

Local structure and dynamics of impurities Fe, Al and Mn in melt beryllium

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Abstract: In this paper, the local structure and dynamics of impurities Fe, Al and Mn in beryllium were investigated on an atomic scale using ab initio molecular dynamics and statistical physics methods. The analysis of the radial distribution function centered on impurity atoms showed that the density of beryllium atoms around Fe and Mn is 8.4% and 8.6% higher than that of beryllium around Al, respectively. The statistics of the measure square displacement of impurity atoms showed that the diffusion coefficients of Al atoms are 114% and 133% larger than those of Fe and Mn atoms in the melt beryllium, respectively. Statistical analysis of velocity autocorrelation function of impurity atom showed that Fe and Mn atoms collided strongly with beryllium atoms in the first coordination layer, indicating that they were tightly surrounded and bound by the surrounding beryllium atoms in the central position, while the beryllium atoms around Al are loosely arranged and have weak binding forces with Al. The analysis of the activity coefficients of the impurities showed that when Fe or Mn entered the melt beryllium, it reduced the free energy of the system, while when Al entered, it increased the system energy. In summary, the interatomic force of BeAl is weak, so they do not form intermetallic compounds, and Al diffuses quickly in beryllium. While BeFe, BeMn have strong interatomic forces, and tend to form more BeFe and BeMn bonds to reduce the free energy of the system, thus Fe, Mn diffuse slowly in beryllium. Ab initio molecular dynamics can be used to forecast the best experimental temperature for the vacuum distillation of beryllium. It provides an efficient and convenient means to guide the purification of beryllium.

Keywords: Beryllium, Purification, Ab initio molecular dynamics, Local structure, Activity coefficients

1 Introduction

High-purity beryllium is often used for the P-type doping of semiconductor materials, and through the formation of PN junctions, multi-purpose devices are prepared, including the first generation of silicon devices, the second generation of

GaAs, InP-based devices, the third generation of GaN, SiC, ZnO devices, etc., and in the infrared field, including the second class of superlattice devices, InSb devices, etc., it is an important doping element for the preparation of semiconductor devices. At present, high-purity beryllium has been listed as a strategic material by many countries. The outer electron configuration of the beryllium nucleus is 2, and the beryllium atom forms four covalent bonds with the surrounding four

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semiconductor atoms, but two holes are formed due to the lack of two valence electrons, so the doped semiconductor formed is a P-type semiconductor. Beryllium has the advantages of small atomic weight, easy migration to replace trivalent metals, high activation rate, and various doping forms. The beryllium acceptor in gallium nitride was the subject of intensive research at the end of the nineties. In 2013, the GaN:Be crystals were grown and a white light emitting diode was fabricated, and obtained much higher quantum efficiency [1]. Denis O. Demchenko et al using theory and experiment addressed a possible p-type doping route of GaN using the Be acceptor [2]. However, high-purity beryllium is difficult to obtain because the melting point of beryllium is high, and the physicochemical properties of impurity atoms vary greatly. Since the development of nuclear energy in the forties of the last century, the demand for pure beryllium has expanded rapidly, coupled with other industrial fields with the increase in the application of the field, the beryllium industry has developed rapidly, and China, Russia, and the United States have a considerable scale of beryllium industrial countries. According to the literature, the United States and the former Soviet Union have prepared 99.99% high-purity beryllium. In 2011, Materion, Inc., USA released UHP-9999 grade beryllium material, which has a purity of 99.99%, but about the manufacturing process of this grade of beryllium, details of the production technology and mechanical and physical properties have not yet been reported. Vacuum distillation is an effective method for purifying beryllium metal, which has the advantages of simple equipment, short operation process and low pollution. According to public reports, both Russia, and the United States use vacuum distillation purification method. Beryllium metal materials are strategically sensitive materials, so the key process and technology for purification is strictly blocked.

According to the results of our preliminary experiments, vacuum distillation can separate most of the impurities in beryllium, but Fe, Mn etc. are difficult to completely remove. There are some basic theoretical problems in the purification process, such as Mn and Be vapor pressure is very different, but it is difficult to remove, while Al and Be vapor pressure are close but easy to separate. Distillation purification includes two physical processes of volatilization and diffusion, the host metal and impurities volatilize on the melt surface, and its volatilization rate is different due to the difference in vapor pressure of different elements, thereby forming a concentration gradient of impurity in the beryllium melt. Under the action of concentration gradient, impurity atoms diffuse and migrate in the beryllium melt. The local microstructure and dynamics characteristics of different impurity atoms in the melt beryllium are key factors affecting their diffusion migration, which in turn affects their separation behavior and purification.

