Evaluation of Microstructure and Glass Transition Temperature of Al-Cu-Cr-Fe-Ni High-Entropy Alloy by Molecular Dynamics Simulation

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Abstract - In this study, the microstructure and glass transition temperature (Tg) of five elements (Al-Cu-Cr-Fe-Ni) high-entropy alloy was evaluated under different Al content by molecular dynamics (MD) simulations. The <NVT> ensemble and COMPASS potential were used. Firstly, the Al-Cu-Cr-Fe-Ni high-entropy alloys were melted at high temperature and were cooled with a high quenching rate furtherly.The radial distribution function (RDF),Wendt-Abraham parameter and X-ray diffractometer (XRD) were used to analyze the change on the microstructure and glass transition temperature (Tg) of Al-Cu-Cr-Fe-Ni high-entropy alloys. Simulation results show that the micro-structure of different aluminum content of AlxCuCrFeNi alloy after fast quenching are all amorphous state. When the aluminum content decreased, the amorphous state are more obviou and the glass transition temperature decreases.

Keywords – High entropy alloy, Mcrostructure, Glass transition temperature, Molecular dynamics.

I. INTRODUCTION

High entropy alloys are multi-component alloys which were composed of n major alloy elements, where n specifically indicates >= 5 [1]. For thousands of years the development of practical alloy systems has been based typically on the use of one principal element as the matrix. Rrecently, Yeh et al. [2] and Inoue [3] have explored an entirely new class of alloys coined as high-entropy alloys which are defined as alloys that have at least five principal elements with concentrations between 5 and 35 at.%. These alloys are found to consist of multiprincipal element solid solution phases typically, as opposed to the anticipated formation of a multitude of complicated and brittle intermetallic compounds [4].

It was found that nanoprecipitation can occur [5] at lower temperatures, when the contribution of the mixing entropy is reduced. These structural characteristics, i.e. multiprincipal element solid solution phases with nanoprecipitation, are found to provide high-entropy alloys with a combination of promising properties, such as high hardness, large work hardening capacity, superior resistance to temper softening, wear, oxidation and corrosion [2–5]. It has been found, however, that the large mixing entropy may not be sufficient to prevent the segregation of some various chemically incompatible elements [2].

Elemental composition is a critical factor in alloy design which will definitely affect the alloying behaviors and properties of high-entropy alloys. Many high-entropy alloy systems have been reported recently, and Al was investigated in most of the cases [6, 7]. The addition of Al was identified to having considerable effects on the structures and properties of high-entropy alloys due to its large atomic radius [8].

In previous studies, high-entropy alloys have demonstrated simple crystal structures, ease of nanoprecipitation, a promising degree of hardness, resistance to temper softening, and wear [9]. Among these alloys, AlCoCrFeNi has shown particular promise for applications in structural and tool industries, making it an ideal candidate material for nanomolds.

Molecular dynamics (MD) simulation can provide the opportunity to understand the nature of the microstructure and the glass formation at an atomic level, which is difficult to access in real experiments or hard to obtain with reasonable precision. There are large number of studies in the MD simulations of glass transitions of metals and binary alloys [10-13].

However, the studies on five elements high entropy alloys are still rare in the international literatures. As a result, we investigate the molecular dynamics to explore the influence of different aluminum contents in high-entropy alloys at high cooling rates on the microstructure of high entropy alloy and the glass transition temperature (Tg) in this paper.

II. PHYSICAL MODEL

A standard MD simulation was performed using the Forcite module in Materials Studio® (Accelrys Software Inc.) with a COMPASS potential function [14] to construct atomistic models of atoms in high entropy alloys.

In the preparation of high entropy alloy substrates, periodic boundary conditions were applied to the models along both the Z axis and transverse directions. The substrates were constructed with from 400 to 500 atoms including Al, Co, Cr, Fe and Ni, and the aluminum content in different composition as shown in Table 1. The size of the substrate was 1.8 nm. The initial physical model of the high-entropy alloy is shown in Fig. 1.

III. SIMULATION OF MOLECULAR DYNAMICS

A. Preparation of AlxCoCrFeNi High-Entropy Alloys

The MD simulation procedure was proceeded according to Haile [15]. Initially, the atoms of the specimen were random positioned at face-centered-cubic (FCC) crystal structure sites, and then relaxed to a position of equilibrium at 2200 K followed by quenching at 0.05K/fs to 300 K. In

this study, the preparation of high-entropy alloys comprised two stages: initial relaxation and quick quenching. In the first stage (melt stage), all atoms were positioned according to an assumed FCC crystal structure and their velocities were randomized to fit a Maxwell-Boltzmann distribution at 2200 K. For the simulation results presented below, the relaxation phase was completed within 20 000 time steps by controlling the bulk temperature and external pressure at a setting temperature and at 0 GPa, respectively, using an <NPT> ensemble [15]. In the second stage, quick quenching was simulated by reducing the environmental temperature by 0.05 K in each time step, when the environmental temperature reached 300 K. The quick quenching phase was completed within 20000 time steps by controlling the external stress at a setting temperature of 300 K using an <NPT> ensemble.

