca  | \( 5.561 \times 10^{-12} \) | 0.080   | 2.34    | 2025.0   |
| Ca-Mg  | \( 5.561 \times 10^{-12} \) | 0.080   | 2.07    | 1307.3   |
| Ca-Al  | \( 5.144 \times 10^{-12} \) | 0.074   | 1.96    | 1656.9   |
| Ca-O   | \( 1.237 \times 10^{-11} \) | 0.178   | 2.99    | 4077.5   |
| Mg-Mg  | \( 5.561 \times 10^{-12} \) | 0.080   | 1.79    | 843.9    |
| Mg-Al  | \( 5.144 \times 10^{-12} \) | 0.074   | 1.68    | 1069.6   |
| Mg-O   | \( 1.237 \times 10^{-11} \) | 0.178   | 2.72    | 2632.2   |
| Al-Al  | \( 4.727 \times 10^{-12} \) | 0.068   | 1.57    | 1355.7   |
| Al-O   | \( 1.196 \times 10^{-11} \) | 0.172   | 2.61    | 3336.3   |
| O-O    | \( 1.919 \times 10^{-11} \) | 0.276   | 3.64    | 8210.2   |

Table 2. Composition of different simulation group.

|       | CaO   | Al_{2}O_{3} | SiO_{2} | MgO   | CaO/Al_{2}O_{3} Ratio |
|-------|-------|-------------|---------|-------|-----------------------|
| R-1   | 31.88 | 53.13       | 5.00    | 10.00 | 0.6                   |
| R-2   | 37.78 | 47.22       | 5.00    | 10.00 | 0.8                   |
| R-3   | 42.50 | 42.50       | 5.00    | 10.00 | 1.0                   |
| R-4   | 46.36 | 38.64       | 5.00    | 10.00 | 1.2                   |
| R-5   | 49.58 | 35.42       | 5.00    | 10.00 | 1.4                   |

3. Results and Discussion

3.1. Pair Distribution Function and Coordination Number

The pair distribution function (PDF) can be used to reveal the short-range order degree of melt slag, thereby establishing a relationship between atomic structure and macroscopic properties [30,31]. This describes the local atomic structure in the spherical range around an atom. The equation describing the PDF is as follows:

\[
g_{ij}(r) = \frac{V N_i N_j}{4 \pi r^2 \Delta r} \sum \langle n_{ij}(r - \Delta r/2, r + \Delta r/2) \rangle
\]

where, \( N_i(N_j) \) is the total number of atoms \( i(j) \) in the system; \( V \) is the volume of the system; \( n_{ij}(r - \Delta r/2, r + \Delta r/2) \) is the number of atoms \( j \) within a radius of \( r \pm \Delta r \) around the atom \( i \). The PDF provide a lot of basic structural information, such as bond length, bond angle and coordination number.

Figure 1 shows the curves of the PDFs of Si-O, Al-O, Ca-O, and Mg-O with various CaO/Al_{2}O_{3} ratios. The abscissa of the first peak position of the PDF represents the average bond length of two atoms. The average bond lengths of Si-O and Al-O were approximately 1.619 and 1.731 Å, respectively. The intensity of the PDF of the first peak of Si-O was the strongest, indicating that the Si-O bond length is the most stable. As the CaO/Al_{2}O_{3} ratio was increased, the bond lengths of Si-O and Al-O did not change. The Si-O and Al-O bonds are covalent bonds in aluminosilicate systems, whereas Ca-O and Mg-O are the ionic bonds. Therefore, aluminosilicate is mainly composed of a network structure of Si, Al, and O. A higher CaO/Al_{2}O_{3} ratio did not notably affect the bonding strength of the network structure. Furthermore, the PDF shows that the bond lengths of Ca-O and Mg-O were approximately 2.344 and 1.969 Å, respectively. The ionic bonding had a very low bonding strength. The variation of the CaO/Al_{2}O_{3} ratio did not affect the ionic bond length.
By integrating the PDF, the average coordination number (CN) of a given atom at a given distance was obtained. Generally, the average coordination number corresponding to the first trough of the PDF is considered to be the coordination number between two atoms, calculated as follows:

$$N_{ij}(r) = \frac{4\pi N_{ij}}{V} \int_{0}^{r} r^2 g_{ij}(r) dr$$  \hspace{1cm} (3)

Figure 2 shows the curve of the coordination number of Si-O and Al-O at various CaO/Al$_2$O$_3$ ratios. The coordination numbers of Si-O or Al-O were determined based on the first minima of the corresponding PDF. The coordination numbers of Si-O and Al-O at various CaO/Al$_2$O$_3$ ratios were in the ranges of 4.082–4.101 and 4.451–4.471, respectively. Therefore, the coordination number of Si-O was closer to 4, indicating that most of the Si-O exists in a tetrahedral structure. The coordination number of Al-O was greater than 4, indicating that Al-O has a structure with multiple coordination numbers in the system. On increasing the CaO/Al$_2$O$_3$ ratio, the coordination number of Si-O slightly increased, indicating that the CaO/Al$_2$O$_3$ ratio slightly weakened the stability of the SiO$_4$ tetrahedron.