Ab initio molecular dynamics combines first principles with molecular dynamics simulation, directly calculates the inter-

action between all atoms through quantum chemical methods, and has high calculation accuracy, which is an effective method for simulating liquid metals. Molecular dynamics simulations using a third generation of charge-optimized many body potential were performed to calculate the solid-liquid interface free energy and its anisotropy of Al [3]. The effects of refractory elements on the structure and diffusivity of liquid Ni and vibrational motion of ions of BaTiO₃ have been investigated by ab initio molecular dynamics simulations [4-5]. To solve the dilemma of accuracy versus efficiency, interatomic potential through machine learning is proposed and has raised much attention now, the molecular dynamics simulations of the deep learning potential was used to investigate the microstructure and property of materials [6-9]. Sheng H.W. et al use a combination of state-of-the-art experimental and computational techniques to resolve the atomic-level structure of amorphous alloys and elucidate the different types of short-range order as well as the nature of the medium-range order [10]. Ab initio molecular dynamics can be used to forecast the best experimental temperature for the vacuum distillation of alloys, and it provides an efficient and convenient means of guiding vacuum metallurgy [11]. Al, Fe and Mn are the main impurities in beryllium, and their trend and evolution in melt beryllium have typical characteristics, so they were selected as the research objects of this paper. In this paper, the local structure and separate behavior of impurities Fe, Al and Mn in beryllium were investigated on an atomic scale using ab initio molecular dynamics and statistical physics methods, the relevant basic theoretical data, including the activity characteristics of impurities Al, Fe and Mn in beryllium, diffusion migration and local microstructure, etc., are clarified, which guide the establishment of beryllium purification methods and processes.

2 Computational methods

The computer simulation experiments in this study were performed by using ab initio molecular dynamics (AIMD) [12-16]. The ab initio molecular dynamics simulation was performed using VASP software. The results of the molecular dynamics simulation were processed by vaspkit software to obtain the microstructure and kinetic properties of the system. The structure is displayed using VESTA software. The ion-electron interaction is described by ultra-soft Vanderbilt pseudopotentials (US-PP) and projector augmented waves (PAWs) with a PW91 exchange-correlation function of the Brillouin zone. The simulations were performed in a canonical ensemble with fixed particle number, volume and temperature (NVT) at the Γ point, and the time step was set at 1 fs. The plane wave cut-off energy was 360 eV, and the energy convergence criterion of self-consistency was 1×10^{-4} meV/atom. The calculation accuracy is selected as normal.

The configuration in this study contains 108 atoms (107 Be atoms and 1 impurity atom), which was expressed as Be₁₀₇M

(M=Fe, Al, Mn). The initial configuration of the above system was constructed with 108 atoms randomly distributed in a cube box and equilibrated at 1573K, 1673K, 1773K, respectively, by the Nose-Hoover thermostat. The densities of the molten Be107M alloy were obtained by some test calculations. Firstly, the density of molten Be107M of 1573K, 1673K, and 1773K are preliminarily estimated by the theoretical density and expansion coefficient, and then the initial configuration is calculated by test. Then, on the basis of the above initial configuration, the NPT ensemble is used to calculate the simulated cell volume under different pressures for 500 steps, with high accuracy and truncation energy of 520eV to obtain the relationship between pressure and volume. Then, the quadratic polynomial is used to fit the pressure volume trend line to obtain the volume under zero pressure [17-20]. The supercell volume at different pressures and temperatures are shown in Fig. 1. Finally the size and density of the simulated cell under zero pressure are calculated, and they are used as the size and density of the simulated system. The size of the supercell used in the calculations and the density of each system obtained at different temperatures are listed in Table 1.

Table 1 The length of cube box and density of four systems at different temperatures under zero pressure: (a) Be108, (b) Be107Al, (c) Be107Fe, (d) Be107Mn

Syst	Temp(K)	Leng(Å)	Density(g/cm ³)	Syst	Temp(K)	Leng(Å)	Density(g/cm ³)
Be108	1573	10.192	1.533	Be107Al	157	10.18	1.564
	1673	10.199	1.529		167	10.20	1.555
	1773	10.203	1.528		177	10.21	1.549
Be107Fe	1573	10.177	1.613	Be107Mn	157	10.19	1.605
	1673	10.187	1.609		167	10.20	1.601
	1773	10.217	1.595		177	10.21	1.593

ar dynamics simulation runs 8000 steps at different temperatures in four systems. The last 4000 steps were analyzed by statistical physics to obtain the characteristic information of impurities in the melt beryllium, including local microstructure and dynamics characteristics.

