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ARTICLE

# Molecular Dynamics Simulation of High-Nitrogen Stainless Steel Brazed by AgCuNi Filler Metal

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**Abstract:** The element diffusion process of the binary systems (Fe-Cu and Fe-Ni) in the vacuum brazing of high-nitrogen stainless steel with AgCuNi filler was investigated by Lammps software for molecular dynamics simulation analysis. Results show that the mutual diffusion phenomena of Fe-Cu and Fe-Ni binary systems are obvious, and the thickness of the diffusion layer is increased with increasing the diffusion time. In the Fe-Cu diffusion process, only the mutual atom diffusion occurs; whereas not only the mutual atom diffusion, but also the formation of mesophase occurs in the Fe-Ni diffusion process. In the Fe-Cu binary system, the mean square displacement (MSD) and diffusion coefficient of Fe atom are greater than those of Cu atom, so the diffusion ability of Fe atom is better than that of Cu atom. Similarly, in the Fe-Ni binary system, MSD and diffusion coefficient of Fe atom are greater than those of Ni atom, so the diffusion ability of Fe atom is also better than that of Ni atom. With increasing the diffusion temperature, MSD and diffusion coefficient of atoms are increased, and their diffusion ability is enhanced.

**Key words:** high-nitrogen stainless steel; brazing; AgCuNi filler metal; molecular dynamics simulation; mean square displacement

High-nitrogen stainless steel is used as a structural component due to its properties of high load-bearing capacity and strong impact resistance. The fusion welding methods, such as laser welding and arc welding, are commonly used to prepare the workpiece of high-nitrogen steel[1-3]. In the medical device field, the corrosion resistance of high-nitrogen steel joint is the key factor[4,5]. It is well known that the deterioration of mechanical properties and corrosion resistance of high-nitrogen steel fusion welded joint are mainly associated with the porosity[6,7]. Additionally, the nitrogen loss and welding cracking are the main defects in the fusion welding[8]. The solid-state welding technique of friction stir welding (FSW) has short-period thermal cycles and can result in low peak temperature, therefore effectively reducing the porosity and welding cracking[8]. Consequently, FSW has been widely used to join the high-nitrogen steel. Moreover, the brazing method is also commonly used to join the high-nitrogen steel, especially in the medical fields. The brazing temperature is the main influence factor during the manufacturing of brazed joint. Therefore, it is very necessary to control the brazing temperature of high-nitrogen steel. In order to find the optimal brazing temperature for high-nitrogen steel, the Lammps software was adopted to conduct the molecular dynamics simulation of the matrix elements in high-nitrogen steel, such as Fe, Cu, and Ni[9-11]. The atomic diffusion process in the Fe-Cu and Fe-Ni binary systems was simulated to analyze the formation of intermetallic compounds in this research.

## 1 Experiment

The base metal was high-nitrogen austenitic stainless steel

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with 0.75wt% nitrogen, which was mainly composed of austenite of uniform size with some crystal twinning, as shown in Fig. 1. The structure of the forged base metal consisted of equiaxial grains. The Ag-Cu-Ni filler metal was prepared through the addition of 0.75wt% nickel into the Ag-Cu eutectic filler with the particle size of ~75 μm.

The atomic diffusion simulation was conducted under the conditions of constant temperature, constant pressure, and fixed number of particles in the system at brazing temperatures of 1123, 1223, and 1323 K, i.e., NPT ensemble was adopted in the process of the atomic diffusion simulation of Fe-Cu binary system[12,13]. For the atomic diffusion simulation of Fe-Ni binary system, the temperatures of 1123, 1223, and 1323 K were selected. The standard atmospheric pressure (0.1 MPa) was adopted because no additional pressure was needed during the brazing test. The diffusion model was constructed with crystalline Fe and crystalline Cu, where the lattice constants of Cu and Fe atoms are 3.614 925 05 and 2.855 324 63 nm, respectively. For the molecular dynamics model of Fe-Ni diffusion, the lattice constant of Ni atom is 3.506 486 nm. The supercells with less atoms were established for the higher calculation efficiency and less calculation time. Approximately 10 reduplicate unit cells were established along the *X*, *Y*, and *Z* directions, separately, to construct the supercell with a small distance between the adjacent crystals. The periodic boundary conditions were applied along three directions, and the potential function of embedded atom method[14,15] was adopted to calculate the interatomic force between the Fe and Cu atoms. The potential function including the Fe-Cu-Ni atomic potential[16-19] was used for the analyses of microstructure evolution and mechanical property changes in the steel of reactor pressure vessel as well as the atomic diffusion simulation in this research.

