



Glass transition of aluminum melt. Molecular dynamics study



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ABSTRACT

Molecular dynamics study of transition from liquid aluminum into an amorphous state is carried out. Different criteria for the glass transition are compared with each other: splitting of the second peak of the radial distribution function, increasing of the number of atoms with icosahedron-like environment, increase of the self-diffusion activation energy, changes in the heat capacity. The discrepancy of the results obtained by different criteria is discovered. Heat capacity dependence on temperature agrees qualitatively with theoretical and experimental results. Influence of cooling rate on the glass transition temperature is studied. Although different methods give different glass transition temperatures, the dependencies on cooling rate may be interpreted in the Volkenstein and Ptitsyn's framework using either Arrhenius or Vogel–Fulcher–Tammann relaxation laws.

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1. Introduction

Amorphous metals have a unique non-crystalline structure. Because of this, they have physical and magnetic properties combining strength and rigidity with the flexibility and toughness which are unusual for polycrystalline solids. It is known that there is a strong dependence of the structure and properties of glasses on the conditions and methods of manufacturing [1–8]. The main problem of the physics of liquid-glass transition is how to distinguish these two noncrystalline phases clearly [9–13]. There is no generally accepted theory describing the details of the glass transition. A large variety of experimental data, corresponding to different laws for different matters, leads to a large number of theories. For example, the glass transition in [6,7,14] is considered in the approximation of mode-coupling theory in the system described by a form of repulsive shoulder potential of solid spheres. This transition is associated with changes of some “order parameter” in [1,2,4,5].

Glass transitions and phase transition have a lot in common. Many properties of material such as the specific heat, compressibility and viscosity undergo a sharp change in a narrow temperature range. Nevertheless, despite some similarity, the glass transition is not a

phase transition in a strict sense. Thus, one can obtain glasses with different internal structure at different cooling rates.

Transition is not accompanied by sharp symmetry change at the molecular level as in the case of the liquid-crystal transition. Both experiments and atomistic simulations for a number of substances confirm that the second peak of the radial distribution function (RDF) begins splitting when the melt viscosity undergoes a sharp change [9]. So, splitting of the second peak of RDF can be used as a criterion for the glass transition.

The molecular dynamics (MD) method allows us to describe physical processes on the atomistic level. One can perform a detailed study of glass and phase transitions and obtain different characteristics of this process [15–21]. RDF is usually used in MD simulations for analysis of the amorphous structure and determining the glass transition. Voronoi polyhedra are used for a more detailed examination of the structure [22,23]. Influence of the number of different types of Voronoi polyhedra on the form of pair correlation functions second peak is considered in [9].

In this paper, glass transition of aluminum melt during isobaric cooling is considered using the MD method. In Section 2, the calculation technique is described. Structural and dynamic criteria for the glass transition are compared with each other in Section 3. Calorimetric criteria for calculation of glass transition temperature on cooling and re-heating are considered in Section 4. Influence of cooling rate on the glass transition temperature is studied in Section 5. Conclusions follow in Section 6

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2. Molecular dynamics model

Aluminum melt cooling is taken as an example to study a glass transition. Liquid aluminum is described by a many-body embedded atom method potential [24]. The potential energy of the system in this approach is represented as follows:

$$U = \sum_{i<j} \varphi(r_{ij}) + \sum_j F(\bar{\rho}_j), \bar{\rho}_j = \sum_{j \neq i} \rho(r_{ij}). \quad (1)$$

The first term is a sum of pair potentials φ over all atom pairs in the system, r_{ij} is the distance between atoms i and j . A non-linear embedding function $F(\rho)$ introduces many-body effects. $\bar{\rho}$ is an effective electron density that is induced by the neighboring atoms at the given atom and $\rho(r_{ij})$ is a contribution from each neighbor. The parametrization of the potential is suggested in [25].

Molecular dynamics method [18,26] is used to study the aluminum melt. Simulations are performed using the LAMMPS MD software [27]. Numerical integration of the equations of motion is done using 1 fs timestep.

To create the initial configuration, atoms are placed at the sites of the fcc lattice with parameter $a_0 = 4.08 \text{ \AA}$. A simulation box $40a_0 \times 20a_0 \times 20a_0$ along the axes x, y, z with the periodic boundary conditions on all axes is used. 32,800 atoms are placed as a thin film which takes half of the volume of the simulation box ($x \geq 20a_0$).

Free space in the simulation box allows the film to change its thickness during the cooling process. That is why the atoms are able to occupy a volume corresponding to a constant near-zero pressure. Therefore, no additional barostat is necessary to apply to maintain the isobaric conditions during cooling or heating processes. This is similar to the real conditions.

At the initial step, random (in magnitude and direction) velocities are given to all the atoms. They are normalized so that the kinetic energy of the atoms corresponds to the temperature $T = 2000 \text{ K}$. A crystal melting process is carried out at 2000 K for the first 900 ps. Then, the system is cooled down to 1500 K and additional equilibration at this temperature takes place.