3 Results and discussion

3.1 Local structure

Radial distribution functions are the most commonly used mathematical language for describing the microstructure of liquid and amorphous materials. The radial distribution function $g_{AB}(r)$ represents the probability of finding class B atoms in a spherical shell with a distance of r from the central atom of class A and a thickness of δr , and the ratio of the probability when class B atoms are evenly distributed throughout the simulated system. $g_{AB}(r)$ can understand the structural information of the system, such as atomic radius, average spacing between atoms, and coordination number, so as to obtain the interrelationship between class A atoms and class B atoms. $g_{AB}(r)$ is defined as [21-23]:

$$g_{AB}(r) = \frac{L^3}{N_A N_B} \left\langle \sum_{i=1}^{N_A} \frac{n_{iB}(r)}{4\pi r^2 \Delta r} \right\rangle \quad (1)$$

In the formula (1), A and B are particle types; N_A and N_B are the number of A particles and B particles, respectively. $n_{iB}(r)$ is the number of B particles in the $r \sim r + \delta r$ spherical shell centered on A particles.

The last 4000 frames of the trajectory of the MD simulation were analyzed, and the radial distribution function centered on the impurity atoms Fe, Al and Mn was obtained in Fig. 2. In the radial distribution function curve, the strength of the first peak reflects the strength of the bond between the impurity atom and the Be atom. It can be seen from Fig. 2 that the first peak strength of Be107Fe and Be107Mn are relatively large, Be108 is in the middle, and Be107Al is the smallest, indicating that Be atoms centered on the Fe and Mn are tightly stacked, and the bond of BeFe and BeMn are strong, while the Be atoms centered on the Al are stacked loosely, and the bond of BeAl is weak. [24]:

$$N_{AB} = \int_0^{r_{\min}} 4\pi \rho_B r^2 g_{AB}(r) dr \quad (2)$$

The coordination number N_{M-Be} of the impurity atom Fe, Al and Mn is calculated by formula (2). To express the compactness of the cluster of B atoms centered on A atoms, the atomic number density ρ_{AB} is defined as the density of the number of B atoms in a sphere with radius r_{\min} from the central atoms A, calculated as [18]:

$$\rho_{AB} = \frac{N_{AB}}{4/3\pi r^3} \quad (3)$$

So far, the structural parameters of melt beryllium are calculated, including the first peak radius r_{\max} , the first valley radius r_{\min} , the coordination number N_{M-Be} , and the atomic number density ρ_{M-Be} , as shown in Table 2, the results show that the Be atoms centered on the Fe and Mn are compact, and the Be atoms centered on the Al are loose.

Table 2 r_{max} , r_{min} , NM-Be, ρ M-Be ($N/\text{\AA}^3$) of the three systems at different temperatures

Syst.	Be107Fe		Be107Al		Be107Mn	
	1573K	1773K	1573K	1773K	1573K	1773K
r_{max}	2.2785	2.2782	2.5281	2.5124	2.3174	2.3813
r_{min}	3.2330	3.2459	3.4265	3.4221	3.2829	3.2853
NM-Be	14.66	14.12	15.87	15.48	14.90	15.14
ρ M-Be	0.10357	0.09857	0.09417	0.09222	0.10050	0.10193

3.2 Dynamics characteristics

3.2.1 Mean square displacement

The diffusion coefficient of the impurities Fe, Mn and Al in melt beryllium was calculated by the mean squared displacement (MSD) method with the Einstein equation [25-27]:

