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The structure of the Zr-Cu-Al melts in the glass forming range of concentrations

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The short-range order structure of the $Zr_{47.5}Cu_{47.5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts was investigated by the method of molecular dynamics simulations. Based on the obtained results, partial pair correlation functions and distributions of partial coordination numbers have been calculated. The main structure parameters determined from these functions were compared with similar parameters for amorphous alloys. According to the results of research, the presence of both homocoordinated and heterocoordinated clusters in the melts, which form a cluster solution, was established. The assumption of the presence of heterocoordinated icosahedral clusters is made.

Keywords: metal alloys, atomic structure, coordination numbers, cluster structure.

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Introduction

Zr-Cu alloys are known to be able to form bulk amorphous alloys by melt crystallization. According to the literature, the compositional interval of formation of amorphous alloys in such systems corresponds to the range of concentrations from 25 at% to 70 at% of copper [1, 2]. In particular, the study of the structure and properties of amorphous alloys of the Zr-Cu system was performed for atomic compositions $Zr_{35}Cu_{65}$, $Zr_{36}Cu_{64}$, $Zr_{40}Cu_{60}$, $Cu_{46}Zr_{54}$, $Cu_{50}Zr_{50}$, $Zr_{55}Cu_{45}$ [3-9]. Many studies have reported the presence of a set of polyhedra in the amorphous phase, many of which are icosahedrons characterized by fifth-order symmetry. According to these works, the high ability to amorphization is due to the presence of atomic clusters formed by icosahedrons.

The structure and properties of amorphous and liquid alloys of the Zr-Cu system were also studied by computer simulation methods. In particular, the authors of [10] concluded that both in the amorphous and in the liquid state, the increase in copper content causes an increase in the degree of icosahedral structure of the short-range order structure. The results of research on the relationship between the glass forming ability, the degree of

icosahedral structure and the amount of free volume for Cu-Zr alloys are presented in [11]. It is shown that these features of the structure cause the slowing down of diffusion processes in the amorphous state. Interesting results were obtained in [12], where the structure of Zr-Cu and Zr-Ni alloys in the liquid state was studied. It was shown that in these melts the formation of different types of short-range order is especially due to the difference in the electronegativities of the components and their electronic structure.

It has been shown that the addition of aluminum to alloys of the Zr-Cu system improves the glass forming ability, which allows to obtain bulk amorphous alloys up to 20 mm in diameter [13, 14]. It was found that the glass forming ability of the $Zr_{65}Cu_{27.5}Al_{7.5}$ alloy compared to the $Zr_{65}Cu_{35}$ is higher and it is shown that the increase in this ability in addition of aluminum is due to reduced diffusion coefficients of copper and aluminum in the melt and low rate of $Zr_2(Cu, Al)$ compound nucleation [15]. This paper also shows that the increase in the glass forming ability is not due to the complexity of crystallization due to the presence of aluminum atoms at the interface between the liquid and amorphous phases. Another study indicates that the high glass forming ability of the Zr-(Cu, Ag)-Al

system alloys is due to the denser local atomic packaging and the smaller difference in Gibbs free energies between the amorphous and crystalline phases [13].

Alloys of the Zr-Cu-Al system have been repeatedly studied for atomic structure and its effect on the glass forming ability, as well as the properties of alloys in the amorphous state [16-19]. In particular, by means of the method of X-ray diffraction studies was found that the addition of 10% aluminum to the $Zr_{66.7}Cu_{33.3}$ alloy does not affect the shape of the pair correlation functions. An increase in the aluminum content leads to a decrease in the number of Zr-Zr and Zr-Cu atomic pairs and an increase in the Zr-Al and Cu-Al pairs [16]. The study of the structure of the $Zr_{48}Cu_{45}Al_7$ amorphous alloy before and after annealing indicates the special role of aluminium atoms in the alloy, which binds copper and zirconium atoms becoming the centers of clusters [17]. X-ray diffraction studies also showed that the addition of aluminum to Zr-Cu alloys leads to the modification of the atomic structure in such a way that as a result of the predominant interaction between aluminum and zirconium, a significant number of Zr-Al atomic pairs are formed, the number of which is much greater than the distribution of atoms in an ideal solution [18]. The study of the atomic structure of amorphous Zr-Cu-Al alloys by EXAFS spectroscopy indicates the existence of icosahedral clusters as the main components that form these alloys [19].