The temperature of the system is changed by the following procedure. First, we choose a cooling rate and calculate the desired linear dependence of temperature on time. The time integration is then performed using the conventional velocity Verlet scheme. The velocities of atoms are rescaled every 100 fs to match the chosen dependence of temperature on time. This method allows us to avoid unphysical temperature oscillations characteristic to the Nose-Hoover thermostat with the rapidly changing target temperature.

For the study of the glass transition, rapid cooling of melt from 1500 K to 300 K is carried out at constant rates. MD simulations are performed for cooling rates from 10^{10} K/s to 10^{13} K/s .

3. Criteria for the glass transition

Two structural criteria and a dynamical one are considered in this section. Changes in the RDF, number of icosahedral clusters and self-diffusion coefficient are monitored during the cooling.

The procedure is repeated for a number of cooling rates. The results are presented below for the rate $|dT/dt| = 10^{13} \text{ K/s}^1$ as an example. They are typical for the all cooling rates considered.

3.1. Splitting of the second peak of RDF

The well-known criterion is splitting of RDF's peak in the second coordination sphere. It corresponds to the appearance of two local maxima and a local minimum between them. This is equivalent to the

existence of r_0 , for which $G(r_0 \pm h) - G(r_0) > 0$, where $G(r)$ is the RDF and h is the spatial step the RDF is calculated with.

The area S enclosed between the tangent drawn at the point r_0 to the graph of RDF and the graph of RDF itself in the second coordination sphere is used as the mathematical measure of peak splitting (Fig. 1):

$$S = \int_{r_1}^{r_2} (G(r) - G(r_0)) dr \quad (2)$$

where r_1 and r_2 are determined from the condition $G(r_{1,2}) = G(r_0)$. There are no such points when RDF is far from splitting, and thus the integral in Eq. (2) reduces to zero for the liquid state.

The results are presented for the whole temperature range in Fig. 2a. The range of the glass transition reveals itself about 900 K.

3.2. Number of icosahedral clusters

Extremely supercooled metallic melts of one-component systems are characterized by an icosahedral short-range order [28,29,30,31]. Recently more and more papers are devoted to the description of metallic glass structure through the packaging of clusters with different local order [32,33,34]. In particular, the 13-atom icosahedral clusters are proposed as a structural unit responsible for the formation of metallic glasses [13,35,36]. Recently their existence in metallic glasses were confirmed experimentally for $Zr_{36}Cu_{64}$ alloy [13] by X-ray scattering. However, it is also shown that not all metal alloys demonstrate increase of number of such clusters at glass transition.

Analysis of the number of icosahedral cluster is carried out at the aluminum melt cooling in this paper. Common Neighbor Analysis (CNA) [37] is used to identify them. This method is also used for identifying crystal lattice short-range order during crystallization. Atom with its nearest neighbors corresponding to the first peak of RDF forms "bonded" pairs. These bonds are classified by the combination of 3 indexes $ijkl$. The first index, j , is the number of neighbors common to both atoms. The second index, k , is the number of bonds between these common neighbors. The third index, l , is the number of bonds in the longest continuous chain formed by k bonds between common neighbors. After it, different types of bonds are associated with different local order. Icosahedral order is characterized by 555 bonds. Number of atoms participating in twelve 555 bonds is thought of as the number of icosahedral clusters N_{ic} .

There are no icosahedral clusters at high temperatures or their quantity differs insignificantly from zero. However, N_{ic} begins to increase

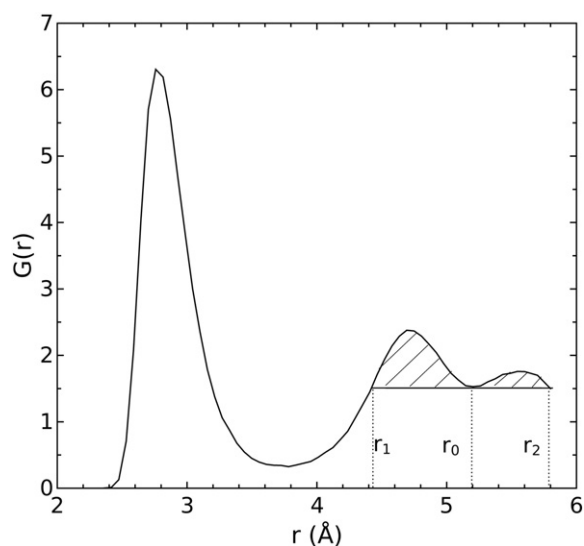


Fig. 1. Splitting of the second peak of radial distribution function. r_0 is determined from the Eq. (2), $r_{1,2}$ satisfy the condition: $G(r_{1,2}) = G(r_0)$.

¹ We will further use absolute values of temperature change rate, the rate itself is negative in the cooling process and positive in the re-heating process.

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