During the diffusion simulation process, the model was relaxed for a period and the temperature was raised to the specified value. The atomic diffusion process was observed through the atom migration and movement at specified temperature.

## 2 Results and Discussion

### 2.1 Fe-Cu and Fe-Ni diffusion processes

The models of Fe-Cu atomic diffusion at 1123 K and 0, 200, 400, and 600 ps are shown in Fig. 2. It can be seen that the gap between Fe and Cu atoms disappears immediately due to the diffusion process. The interface is located at 30 nm on the *Z* axis. It is found that with the atomic diffusion proceeding, the thickness of diffusion area is gradually increased. The Fe atoms firstly start to diffuse into the Cu lattice, then the Fe atoms on the side of the Cu lattice are increased, and only a small number of Cu atoms are diffused into the Fe lattice. The atom content curves of the Fe-Cu diffusion system are basically linear, indicating that these atoms are only diffused into each other in the holding process without the generation of mesophase.

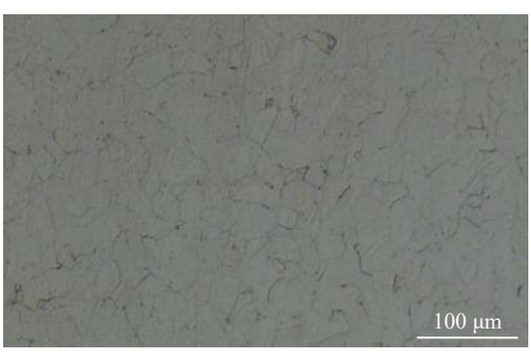


Fig.1 Microstructure of high-nitrogen austenitic stainless steel

The models of Fe-Ni atomic diffusion at 1123 K and 0, 200, 400, and 600 ps are shown in Fig.3. It can be seen that the gap between Fe and Ni atoms disappears immediately. Then the diffusion and migration processes are observed at the interface. The interface is also located at 30 nm on the *Z* axis. The thickness of diffusion area is gradually increased with the atomic diffusion proceeding. The Fe-Ni atomic diffusion diagram usually shows the asymmetric characteristic: more Fe atoms are diffused into the Ni lattice, while only a small number of Ni atoms are diffused into the Fe lattice. It can also be inferred that the diffusion ability of Fe atoms is stronger than that of Ni atoms in the Fe-Ni binary diffusion system. The diffusion area of Fe-Ni system is thicker than that of Fe-Cu binary system under the same conditions.

Moreover, compared with those of the Fe-Cu diffusion system, the atom content curves of Fe-Ni system have a section with smaller slope in the diffusion area. According to Ref. [16], a mesophase is formed in this area during the diffusion process, namely Fe-Ni compound. It is well known that the intermetallic compounds of FeNi and $FeNi_3$ can be easily formed according to the Fe-Ni binary phase diagram, which is consistent with the results of the molecular dynamic simulation.

### 2.2 Atomic potential energy in diffusion process

Based on the molecular dynamics simulation, the potential energy of each atom during the diffusion process can be calculated. The results of atomic potential energy in Fe-Cu diffusion process are shown in Fig. 4. The potential energy of atom at the equilibrium position is zero. When the atom is away from the equilibrium position, the potential energy becomes negative. The smaller the distance between atoms, the greater the deviation from equilibrium position and the greater the absolute value of atomic potential energy. It can be seen that the absolute potential energy value of Fe atoms is greater than that of Cu atoms, which means that the Fe atoms are further away from the equilibrium position and more unstable. The average values of the atomic potential energy of Fe and Cu atoms at different temperatures are shown in Table 1. For the Fe and Cu atoms under the same conditions, the absolute value of atomic potential energy is increased with increasing the temperature, and the atoms become more unstable.

The atomic potential energies of Fe and Ni atoms in diffusion process at different temperatures after 600 ps are