The icosahedral structure of amorphous and liquid alloys can be studied in more detail by computer simulation methods. Currently, there are many works in which the structure of liquid and amorphous alloys by the method of molecular dynamics has been studied. Many of them confirmed the experimental results on the existence of icosahedral clusters in amorphous alloys of the Zr-Cu-Al system. In addition, the presence of interpenetrating chains based on icosahedral clusters is indicated [20-22]. It is shown that the addition of aluminum to the base alloy Zr-Cu is particularly effective, which is manifested not only in a significant increase in the number of icosahedral clusters, but also in their improved symmetry, as well as their expanded spatial connectivity and the appearance of intermediate order in the structure [20]. There are also studies that shed light on the amorphization process itself, and the molecular dynamics method explains the better ability to amorphize the Zr-Cu-Al ternary alloy compared to the Zr-Cu binary base alloy.

Thus, as we can see from previous studies, amorphous alloys of the Zr-Cu-Al system, as well as similar melts near the melting temperature are characterized by a predominant interaction of atoms of different types, which leads to the formation of icosahedral clusters. However, due to the significant intensity of interatomic bonds between different kind of atoms, it is possible to assume the presence of residues of icosahedral clusters and inhomogeneous short-range order structure in the liquid state at temperatures much higher than the melting point. In view of this, the structure of Zr-Cu-Al melts at temperatures above the melting point of the most refractory component of the alloy, zirconium, was investigated in this work by the method of molecular dynamics. Simulations were performed in the temperature range 2000K - 2500K.

I. Methods of molecular dynamic simulations

The structure of Zr-Cu-Al melts was studied by the molecular dynamics simulation method using the LAMMPS package [23]. The simulations were performed at a constant temperature and volume in the NVT ensemble. The initial arrangement of atoms in a model cell of cubic shape was chosen randomly, taking into account the experimental density of melts and radii of atoms. The interaction between atoms has been described using the EAM potential proposed for the Al-Cu-Fe-Mg-Si system [24]. According to this method, the total energy of the atomic system can be calculated by the equation:

$$E = \sum_{i=1}^N \{F(\bar{\rho}_i) + \sum_{i \neq j}^N S_{ij} \varphi_{ij}(r_{ij})\} \quad (1)$$

The initial velocities of atoms were set according to the Maxwell-Boltzmann distribution for each of the temperatures studied. The step of integrating the equations of motion was 2 fs.

The resulting atomic configurations were used to calculate partial pair correlation functions, mean coordination numbers and their distributions. Partial pair correlation functions were calculated from the coordinates of atoms and the distance between them in the model cell:

$$g(R_n) = \frac{V(L)h_n}{2\pi N(L)^2 R_n^2 \Delta R} \quad (2)$$

$V(L)$ is the volume of the spherical layer in which the atoms are considered, h_n – the number of pairs of atoms in the range of distances for which $(n-1)\Delta R \leq R_{ij} \leq n \Delta R$ at a distance

$$R_n = \left(\frac{n-1}{2}\right) \Delta R \quad (3)$$

The Open Visualization Tool (OVITO) was used for visual analysis of atomic configurations [25].