3.2. Evolution of Oxygen Units

The microstructure of the refining slag is composed of network structure of three atoms: Si, Al, and O. The main structural units were SiO$_4$ tetrahedron and AlO$_4$ tetrahedron.
Oxygen plays an important role in the micro network structure. Oxygen units can be mainly divided into four types: free oxygen (FO), non-bridging oxygen (NBO), bridging oxygen (BO), and tricluster oxygen (TO) [32,33]. Illustrations of the different oxygen units are shown in Figure 3. FO does not participate in the formation of a tetrahedral structure and is freely present in the network structure. NBO exists at the corners of the tetrahedral structure, and only connects one Si or Al atom. BO connects two tetrahedrons, and acts as a connection point, including three types: Si-O-Si, Si-O-Al, and Al-O-Al. The higher the BO content, the more tetrahedrons are connected together, and the greater the degree of polymerization (DOP). TO is oxygen that connects three tetrahedral structures and can be divided into four forms: OSSS, OSSA, OSAA, and OAAA. The higher the content of TO, the more complex the structure of the system.

Figure 3. Illustration of different oxygen units.

Figure 4 shows the evolution of different oxygen units with various CaO/Al₂O₃ ratios. The increase of the CaO/Al₂O₃ ratio caused a decrease of FO and NBO content, and an increase of BO and TO content. Therefore, higher CaO/Al₂O₃ ratio promoted the conversion of FO and NBO to BO and TO. The CaO/Al₂O₃ ratio increased the complexity of the system, thereby increasing the degree of polymerization of the system.

Figure 4. Evolution of different oxygen unit with various CaO/Al₂O₃ ratios.
From the content analysis, at a higher CaO/Al₂O₃ ratio, the overall CaO content of the system increased. Previous studies have shown that CaO has the effect of a depolymerization system, and an increase in CaO usually causes a decrease in the degree of polymerization of the system [34,35]. However, in the content range of this research system, a low SiO₂ content means that the main structure of the network is Al₂O₃. The normal slag structure composed of SiO₄ tetrahedron is relatively stable. The appearance of Al₂O₃ changes the silicate network, with some Al³⁺ ions replacing Si⁴⁺ ions. The difference in the charge of Al³⁺ ions and Si⁴⁺ ions, requires an additional amount of IA (The first main group atom) and IIA (The second main group atom) cations in the slag to provide charge compensation for Al³⁺ ions. Ions that provide charge compensation are no longer used as network modifiers. Therefore, in this paper, the increase of the CaO/Al₂O₃ ratio produces a large number of Ca²⁺ ions in the system, which provides sufficient charge compensation. Hence, more Al³⁺ ions can form AlO₄ tetrahedron structures, which promote the transformation of FO + NBO to TO + BO.

Here, BO are grouped into three types namely: Al-O-Al, Al-O-Si, and Si-O-Si. The specific evolution is shown in the Figure 5. The content of Al-O-Si and Si-O-Si remained basically constant, and the content of Al-O-Al increased at higher CaO/Al₂O₃ ratios. The reason for this is that the SiO₂ content remains constant in all systems. The increase of the CaO/Al₂O₃ ratio provides more charge compensation for Al³⁺ ions, which leads to more Al₂O₃ participating in the construction of the network structure.

![Figure 5. Evolution of different bridging oxygens with various CaO/Al₂O₃ ratios.](image)

In this paper, TO only has three forms of O-AAA, O-AAS, and O-ASS. The specific variation is shown in Figure 6. Because the SiO₂ content is relatively small, no O-SSS TO appeared in this research system. From the results in the figure, it indicated that as the CaO/Al₂O₃ ratio was increased, the O-ASS content of two SiO₄ tetrahedron does not change. The O-AAS and O-AAA content showed an upward trend, and the O-AAS content with SiO₄ tetrahedron participation increased more slowly. Thus, the presence of SiO₄ slightly prevented Al₂O₃ from forming TO. When only Al₂O₃ was present in the system, the TO of the system increased rapidly as the CaO/Al₂O₃ ratio was increased.
3.3. Distribution of n-Coordinated Al and $Q_n$