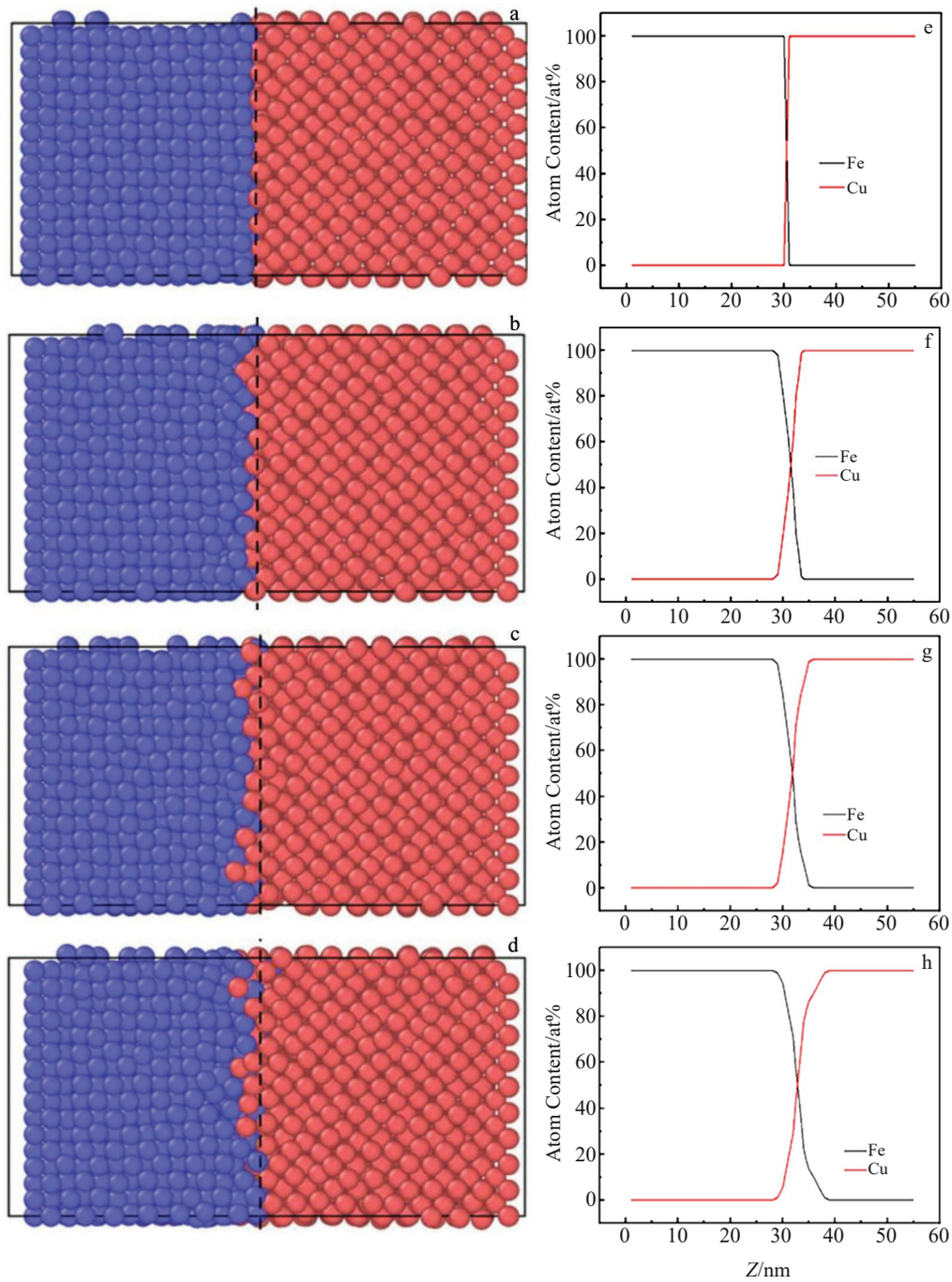


Fig.2 Atomic distribution models (a~d) and atom contents along *Z* axis (e~h) of Fe-Cu binary system at 1123 K and different diffusion time: (a, e) 0 ps, (b, f) 200 ps, (c, g) 400 ps, and (d, h) 600 ps

shown in Fig. 5. It can be seen that the absolute potential energy value of Fe atoms is smaller than that of Ni atoms, which means that the atoms start to move and get away from the equilibrium position since the diffusion occurs. Additionally, the Ni atoms are further away from the equilibrium position and become more unstable. The average values of atomic potential energy of Fe and Ni atoms at different temperatures after 600 ps are shown in Table 2. For the Fe and Ni atoms under the same conditions, the result of atomic potential energy agrees well with that obtained in Fe-Cu diffusion simulation. With increasing the temperature, the absolute value of atomic potential energy is increased, the atoms become more unstable, the barrier is easier to break, and finally the atoms are diffused.