$$(4)$$

Where $r_i(t)$ and $r_i(0)$ are the coordinates of particle i at the arbitrary origin of time and t later, respectively. The MSD curves of the impurity atom Fe, Mn and Al at 4000 fs-4500 fs, 5000 fs-5500 fs, 6000 fs-6500 fs, 7000 fs-7500 fs at 1673 K are shown in Fig. 3. In this statistic, the displacement of the same impurity atom was tracked and analyzed. Statistics at different time starts of the same impurity atom are independent statistics, equivalent to statistics for large groups of particles. To improve the statistical accuracy and avoid the influence of statistical noise, we calculated the diffusion coefficient every 500 steps (0.5 ps) and obtained the total average values. It can be seen from the figure that Al atoms move fastest in melt beryllium, while Fe atoms move slower and Mn atoms move the slowest. The diffusion coefficient of the impurities Fe, Mn and Al in melt beryllium at 1673 K calculated according to formula (6) are shown in table 3. The diffusion coefficients of Fe, Mn, and Al atoms in melt beryllium at 1673K are $0.367 \text{\AA}^2/\text{ps}$, $0.337 \text{\AA}^2/\text{ps}$, and $0.787 \text{\AA}^2/\text{ps}$, respectively. The results show that Al diffuses fast in melt beryllium, while Fe and Mn diffuse slowly. The results of the dynamics analysis of the impurity atoms are consistent with the results of the above-mentioned local microstructure analysis. In the melt beryllium, the beryllium atoms around the Al atom are loosely arranged and the binding force is weak, so Al diffuses quickly; The beryllium atoms around Fe and Mn atoms are closely arranged and have strong binding forces, so Fe and Mn diffuse slowly.

Fig. 3 MSD of four independent statistics: (a) 4000 fs – 4500 fs, (b) 5000 fs - 5500 fs, (c) 6000 fs -6500 fs, (d) 7000 fs -7500 fs at 1673 K

Table 3 MSD and diffusion coefficient (D) of impurities Fe, Mn, and Al in melt beryllium of 1673K

	Fe		Mn		Al	
	1.1	1.09	0.7	0.8	1.3	2.2
MSD (\AA^2)	3	09	6	8	6	10
		4			6	4
		0			4	9
Avg (\AA^2)	1.10		1.01		2.36	
D ($\text{\AA}^2/\text{ps}$)	0.367		0.337		0.787	

3.2.2 Velocity autocorrelation function

The time correlation function is an important quality of thermodynamic systems, which indicates the degree of correlation between a specific physical quantity at the current moment and a physical quantity at an earlier time, reflecting the causal relationship between them, and is closely related to the migration and mass transfer process of thermodynamic system. The velocity autocorrelation function (VACF) is defined as the degree to which the current velocity of a particular atom in a simulated system correlates with the velocity at an earlier time [28]:

$$\text{VACF}(t) = V_i(t) \cdot V_i(t_0+t) \quad (5)$$

In this experiment, to improve the statistical accuracy and avoid the influence of statistical noise, VACF statistics were performed for different time starting points of the same impurity atom, and the time periods of four independent statistics were 4000fs-4500fs, 5000fs-5500fs, 6000fs-6500fs, 7000fs-7500fs, as shown in Fig. 5. As can be seen from the figure, the velocity of the impurity atom is completely auto-correlated at $t=0$, and the velocity autocorrelation function value is 1. When t is greater than 0, the autocorrelation of the atom quickly disappears, decays to 0, and then oscillates rapidly with a large margin. It indicates that the impurity atom loses all momentum in a strong collision and bounces in the opposite direction of the pre-collision momentum. VACF reacts the momentum exchange of impurity atom when it collides with the surrounding Be atoms, which is only related to the characteristics of Be atoms with direct collision and momentum exchange in the first coordination layer, and has less relationship with the characteristics of Be atoms outside the first coordination layer where there is no direct momentum

exchange and only indirect momentum exchange. Looking at the four independent statistics in Fig. 5, Al atoms have the slowest initial velocity decay and weakest rebound, while Fe atoms decay faster and rebound strongly, and Mn decay is the fastest and rebound is the strongest. It shows that Fe and Mn are tightly wrapped in the center position by the coordination atom Be and oscillate at a higher frequency, and it is difficult to break away from the encirclement formed by the coordination atom Be. The beryllium atoms around Al are looser, the collision effect is weak, and the rebound is also weak. The analysis of velocity autocorrelation function is consistent with the conclusions of local microstructure analysis and diffusion coefficient analysis.

The statistics of the time when the initial momentum of the impurity atom decayed to zero from above-mentioned velocity autocorrelation function are shown in Table 4. In the four statistics, the average time when the momentum of impurity atom decays to zero at 1673K were 52fs, 45.5fs, and 41fs, respectively, indicating that the beryllium atoms around Al are loosely arranged and have weak binding forces, while the beryllium atoms around Fe and Mn are tightly arranged and have strong binding forces.