The evolution of different types of Al atoms is shown in Figure 7. The coordination number of Al-O was mainly concentrated over four coordination types, namely 2, 3, 4, and 5-coordinated Al, which accounted for more than 90% of the total content. The content of the 1- and 6-coordinated Al changed very little, indicating that an increase of the CaO/Al$_2$O$_3$ ratio does not notably affect the formation of these two complexes. However, at higher CaO/Al$_2$O$_3$ ratio, the contents of 4- and 5-coordinated Al tended to increase, whereas the contents of 2- and 3-coordinated Al decreased. This results can be explained by the increase of CaO/Al$_2$O$_3$ promoting conversion of 2- and 3-coordinated Al to 4- and 5-coordinated Al. A higher CaO/Al$_2$O$_3$ ratio provides more Ca$^{2+}$ for the system, which gives sufficient charge compensation. Thus, the content of 4- and 5-coordinated Al increases. The 4-coordinated Al with sufficient charge compensation is relatively stable and hence, it has the highest content.
The DOP can be also described by $Q_n$, which is related to the properties of the refining slag ($n$ is the number of bridging oxygens within a tetrahedron). The variation of $Q_n$ for Si and Al at different CaO/Al$_2$O$_3$ ratios is shown in Figure 8. Figure 8a shows that the $Q_n$ value for Si is mainly composed of two forms of $Q_0$ and $Q_4$. The remaining $Q_n$ contents are relatively low and are less strongly affected by the CaO/Al$_2$O$_3$ ratio. The two forms of $Q_0$(Si) and $Q_4$(Si) mutually transformed on changing the CaO/Al$_2$O$_3$ ratio. The evolution of $Q_n$(Al) is shown in Figure 8b. The contents of both $Q_0$(Al) and $Q_3$(Al) were less affected by the variation of the CaO/Al$_2$O$_3$ ratios. When the CaO/Al$_2$O$_3$ ratio was increased, the contents of $Q_1$(Al) and $Q_2$(Al) tended to decrease, while the contents of $Q_4$(Al) and $Q_5$(Al) increased. Therefore, the conversion of $Q_1$(Al) and $Q_2$(Al) to $Q_4$(Al) and $Q_5$(Al), gradually increased the complexity of the system, and the DOP of the system gradually increased.

![Figure 8. Variation of $Q_n$ with different CaO/Al$_2$O$_3$ ratios: (a) $Q_n$ of Si, and (b) $Q_n$ of Al.](image)

**3.4. Analysis of Bond Angle**

Previous studies have identified aluminosilicates are composed of microscopic network structures with the three elements of Si, Al, and O as connection points. To clarify the microstructural composition of the system, the distribution of bond angles is shown in Figure 9. Because the bonds formed by Ca, Mg, and O are generally ionic bonds, the bond angles of Ca, Mg, and O are not analyzed. Through the analysis of the previous chapters, it has been proved that the microstructural units in aluminosilicate are mainly SiO$_4$ and AlO$_4$. As can be seen from Figure 9a, the bond angles of O-Si-O are mainly distributed around 105.5°, whereas the bond angle O-X-O of a standard tetrahedron is 109°. The reason for this effect is a small content of structures other than SiO$_4$ in the microstructural unit composed of Si. However, the SiO$_4$ tetrahedral structure was the most stable in the aluminosilicate melt. As observed from Figure 9b, the O-Al-O bond angles were distributed in the range of 73.5° and greater. This is because of the existence of multiple types of Al-O coordination. Hence, the Al-O structure unit is unstable. As can be seen from Figure 9c,d, the bond angles of Al-O-Al and Al-O-Si are distributed around 88.5° and 96.5°, respectively. In summary, the change of CaO/Al$_2$O$_3$ ratio does not change the bond angles of the microstructure of the system.
3.5. The Variation of Slag Transport Properties

The average displacement of different atoms in a certain period of time is calculated by examining the atomic displacements over a certain period of time. The calculation formula is as follows:

\[ MSD = \left\langle |r(L) - r(0)|^2 \right\rangle = \frac{1}{NM} \sum_{i} \left| r_i(t_k + L) - r_i(t_k) \right|^2 \]  \( (4) \)

where \( N \) is the total number of atoms, \( L \) is the time parameter. \( M \) is the number of the time series; \( t_k \) is the start time of the Kth time; \( r_i(t_k + L) \) and \( r_i(t_k) \) represent the positions of the atoms at time \( t_k + L \) and \( t_k \), respectively. The self-diffusion coefficient of an atom can be calculated by the Einstein’s equation. The specific calculation is as follows:

\[ D = \frac{1}{6L} MSD \]  \( (5) \)