### 2.3 Mean square displacement and diffusion coefficient

The mean square displacement (MSD) indicates the change of particles in time and space, which can be used to explain the diffusion ability of Fe and Cu atoms and to calculate the diffusion coefficient, as follows:

$$\mathrm{MSD} = \frac{1}{N}\sum_{i=1}^{N}|r_i(t) - r_i(0)|^2 \quad (1)$$

where $r_i(t)$ is the position of atom $i$ at time $t$; $r_i(0)$ is the

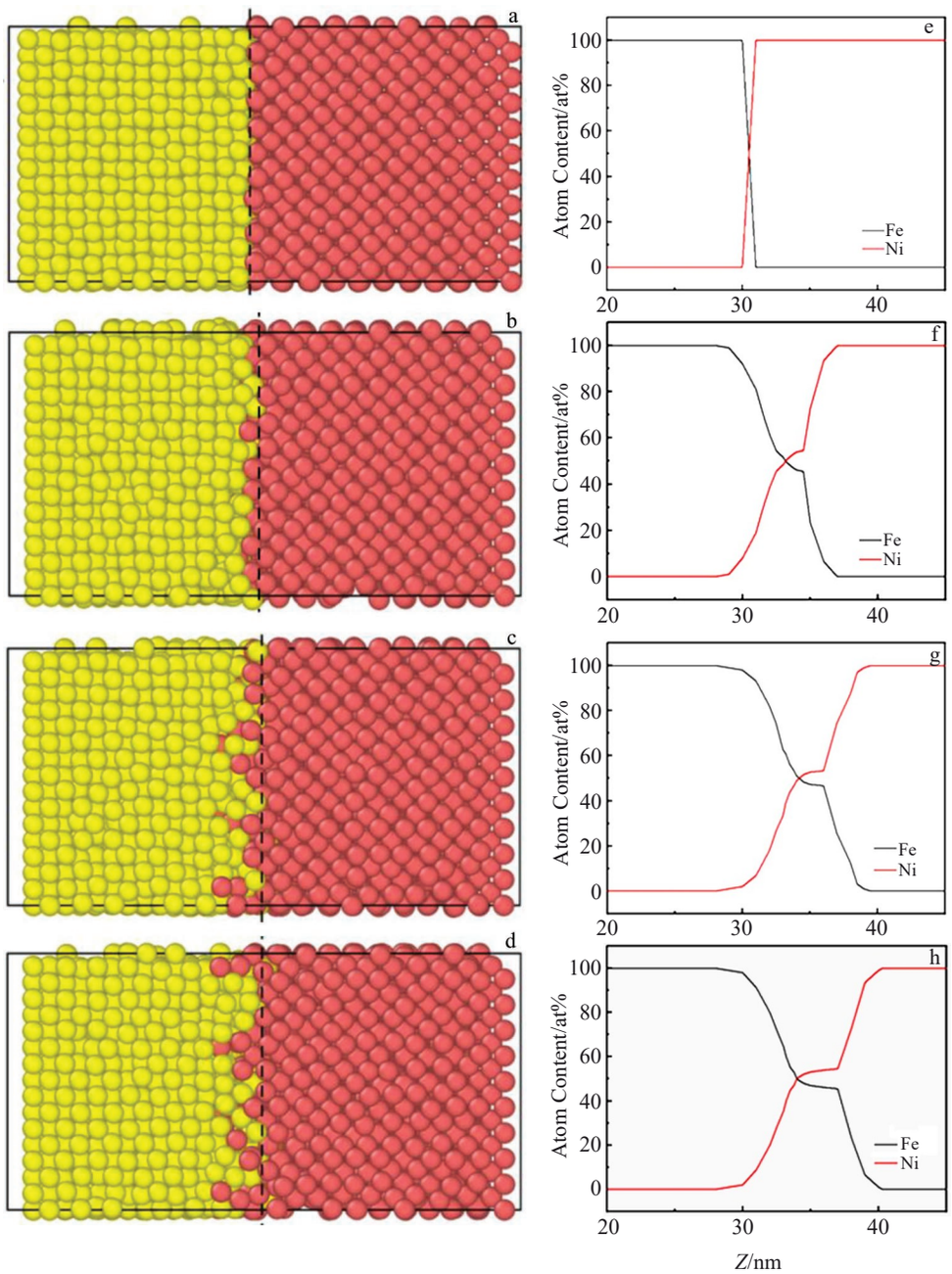


Fig.3 Atomic distribution models (a~d) and atom contents along *Z* axis (e~h) of Fe-Ni binary system at 1123 K and different diffusion time: (a, e) 0 ps, (b, f) 200 ps, (c, g) 400 ps, and (d, h) 600 ps

position of atom *i* in the initial state; *N* is the number of atoms.