3.3 Activity characterization

In a thermodynamic system, partial molar free energy ($\frac{\partial G}{\partial N_i}$) is a quantity of strength that represents the intrinsic properties of the material system. The partial molar free energy of component i ($\frac{\partial G}{\partial N_i}$) has the following relationship with the activity coefficient γ_i of component i in this system [29-31]:

$$RT \cdot \ln \gamma_i = \frac{\partial G}{\partial N_i} \quad (6)$$

The activity coefficients of the impurities Fe and Al in beryllium were 0.055 and 4.71 respectively [32], thus,

$$\frac{\partial G}{\partial N_{Fe}} < 0 \quad \frac{\partial G}{\partial N_{Al}} > 0 \quad (7)$$

The intrinsic properties of the Be107M (M=Al, Fe, Mn) system represented by Equation (7) are: Al atom enters the beryllium matrix to increase the free energy of the system, Al-Be interatomic attraction is less than Be-Be atomic attraction, Al atom has the effect of repelling beryllium matrix, the system tends to form less Al-Be bonds to reduce energy, Al and Be are not miscible, do not form intermetallic compounds, while Fe atom enters the beryllium matrix, the free energy of the system decreases, the attraction between Fe-Be atoms is greater than the attraction between Be-Be atoms, Al atom have the effect of shrinking the beryllium matrix, the system tends to form more Fe-Be bonds to reduce energy, Fe and Be are easy to form solid solutions or intermetallic compounds.

There is no interatomic activity coefficient of Be and Mn in the references, and the data are insufficient, so only qualitative estimation can be made. According to the theoretical results of this paper, the interaction force between Be, Mn atoms is stronger than that between Be, Be and Mn, Mn, which is a negative deviation, and this kind of state is characterized by the existence of solid solutions and compounds below the liquidus line. Although the vapor pressure of Be and Mn is very different, it has a strong force with Be. In the vacuum distillation process, Mn is separated by accumulation in the residue.

4 Conclusion

The outer electronic structure of atom determines characteristics of interaction force between atoms. The interaction force of BeAl are weaker than that of BeBe, and the system tends to form fewer BeAl bonds to reduce the system energy. Al tend to be immiscible with beryllium and do not form intermetallic compounds. In the melt beryllium, the Be atoms around the Al are stacked loosely, thus Al diffuses and migrates quickly. The interaction force of BeFe, BeMn are stronger than that of BeBe. The system tends to form more BeFe and BeMn bonds to reduce the free energy of the system. Fe and Mn are easy to form intermetallic compounds or solid solutions with beryllium. In melt beryllium, Fe or Mn atoms are tightly wrapped and bound by the surrounding Be atoms, so they diffuse and migrate slowly. Ab initio molecular dynamics can be used to forecast the best experimental temperature for the vacuum distillation of beryllium. It provides an efficient and convenient means to guide the purification of beryllium.

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熔体铍中杂质 Fe、Al 和 Mn 的局域结构和动力学过程

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摘要: 本文采用从头算分子动力学和统计物理方法, 从原子尺度上研究了铍中杂质 Fe、Al 和 Mn 的局域结构和动力学过程。以杂质原子为中心的径向分布函数分析表明, Fe 和 Mn 周围的铍原子密度分别比 Al 周围的铍原子密度高 8.4% 和 8.6%。杂质原子平方位移的测度统计表明, 熔融铍中 Al 原子的扩散系数分别比 Fe 和 Mn 原子大 114% 和 133%。杂质原子速度自相关函数的统计分析表明, Fe 和 Mn 原子在第一配位层与铍原子发生强烈碰撞, 表明它们在中心位置被周围的铍原子紧密包围和束缚, 而 Al 周围的铍原子排列松散, 与 Al 的结合力较弱。杂质活度系数分析表明, 当 Fe 或 Mn 进入熔体铍时, 它降低了体系的自由能, 而当 Al 进入时, 它增加了体系能量。综上所述, BeAl 原子间作用力较弱, 因此它们不会形成金属间化合物, 并且 Al 在铍中扩散迅速。而 BeFe、BeMn 具有很强的原子间作用力, 并且倾向于形成更多的 BeFe 和 BeMn 键以降低体系的自由能, 因此 Fe、Mn 在铍中扩散缓慢。从头算分子动力学可用于预测铍真空蒸馏的最佳实验温度。它为指导铍的提纯提供了一种有效且方便的方法。

关键词: 铍; 纯化; 从头算分子动力学; 局域结构; 活度系数

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