II. Results and discussion

The study of the structure of melts of the Zr-Cu-Al system was carried out on the basis of analysis of atomic configurations obtained by molecular dynamics at different temperatures (Figs. 1 and 2) as well as quantitative characteristics calculated using these configurations. Visual analysis of atomic configurations shows, at first glance, the predominant interaction of Zr-Zr and Cu-Cu atoms, while the analysis of thermodynamic functions and phase diagrams shows a tendency to form an atomic solution or pairs of Zr-Cu atoms. On the other hand, the addition of aluminum should be accompanied by its predominant interaction with zirconium. Indeed, according to the literature data, the enthalpy of mixing for the Zr-Al system reaches -44 kJ / mol, while in the case of Zr-Cu and Cu-Al alloys it is much lower, and is -23 kJ / mol and -1 kJ / mol respectively [26]. It should also be noted that according

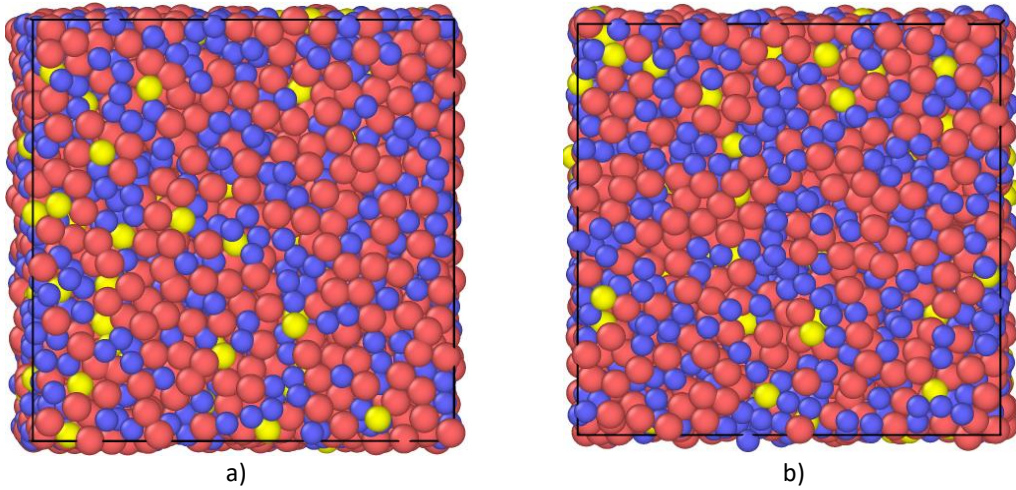


Fig. 1. Distribution of atoms in the model cell of the $Zr_{47.5}Cu_{47.5}Al_5$ melt at temperatures $T = 2000K$ (a) and $T = 2500K$ (b)

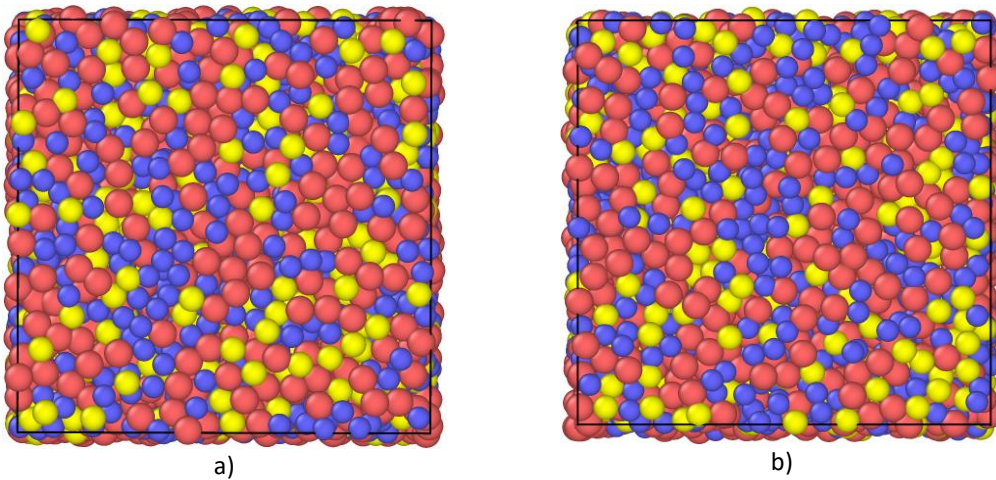


Fig. 2. Distribution of atoms in the model cell of the $Zr_{40}Cu_{40}Al_{20}$ melt at temperatures $T = 2000K$ (a) and $T = 2500K$ (b)

to the already established laws of temperature dependence of the structure of melts, in the liquid state at the temperatures at which the structure was modeled, more expected to form an atomic solution of alloy components close to ideal, which in our case is not observed.