Fig. 6 shows MSD values of Fe and Cu atoms in the diffusion process. The direct proportion indicates that the Fe and Cu atoms have obvious diffusion behavior at these three brazing temperatures. With increasing the ambient temperature of diffusion simulation, MSD value is increased. MSD values of Fe and Cu atoms at different time are shown in Table 3. It can be seen that MSD values of Fe atoms are generally higher than those of Cu atoms at 1323 K. Therefore, it can be concluded that the diffusion ability of Fe atoms is better than that of Cu atoms in the Fe-Cu binary diffusion system.

Additionally, MSD values of Fe and Ni atoms are increased linearly with the diffusion proceeding at different brazing temperatures, as shown in Fig. 7. However, the slope of MSD curves of Fe and Ni atoms has certain changes at 1123 and 1223 K and 240 ps. After that, they are stable at 1123 and 1223 K when the diffusion time is 240~600 ps, indicating the stable diffusion of Fe and Ni atoms. Besides, MSD values of Fe and Ni atoms are also increased with increasing the temperature at the same diffusion time, suggesting that the average diffusion displacement of the particles is increased and the diffusion movement of the particles becomes more active with increasing the temperature. MSD values of Fe and Ni atoms at different time are shown in Table 4. It can be seen that MSD values of Fe atoms are generally higher than those

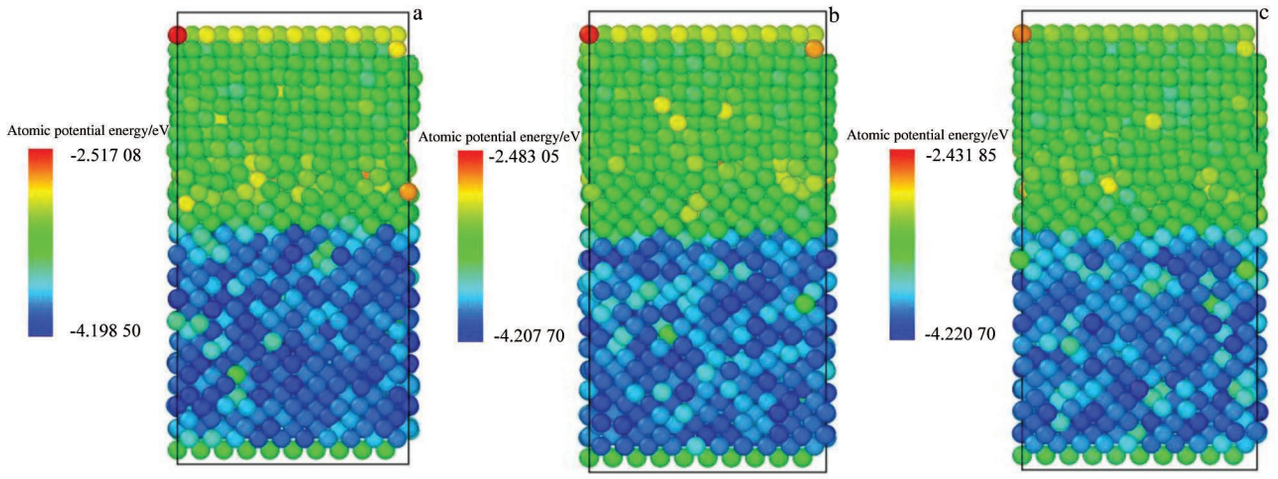


Fig.4 Fe-Cu atomic potential energies at 1123 K (a), 1223 K (b), and 1323 K (c) after 600 ps

**Table 1 Average atomic potential energies of Fe and Cu atoms at different temperatures after 600 ps (eV)**

| Temperature/K | 1123 | 1223 | 1323 |
|---|---|---|---|
| Fe | −3.8522 | −3.8717 | −3.8829 |
| Cu | −3.2708 | −3.2881 | −3.2975 |

of Ni atoms at the same temperature, which means that the diffusion ability of Fe atoms is better than that of Ni atoms in the Fe-Ni binary diffusion system. Moreover, these results also agree with the atomic diffusion models.