Table 1. The different composition of high-entropy alloys

Composition	Al	Cu	Cr	Fe	Ni	Total number of atoms
$Al_0CuCrFeNi$	0	100	100	100	100	400
$Al_{0.3}CuCrFeNi$	30	100	100	100	100	430
$Al_{0.5}CuCrFeNi$	50	100	100	100	100	450
AlCuCrFeNi	100	100	100	100	100	500

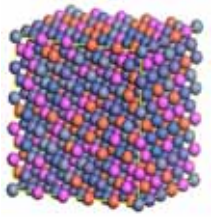


Fig 1. Physical model of high entropy alloys

B. Radial Distribution Function

The radial distribution function (RDF), $g(r)$, is an important structural characteristic, indicating the probability of finding a particle within a distance r from another particle, and is a useful tool for describing the structure of a system. The expression of the radial distribution function is as follows [15]:

$$g(r)=\frac{\sum_{k=1}^{M} N_k(r,\Delta r)}{M\left(\frac{1}{2}N\right)\rho V(r,\Delta r)} \quad (1)$$

where, $V(r, \Delta r)$ is the spherical shell volume of radius r with a thickness of Δr; $N(r,\Delta r)$ is the number of particles within the volume of the shell $V(r,\Delta r)$; M is the number of shells in the system; N is the total number of particles; and ρ is the density of the system.

In a solid, the radial distribution function has an infinite number of sharp peaks whose separation and height are characteristic of a lattice structure. At short distances (less than the atomic diameter), $g(r)$ is zero, due to strong repulsive forces. At long distances, $g(r)$ approaches unity indicating a lack of long-range order.

Radial distribution can be measured experimentally by X-ray diffraction. The regular arrangement of the atoms in a crystal provides characteristic X-ray diffraction patterns with bright, sharp spots. The X-ray diffraction pattern is analyzed to estimate an experimental distribution function for comparison with the results obtained in the simulation.

C. Wendt-Abraham Parameter

The parameter used to dene the glass transition temperature is the Wendt-Abraham parameter [16] defined by (2), The R^{WA} being the value of g(r) at the rst minimum (maximum).

$$R^{WA}\left(\frac{g_{\min}}{g_{\max}}\right)=\frac{g_{\min}}{g_{\max}} \quad (5)$$

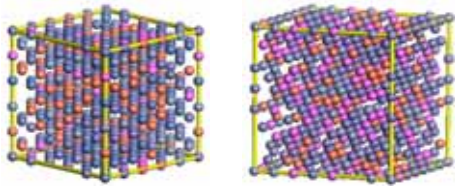
This parameter stresses the local position of g(r), permits a direct comparison between structural and will give the right sequence of the glass transition temperature.

V. RESULTS AND DISCUSSIONS

A. The Effect of Different Lattice Structure

To understand the influence of the initial lattice structure of high entropy alloys on simulation results, the body-centred-cubic (BCC) lattice and the face-centred-cubic (FCC) lattice was used and shown in Fig. 2. The system temperature raised from 300 K to 3200K, and then cooled to 2300K, then performed RDF analysis after the system relaxation. The RDF analysis results were shown in Fig. 3.

From Fig. 3, the RDF distribution is insignificant from the different lattice structure constructed AlCuCrFeNi multi-alloy in the liquid, which can be proved using the BCC or FCC lattice structure to create a physical model will not affect the overall results of the study. Therefore, FCC lattice structure was used to create the physical model in this paper.