Given the above mentioned facts, it is necessary for a detailed study of atomic structure of alloys, which was carried out by us on the basis of quantitative analysis of pair correlation functions and structural parameters determined from them. Partial pair correlation functions were obtained on the basis of information about the distribution of atoms in the model cell using equation (2). Figure 3a shows the indicated functions for pairs of atoms of the same type for $Zr_{47.5}Cu_{47.5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts, as well as for atoms of different types (Fig. 3, b).

A characteristic feature of partial pair correlation functions for atoms of the same type is that the most probable interatomic distances in alloys practically coincide with similar distances for pure components. This feature is clear for pairs of Zr-Zr and Cu-Cu atoms, given that their content is dominant in the melts and there is mainly a predominant neighborhood of these atoms. As for the pairs of Al-Al atoms, as we can see there are pairs of atoms, the distances between which are close to the interatomic distances of liquid aluminum, but the number of such pairs of atoms is less than others. The described

facts indicate the presence in the melts, even at high temperatures, clusters based on atoms of the same type.

Partial pair correlation functions calculated for pairs of atoms of different types (Fig. 3, b) also indicate a good correlation in the arrangement of these atoms. Analyzing the most probable interatomic distances in this case, we see that they, as in the case of distances between atoms of the same type, are less than the sum of the atomic radii of the elements (Table 1). In addition, the distances between atoms in the amorphous state are also smaller compared to the sum of the metal radii.

The decrease in interatomic distances in the studied alloys can be explained by the atypical charge distribution between aluminum and zirconium atoms, and partly between aluminum and copper, and the formation of a specific sp-d interaction. In this case, the predominant interaction between pairs of Al-Zr and Al-Cu atoms can lead to the formation of close to icosahedral clusters with an aluminum atom in the center. The presence of such clusters, as well as Cu-centered icosahedrons, was found in the amorphous alloy $Cu_{46}Zr_{47}Al_7$ [20].

Temperature dependences of partial interatomic distances indicate nonmonotonic change of some of them (Fig. 4). Physical interpretation of such results is possible only in the case of a comprehensive analysis of changes in interatomic distances and coordination numbers.

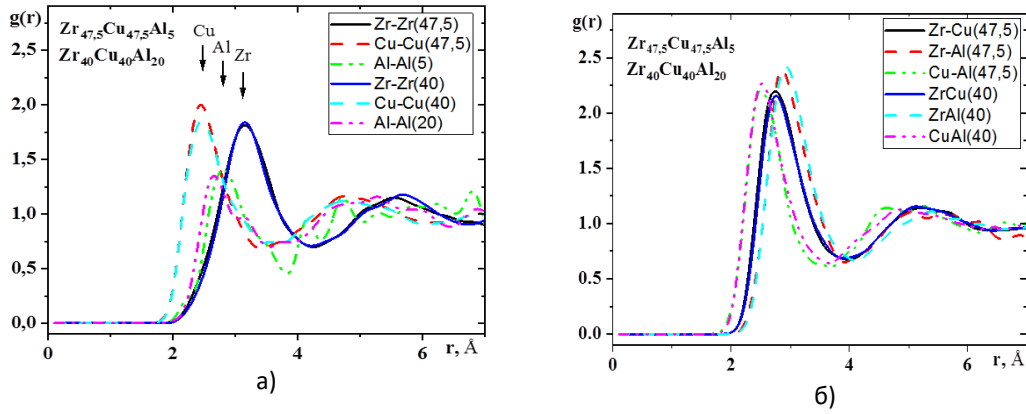

Fig. 3. Partial pair correlation functions of $Zr_{47.5}Cu_{47.5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts.

Table 1

 The main structure parameters of the $Zr_{47.5}Cu_{47.5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts in comparison with similar amorphous alloys.