Diffusion coefficient ($D$) is usually used to indicate the atom diffusion rate quantificationally, and it can be obtained

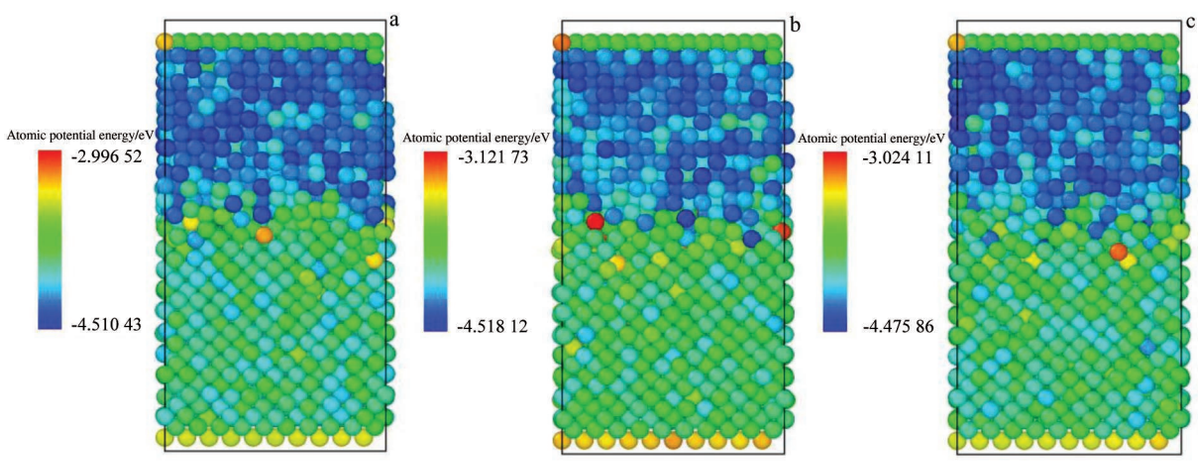


Fig.5 Fe-Ni atomic potential energies at 1123 K (a), 1223 K (b), and 1323 K (c) after 600 ps

through MSD, as expressed by the Einstein diffusion equation[17]:

$$D = \lim_{t\to\infty} \frac{1}{2N_0 t} \left| r_i(t) - r_i(0) \right|^2 \quad (2)$$

where $D$ is the diffusion coefficient of the particle; $N_0$ is the dimension of simulation system and it is set as 3 in this research. Therefore, the relationship between MSD and diffusion coefficient can be obtained, as expressed by Eq.(3):

$$D = \frac{1}{2N_0} \lim_{t\to\infty} \frac{\mathrm{MSD}}{t} = \lim_{t\to\infty} \frac{\mathrm{MSD}}{6t} \quad (3)$$

The diffusion coefficients of Fe and Cu atoms at different temperatures can be obtained, as shown in Fig. 8a. It can be seen that the diffusion coefficients of both atoms are increased significantly with increasing the temperature, i. e., the higher the temperature, the greater the atom diffusion rate. This is because the high temperature leads to the unstable atoms and low barrier for diffusion initiation. As for the Fe atom, the diffusion coefficients at 1123, 1223, and 1323 K are $2.12\times10^{-9}$, $2.97\times10^{-9}$, and $3.55\times10^{-9}$ $m^2/s$, respectively; as for the Cu atom, the diffusion coefficients at 1123, 1223, and 1323 K are $2.062\times10^{-9}$, $2.753\times10^{-9}$, and $3.457\times10^{-9}$ $m^2/s$, respectively. Therefore, it can be concluded that the diffusion ability of Fe atoms is better than that of Cu atoms at the same temperature. The radius of Fe atom is smaller than that of Cu atom, indicating the easier diffusion initiation of Fe atom in the binary system.

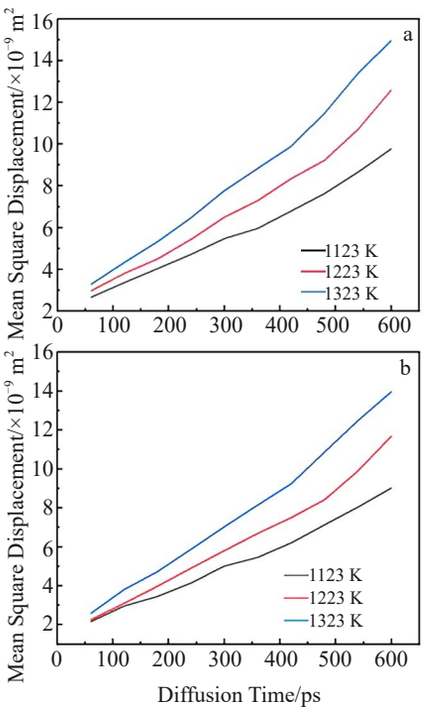


Fig.6 MSD values of Fe (a) and Cu (b) atoms at different temperatures in diffusion process