The variation of the MSD of each element indicated that the system was in a molten state. Changes of the atoms in the five sets of simulations were basically similar, only the MSD of R-3 is shown in Figure 10a. Through variation of the MSD, the diffusion capacity of different elements decreased in the order: Mg > Ca > O > Al > Si. Mg and Ca only formed ionic bonds with O. Therefore, the diffusion capacities of Mg and Ca atoms were the highest. The atomic radius of Mg was smaller than that of Ca, such that the diffusion capacity of Mg was highest. Because of the nature of SiO\(_4\) and AlO\(_4\) structure units in the aluminosilicate, the diffusion capacities of Si, Al, and O are ranked lower. In general, the diffusion capacity of O ranked between that of Si and Al. However, in this system, the high content of FO gave a diffusion capacity of O that is markedly higher than that of Al and Si. Because SiO\(_4\) tetrahedron are more stable, the diffusion capacity of Al is higher than that of Si.

In addition, changes of the MSD cause variation of the self-diffusion coefficient of each element, as shown in Figure 10b. As the CaO/Al\(_2\)O\(_3\) ratio increases, the self-diffusion coefficient of every atom basically showed a downward trend. Hence, an increase in the CaO/Al\(_2\)O\(_3\) ratio caused the overall diffusion capacity of the system to decrease, resulting in a decrease in the fluidity of the system. In previous experimental studies, it has been found that the viscosity of the slag will increase with the increase of the CaO/Al\(_2\)O\(_3\) ratio.
when the alkalinity remains unchanged, as shown in Figure S1. Ca\(^{2+}\) not only acts as a network modifier ion in aluminosilicate system (the network modifier ion will destroy the microstructure network). Meanwhile, Ca\(^{2+}\) ions can also act as a charge compensator to provide charge compensation for [AlO\(_4\)]\(^{5-}\), thereby making microscopic network structure more stable. Once acted as a network modifier, it can no longer act as a charge compensator. In addition, Al\(_2\)O\(_3\) is an amphoteric oxide, which can act as a network former and a network modifier in the slag. In the case of sufficient CaO content, more Al\(_2\)O\(_3\) can be promoted as a network former, thereby increasing the degree of polymerization. Hence, the viscosity of the slag will increase. In order to further verify the correctness of the results, this paper further calculated the melting point of the research system, as shown in Figure S2. As the CaO/Al\(_2\)O\(_3\) ratio increases, the melting point of the system tends to rise. This will also cause the fluidity of the slag to deteriorate.

![Figure 10. Variation of (a) mean square distribution and (b) self-diffusion coefficients of different atoms with various CaO/Al\(_2\)O\(_3\) ratios.](image)

### 4. Conclusions

Here, molecular dynamics simulation methods are used to study the effects of CaO/Al\(_2\)O\(_3\) ratio on the structural properties of refining slag. The conclusions are as follows:

1. The micro-structure of refining slag is composed of micro network structure of Si-O and Al-O covalent bonds, with Ca-O and Mg-O ionic bonds. The bond lengths of Si-O, Al-O, Ca-O, and Mg-O in the refining slag system were 1.619Å, 1.731Å, 2.344Å, and 1.969Å, respectively. An increase in the CaO/Al\(_2\)O\(_3\) ratio did not affect the bond length.

2. At higher CaO/Al\(_2\)O\(_3\) ratios, more charge compensating Ca\(^{2+}\) ions entered the slag, which provided sufficient charge compensation for Al\(^{3+}\). Therefore, more Al\(^{3+}\) acted as a network former in the slag, which promotes conversion of FO and NBO to TO and BO, thereby increasing the degree of polymerization of the system.

3. A higher CaO/Al\(_2\)O\(_3\) ratio induced more 2- and 3-coordinate Al to convert to 4- and 5-coordinate Al. The introduction of more CaO provided a charge compensator for Al\(^{3+}\) ions, which increased the 4- and 5-coordinate Al content.

4. In the micro-structure of refining slag system, the bond angles of O-Si-O, O-Al-O, Al-O-Al, and Al-O-Si are approximately 105.5°, 73.5°, 88.5°, and 96.5°, respectively. The CaO/Al\(_2\)O\(_3\) ratio did not affect the bond angle.

5. Through analysis of the mean square distribution of various atoms, the self-diffusion capacities of different atoms decreased in the order: Mg > Ca > O > Al > Si. A higher CaO/Al\(_2\)O\(_3\) ratio led to the conversion of the simple microstructure to a more complex microstructure (such as, FO and NBO to TO and BO), thereby causing a decrease of the diffusion capacity in the refining slag system.
Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3905/min11040398/s1, Figure S1: The variation of refining slag viscosity with different CaO/Al2O3 ratio., Figure S2: The variation of melting point of refining slag with different CaO/Al2O3 ratio. References [34,35] are cited in the Supplementary Materials.

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