Atomic pairs	The sum of atomic radii, Å	$Zr_{47.5}Cu_{47.5}Al_5$ (liquid state)		$Zr_{47.5}Cu_{47.5}Al_5$ (amorphous state [27])		$Zr_{40}Cu_{40}Al_{20}$ (liquid state)		$Zr_{40}Cu_{50}Al_{10}$ (amorphous state [19])	
		r_{ij} , Å	Z_{ij}	r_{ij} , Å	Z_{ij}	r_{ij} , Å	Z_{ij}	r_{ij} , Å	Z_{ij}
Zr-Zr	3.2	3.16	7.28	3.17	7.4	3.16	5.98	3.15	8.8
Cu-Cu	2.56	2.49	3.98	2.53	4.0	2.44	3.4	2.54	3.7
Al-Al	2.86	2.74	0.3	2.84	0.1	2.83	1.64	-	-
Zr-Cu	2.88	2.78	6.61	2.83	6.7	2.76	5.4	2.68	2.8
Zr-Al	3.3	2.94	0.7	2.99	0.8	2.88	2.97	2.99	3.2
Cu-Al	2.71	2.51	0.47	2.53	0.5	2.49	2.03	3.31	2.2

According to the generally accepted methodology, based on the distributions of atomic coordinates obtained from the simulation results, we calculated the distributions of partial coordination numbers (Figs. 5, 6). These distributions are probabilistic data on the presence of a certain number of atoms in the first coordination sphere of an atom placed in the center of the sphere. As can be seen from the figures, the distributions of the coordination numbers Zr-Zr, Cu-Cu, Zr-Cu and Cu-Zr are symmetric, and their most probable values can be determined by the position of the maximum of the distribution. On the other hand, due to the low content of aluminum, the partial coordination numbers with its participation in the $Zr_{47.5}Cu_{47.5}Al_5$ melt are close to one, indicating the presence of only one aluminum atom in the first coordination sphere of the alloy components. For the $Zr_{40}Cu_{40}Al_{20}$ melt in which the aluminum content is higher, the distribution of Al-Al coordination numbers is symmetric with a pronounced maximum near two. In this case, each aluminum atom can be surrounded by two aluminum atoms, which indicates the existence of chains of these atoms, which can be visually seen in Figure 2.

The distribution of coordination numbers Zr-Zr and Cu-Cu indicates the possibility of the existence of homocoordinated clusters of zirconium and to a lesser extent copper, as evidenced by the presence of coordination numbers close in value as in pure components.

In addition to homocoordinated clusters in the $Zr_{47.5}Cu_{47.5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts, a significant part of heterocoordinated clusters is also present. This is indicated by the significant number of atoms of copper,

zirconium and aluminum surrounded by atoms of other types. Taking into account the results shown in Figures 5, b and 6, b we can state that in the first coordination sphere of such atoms the fraction of different kind of atoms decreases according to the sequence $Zr \rightarrow Cu \rightarrow Al$. However, there is another possibility to provide the existence of a close to heterocoordinated distribution of atoms in the presence of clusters based on pure components in the melt. In particular, this is possible when such clusters form a branched quasi-two-dimensional structure. In this case, due to the large surface area of the clusters, different kind of atoms may have as many nearest neighbors belonging to homocoordinated clusters that coincide with our results. In our opinion, in the studied $Zr_{47.5}Cu_{47.5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts each of these models can be realized with an advantage towards one or the other, depending on the atomic composition of the melt.

Given the wide range of values of coordination numbers for different kind of atoms, we can assume the presence in the studied melts of structural units of several types, and the melts themselves can be represented as cluster solutions in which heterocoordinated clusters are located in a matrix of quasi-two-dimensional clusters. In this case, the clusters based on the components of the alloy are obviously not spherically symmetric, but form an interpenetrating framework in the cavities of which are located heterocoordinated structural units.