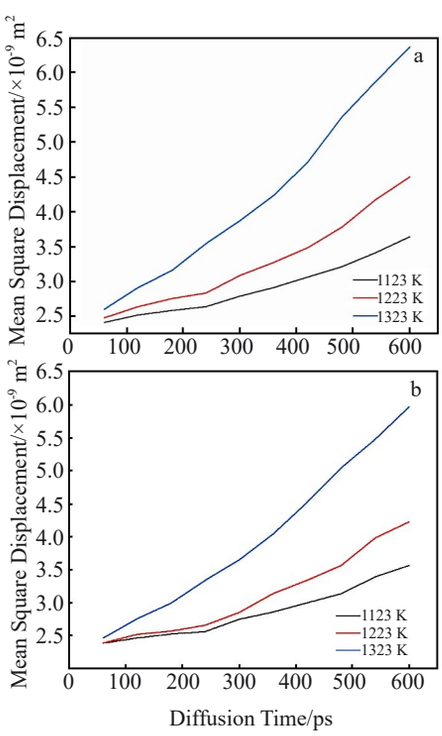


Fig.7 MSD values of Fe (a) and Ni (b) atoms at different temperatures in diffusion process

**Table 2 Average atomic potential energies of Fe and Ni atoms at different temperatures after 600 ps (eV)**

| Temperature/K | 1123 | 1223 | 1323 |
|---|---|---|---|
| Fe | −3.8532 | −3.8731 | −3.8847 |
| Ni | −4.1838 | −4.1985 | −4.2066 |

**Table 3 MSD values of Fe and Cu atoms during diffusion process at 1323 K (×10⁻⁹ m²)**

| Time/ps | 60 | 180 | 300 | 480 | 600 |
|---|---|---|---|---|---|
| Fe | 3.277 39 | 5.332 43 | 7.777 02 | 11.488 30 | 14.978 50 |
| Cu | 2.565 87 | 4.734 97 | 7.038 57 | 10.875 40 | 13.749 50 |

**Table 4 MSD values of Fe and Ni atoms during diffusion process at 1323 K (×10⁻⁹ m²)**

| Time/ps | 60 | 180 | 300 | 480 | 600 |
|---|---|---|---|---|---|
| Fe | 2.608 47 | 3.165 28 | 3.871 63 | 5.369 35 | 6.362 80 |
| Ni | 2.464 61 | 4.734 97 | 3.648 64 | 5.045 19 | 5.971 90 |

However, MSD is not always proportional to the diffusion time at 1123 and 1223 K in the Fe-Ni binary diffusion system. Therefore, the diffusion coefficients are solved at 1123 and 1223 K within 240~600 ps, as shown in Fig. 8b. It can be seen that with increasing the temperature, the diffusion coefficients of two atoms are also increased significantly, i. e., the higher the temperature, the greater the atom diffusion rate. For the Fe atom, the diffusion coefficients at 1123, 1223, and 1323 K are $4.73\times10^{-10}$, $7.63\times10^{-10}$, and $11.70\times10^{-10}$ $m^2/s$, respectively; as for the Ni atom, the diffusion coefficients at the temperature of 1123, 1223, and 1323 K are $4.53\times10^{-10}$, $7.33\times10^{-10}$, and $10.92\times10^{-10}$ $m^2/s$, respectively. Therefore, it can be seen that the diffusion ability of Fe atoms is better than that of Ni atoms at the same temperature.

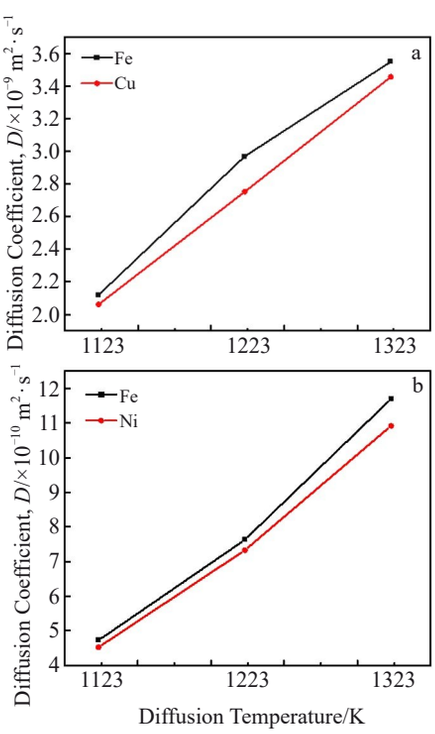


Fig.8 Relationships between diffusion coefficient $D$ and diffusion temperature in Fe-Cu (a) and Fe-Ni (b) binary systems

## 3 Conclusions

1) The obvious mutual diffusion phenomenon can be observed during the molecular dynamics simulation of the diffusion in Fe-Cu and Fe-Ni binary systems. The thickness of diffusion layer is increased with the diffusion proceeding. There is only mutual diffusion process in the Fe-Cu binary system, while in the Fe-Ni binary system, the mutual diffusion process and the generation of Fe-Ni compound mesophase occur.