(a) (b)

Fig. 2 Physical model of high entropy alloys with (a) FCC structure (b) BCC structure

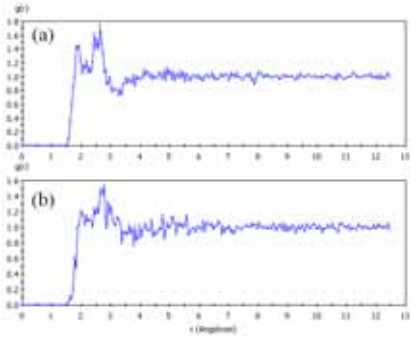


Fig. 3 The RDF patterns of different lattice model at 2300K (liquid phase) (a) FCC (b) BCC

B. The Influence of Al Content on the Microstructure of High Entropy Alloys

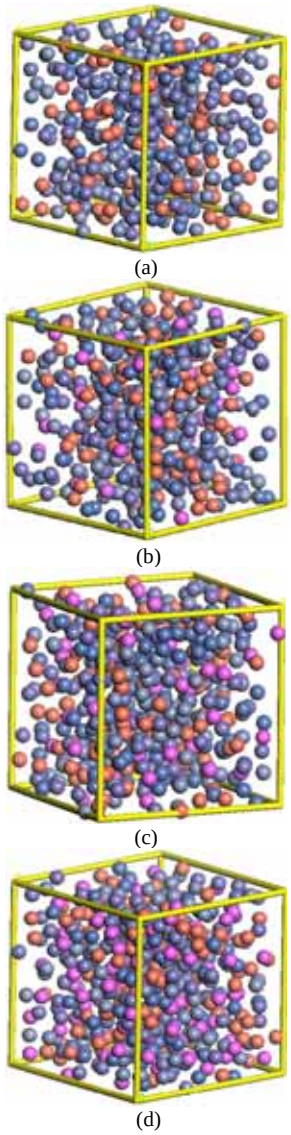
After rapid cooling to 300 K from 2300 K, the appearance structure of high entropy alloys with different aluminium content are shown in Fig. 4. And analysis results

of RDF and XRD were shown in Fig. 5 and Fig. 6, respectively.

From Fig. 4, the microstructure of high entropy alloys are arranged at random. Therefore, only by the appearance can not understand the effect of increase of aluminium content on the microstructure.

From Fig. 5, with the content of Al increases, the intesity in RDF distribution of high entropy alloys becomes more obvious, here that with the aluminum content increases and the regularity of microstructure of the high entropy alloys showed slightly increased also.

From Fig. 6, with the content of Al increases, the major peaks at XRD wrere splitted from three peaks to five peaks. The phenomenone indicates that the higher composition of aluminium was introduced, the XRD pattern presented characteristic peak of five elements composition were more obvious.



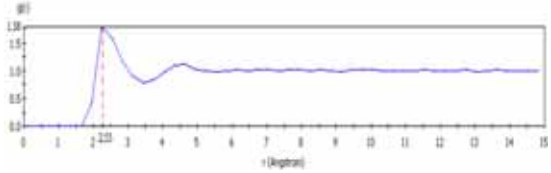
(a)

(b)

(c)

(d)

Fig. 4 The structure of high entropy alloys with different aluminum content at room temperature 300K (a) $Al_0CuCrFeNi$ (b)$Al_{0.3}CuCrFeNi$ (c)$Al_{0.5}CuCrFeNi$ (d) $Al_{1.0}CuCrFeNi$



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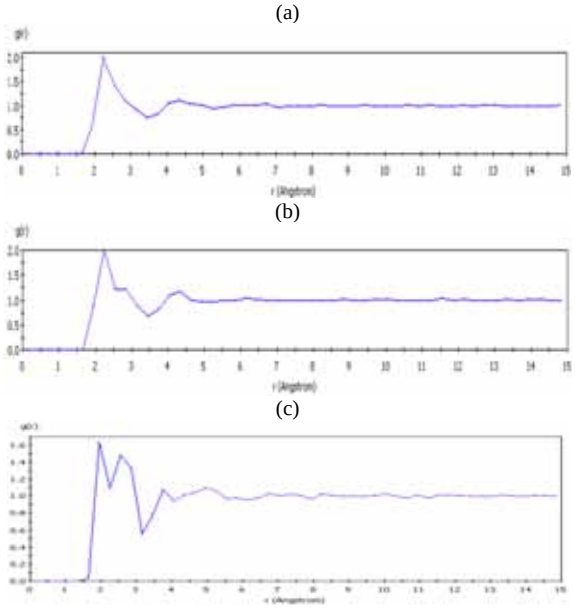

(b)