Temperature dependences of partial coordination numbers and interatomic distances indicate the rearrangement of the short-range order structure, which leads both to the homogenization of melts at heating and

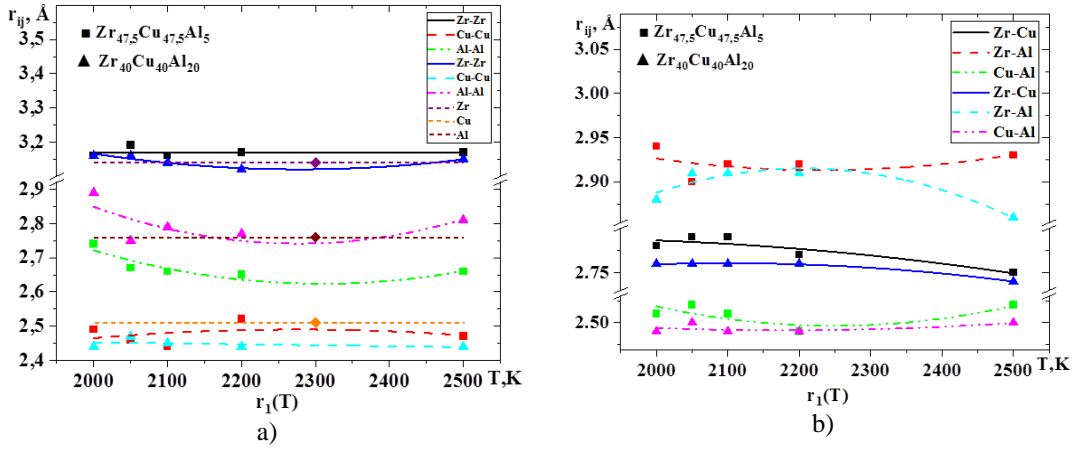


Fig. 4. The most probable partial interatomic distances for $Zr_{47.5}Cu_{47.5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts.

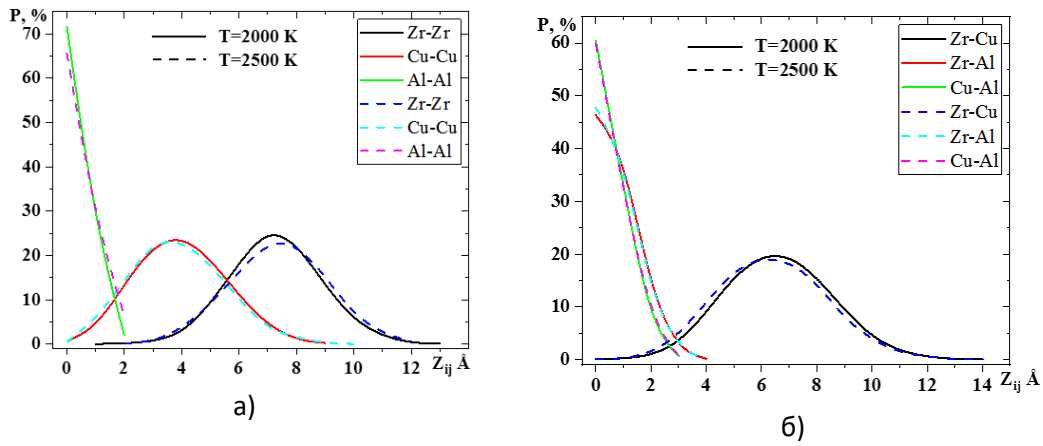


Fig. 5. Distribution of partial coordination numbers of $Zr_{47.5}Cu_{47.5}Al_5$ melt at different temperatures.