2) The higher the diffusion temperature, the greater the mean square displacement (MSD), the larger the diffusion coefficient, and the better the diffusion ability. In the Fe-Cu binary system, MSD and diffusion coefficient of Fe atom are greater than those of Cu atom, indicating that the diffusion ability of Fe atom is better than that of Cu atom. Similarly, in the Fe-Ni binary system, MSD and diffusion coefficient of Fe atom are greater than those of Ni atom, indicating that the diffusion ability of Fe atom is also better than that of Ni atom.

## References


1 Nishimoto K, Mori H. *Science and Technology of Advanced Materials*[J], 2004, 5(1-2): 231

2 Zhang H, Wang D, Xue P *et al. Materials & Design*[J], 2016, 110: 802

3 Zhao L, Tian Z L, Peng Y. *ISIJ International*[J], 2007, 47(12): 1772

4 Baba H, Katada Y. *Corrosion Science*[J], 2006, 48(9): 2510

5 Bayoumi F M, Ghanem W A. *Materials Letters*[J], 2005, 59(26): 3311

6 Lin Z, Tian Z L, Yun P *et al*. *Journal of Iron and Steel Research International*[J], 2007, 14(5): 259

7 Li J G, Li H, Peng W *et al*. *Materials Characterization*[J], 2019, 149: 206

8 Wang Q, Zhang S Y, Lin T S *et al*. *Progress in Natural Science: Materials International*[J], 2021, 31(1): 129

9 Del Rio E, Ampedro J M, Dogo H *et al*. *Journal of Nuclear Materials*[J], 2011, 408(1): 18

10 Wu H Y, Dong Y P, Li Y L *et al*. *Materials Science and Engineering A*[J], 2021, 803: 140 711

11 Li Y L, Liu Y R, Yang J. *Optics & Laser Technology*[J], 2020, 122: 105 875

12 Long Fei, Zhang Guanxin, He Peng *et al*. *Rare Metal Materials and Engineering*[J], 2020, 49(2): 385

13 Yang Yao, Liang Yuxin, Li Shaorong *et al*. *Rare Metal Materials and Engineering*[J], 2020, 49(8): 2636

14 Martyna G J, Klein M L, Tuckerman M. *Journal of Chemical Physics*[J], 1992, 97(4): 2635

15 Nosé S. *Molecular Physics*[J], 2002, 100(1): 191

16 Gervilla V, Zarshenas M, Sangiovanni D G *et al*. *The Journal of Physical Chemistry Letters*[J], 2020, 11(21): 8930

17 Bonny G, Castin N, Terentyev D. *Modelling and Simulation in Materials Science and Engineering*[J], 2013, 21(8): 85 004

18 Chang Jinbao, Zhao Yingli, Ji Shuang *et al*. *Heat Treatment of Metals*[J], 2020, 45(2): 120 (in Chinese)

19 Ouyang Yifang, Wu Jizheng, Zheng Minghui *et al*. *Computational Materials Science*[J], 2018, 147: 7


# 银铜镍钎料钎焊高氮不锈钢的分子动力学模拟


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**摘　要**：利用Lammps软件对AgCuNi钎料真空钎焊高氮不锈钢的相关二元体系（Fe-Cu和Fe-Ni）元素扩散过程进行分子动力学模拟。结果表明，Fe-Cu和Fe-Ni二元体系相互扩散现象明显，扩散层厚度随着扩散时间增加而增加。在Fe-Cu体系的扩散过程中只有原子相互扩散，但Fe-Ni体系的扩散过程中既有原子扩散又有中间相生成。在Fe-Cu二元体系中，Fe原子的均方位移和扩散系数均大于Cu原子，因此Fe原子的扩散能力大于Cu原子。在Fe-Ni二元体系中，Fe原子的均方位移和扩散系数都大于Ni原子，因此Fe原子的扩散能力也大于Ni原子。随着扩散温度升高，原子均方位移和扩散系数增大，扩散能力越强。



**关键词**：高氮不锈钢；钎焊；AgCuNi钎料；分子动力学模拟；均方位移



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