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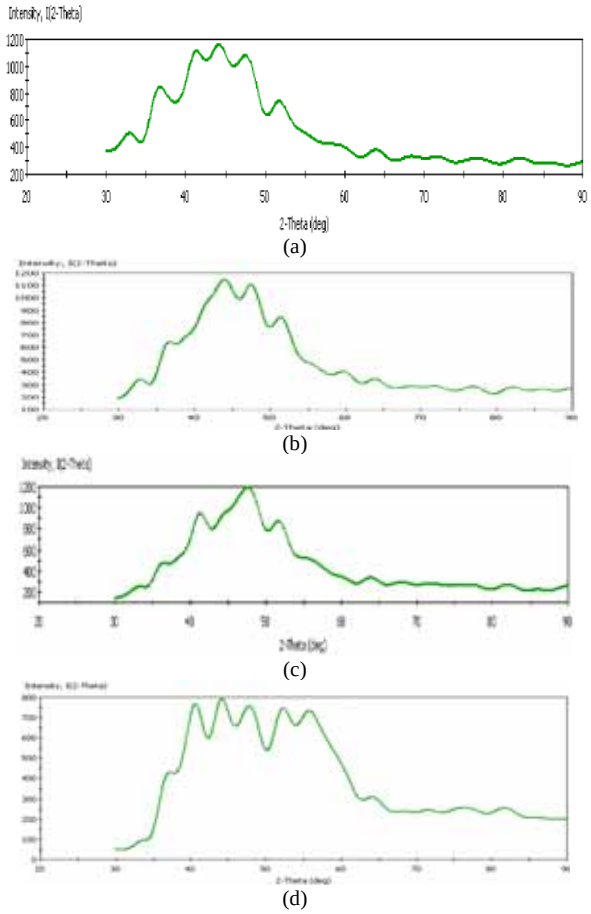
(d)

Fig. 5 The RDF patterns of high entropy alloys with different aluminium content at room temperature 300K (a) $Al_0CuCrFeNi$ (b)$Al_{0.3}CuCrFeNi$ (c)$Al_{0.5}CuCrFeNi$ (d) $Al_{1.0}CuCrFeNi$



(a)

(b)

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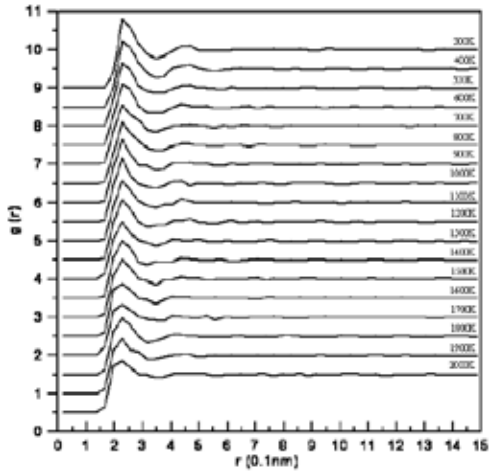
Fig. 6 The XRD patterns of high entropy alloys with different aluminium content at room temperature 300K (a) $Al_0CuCrFeNi$ (b)$Al_{0.3}CuCrFeNi$ (c)$Al_{0.5}CuCrFeNi$ (d) $Al_{1.0}CuCrFeNi$

C. The Influence of Al Content on the Tg of High Entropy Alloys

The high entropy alloys with different aluminium content were rapidly cooled from 2200K to 300K gradiently and the RDF analysis was carried out also with a set of 100K. The combined RDF patterns were shown in Fig. 7.

Fig 8 displays the relation between R^{WA} and temperature and the Wendt-Abraham transition temperature Tg can be determined. The results are shown in Table2.

The glass transition temperature gets higher with the reducing of Al contents. When the Al content is high, the distortion between five element atoms is large, and glass is formed earlier than lower Al content spieces.



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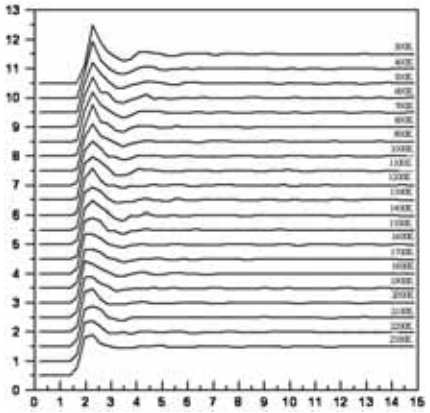

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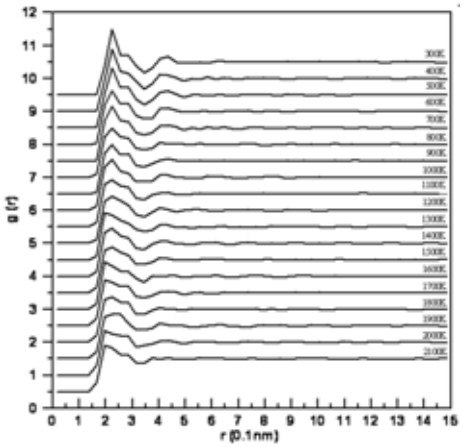