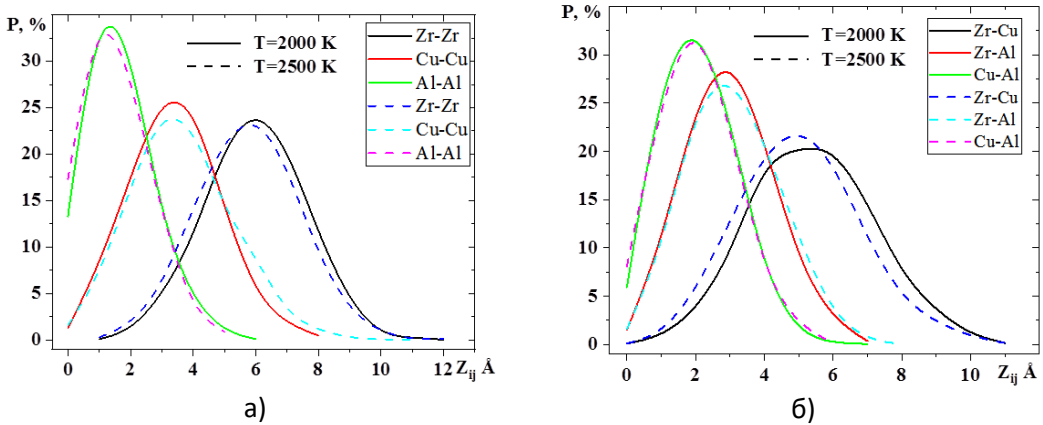


Fig. 6. Distribution of partial coordination numbers of $Zr_{40}Cu_{40}Al_{20}$ melt at different temperatures.

to a decrease in the density of atomic ordering. It should be noted that these changes are differently expressed in melts with different aluminum content, indicating its significant impact on the processes of interatomic interaction. A decrease in the coordination numbers of Zr-Zr and Cu-Cu at temperatures above 2200 K indicates the dissolution of clusters with predominant interaction of one kind of atoms in melts, which leads to an increase in the number of these atoms near aluminum and an increase in fraction of heterocoordinated clusters.

On the other hand, the observed decrease in

coordination numbers with a simultaneous decrease in interatomic distances may indicate a redistribution of atoms in such a way that there is a transition from a short-range order structure close to FCC in the crystalline state to BCC-like structural units. In particular, as pointed out by V. Goldschmidt [28], reducing the coordination number from 12 to 8 reduces the interatomic distances by 3 percent, and reducing the coordination number from 12 to 4 leads to a decrease in interatomic distances by 12 percent. This can be explained by the increase in the forces of interatomic interaction per atom, with a decrease in the