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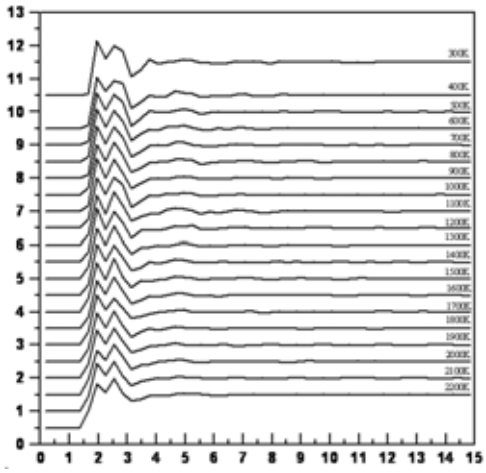
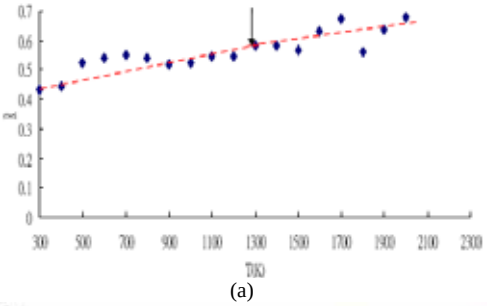


(d)

Fig. 7 The RDF patterns of high entropy alloys with different aluminium content from 2200K to 300K (a) $Al_0CuCrFeNi$ (b)$Al_{0.3}CuCrFeNi$ (c)$Al_{0.5}CuCrFeNi$ (d) $Al_{1.0}CuCrFeNi$



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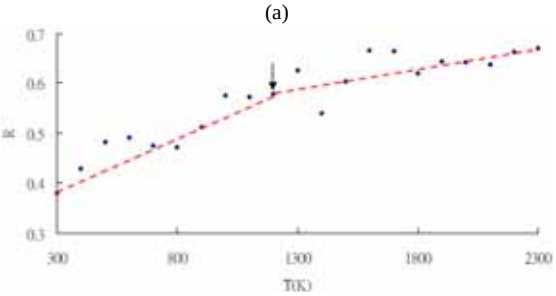

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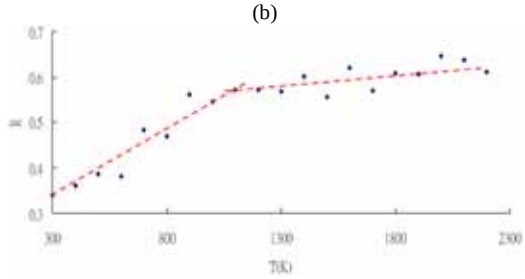

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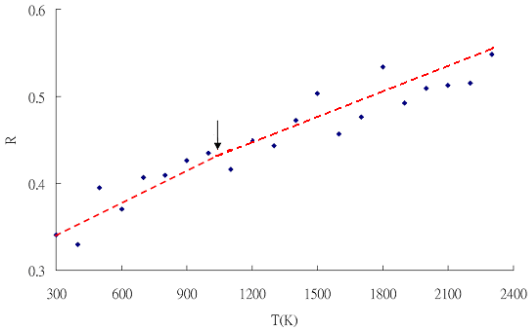

(d)

Fig. 8 The RWA （R=gmin /gmax） with different aluminium content from 2200K to 300K (a) $Al_0CuCrFeNi$ (b)$Al_{0.3}CuCrFeNi$ (c)$Al_{0.5}CuCrFeNi$ (d) $Al_{1.0}CuCrFeNi$

Table 2 The Tg at different Al contens in high-entropy alloys

Aluminium content (%)	Tg (K)
0	1300
30	1200
50	1100
100	1050

VI. CONCLUSIONS

In this study, the microstructure and glass transition temperature (Tg) of five elements (Al-Cu-Cr-Fe-Ni) high-entropy alloy with different Al content was successfully evaluated by MD simulations. The RDF, Wendt-Abraham parameter and X-ray diffractometer (XRD) were used to analyze the variation on the microstructure and Tg of Al_x-Cu-Cr-Fe-Ni high-entropy alloys.

The simulation results show that the microstructures of the different aluminum content of AlxCuCrFeNi high-entropy alloy after fast quenching are amorphous state.

According to the XRD analysis, the greater content of aluminum will enhence the characteristic peak of five elements composition (the major peaks were splitted from three to five).

In the final, the content of Al also shows the influence in Tg. As the aluminum content increases, the glass transition temperature of AlxCuCrFeNi high-entropy alloy decreases.

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