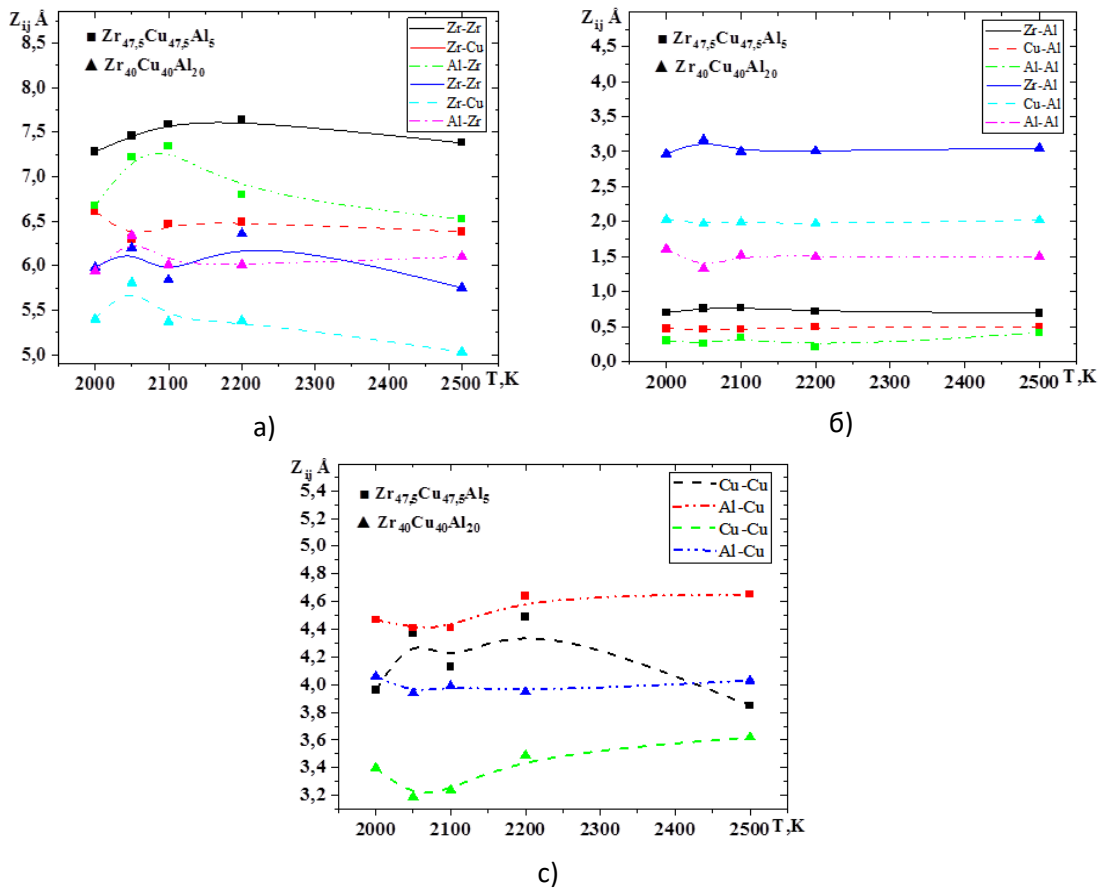


Fig. 7. Temperature dependences of the most probable coordination numbers of $Zr_{47,5}Cu_{47,5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts.

coordination number. Therefore, according to our results, we can assume the transformation of highly coordinated clusters into low-coordinated with a large number of cavities commensurate with the size of the atom. This assumption is also confirmed by an atypical change in the Zr-Zr partial coordination numbers. As we can see in Figure 7a, at temperatures above 2200 K the Zr-Zr coordination numbers decrease, while the number of zirconium's nearest neighbors in the vicinity of aluminum atoms also decreases, and in the vicinity of copper does not change. In this case, obviously, there is a destruction of highly coordinated clusters based on zirconium and an increase in the number of clusters of smaller size.

Conclusions

The structure of $Zr_{47,5}Cu_{47,5}Al_5$ and $Zr_{40}Cu_{40}Al_{20}$ melts in the temperature range of 2000K-2500K was studied by the method of molecular dynamics simulations. It is shown that the structure of these melts in the studied temperature range is close to the structure of amorphous alloys of similar composition. According to research results, even at high temperatures, the presence of clusters based on zirconium and copper with the proximity of atoms of the same variety has been established. Aluminum, in turn, can be surrounded in most cases by only two aluminum atoms, which indicates the existence of chains of these atoms in the melts $Zr_{40}Cu_{40}Al_{20}$.

The analysis of partial interatomic distances suggests

that their decrease in the studied alloys can be explained by the atypical charge distribution between aluminum and zirconium atoms, and partly between aluminum and copper, and the formation of a specific sp-d interaction. In this case, the predominant interaction between pairs of Al-Zr and Al-Cu atoms may lead to the formation of structural units close to icosahedral clusters with an aluminum atom in the center.

Given the wide range of values of coordination numbers for different kind of atoms, we can assume the presence in the studied melts of structural units of several types, and the melts themselves can be represented as cluster solutions in which heterocoordinated clusters are located in a matrix of quasi two-dimensional cluster formations based on pure components. In this case, the clusters based on the components of the alloy are obviously not spherically symmetric, but form an interpenetrating framework in the cavities of which are located heterocoordinated structural units.

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Структура розплавів системи Zr-Cu-Al в інтервалі концентрацій схильних до аморфізації.

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Методом молекулярної динаміки досліджено структуру ближнього порядку розплавів $Zr_{47,5}Cu_{47,5}Al_5$ та $Zr_{40}Cu_{40}Al_{20}$. На основі отриманих результатів розраховано парціальні парні кореляційні функції та розподіли парціальних координаційних чисел. Основні структурні параметри визначені з вказаних функцій порівняли з аналогічними параметрами для аморфних сплавів. За результатами досліджень встановлено наявність в розплавах як гомокоординованих так і гетеро координованих кластерів, які формують кластерний розчин. Зроблено припущення про наявність гетерокоординованих ікосаедричних кластерів.

Ключові слова: металеві сплави, атомна структура, координаційні числа, кластерна будова.