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Medium range order evolution in pressurized sub-T_g annealing of $Cu_{64}Zr_{36}$ metallic glass

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ABSTRACT

Molecular dynamics (MD) simulations have been widely used to study the structure of metallic glasses (MGs) at atomic scale. However, ultrafast cooling rates in MD simulations create structures that are substantially under-relaxed. In this study, we introduce long-term pressurized annealing up to 1 μs slightly below the glass-transition temperature, T_g, in MD simulation, which effectively relaxes the structure of $Cu_{64}Zr_{36}$ MG toward experimental conditions. It is found that applying hydrostatic pressure up to 2 GPa relaxes the MG to low-energy states whereas higher pressures retard relaxation events. In the sample annealed at 2 GPa pressure, equivalent cooling rate reaches to 3.7×10^7 K/s, which is in the order of melt spinning experiments. Furthermore, the correlation between structural relaxation and medium-range structures formed by interpenetrating connection of icosahedra is studied. Clustering coefficient analysis shows that in the samples prepared at high cooling rates, interpenetrating icosahedral networks are fragmented. However, as the glass relaxes toward real conditions, a unified network develops throughout the glassy structure, which acts as a solid-like backbone. The morphology of such a network differs from string-like fashion proposed in the previous studies.

1. Introduction

Metallic glasses are characterized by metallic bonds with no long-range translational order [1], which provides them with unique properties such as outstanding strength [2,3], high fracture toughness [4,5], superior thermoplastic formability [6] and good biocompatibility [7,8] which make them a promising candidate material for a wide range of applications. Several experimental [9,10] and molecular dynamics (MD) [11–16] investigations revealed that, despite lacking long-range order, MGs do have short range (SRO) and medium range ordered (MRO) structures in the atomic scale. It has been shown that full icosahedron (FI), with voronoi index of $< 0,0,12,0 >$, has the lowest dynamics among all SRO structures and its population rapidly increases in supercooled liquid region during cooling [11,17,18]. In addition, icosahedra exhibit stronger self-aggregation tendency than other types of clusters and form a connected network in the MG structure, the so-called MRO [19–21]. Recent MD investigations show that MRO in MGs has close correlation with slow dynamics and mechanical properties [14,22–26]. Meanwhile, many theories, such as the theory of random first order-transitions [27] and spin glass approaches [28] predict the existence of rapidly growing static length scales in MGs to describe the rapidly increase of the viscosity near glass transition temperature.

While it is widely believed that the MRO has close correlation with unusual properties of MGs, it is still not clear how this complex structure develops and what factors affect its evolution. The MRO in MGs is difficult to study by microscopy or diffraction measurements because the higher-order correlations (fourth or higher peaks in PDF) are lost when Fourier transforming the diffraction data to the pair-distribution functions. However, the splitting in the second peak observed by diffraction experiments has been attributed to MRO structures via MD simulation [29]. Up to now, many of the MRO models in MGs have been derived from MD simulations. However, ultrafast cooling rates in MD simulations ($\sim 10^9$ K/s or higher [11]), almost three orders of magnitude higher than experimental ones ($\sim 10^3$–10^7 K/s [30]), result in structures that are substantially under-relaxed. Therefore, efficiently relaxing the structure in MD simulation is required to construct models closer to real condition.

Experimental and simulation studies [31–35] have demonstrated that sub-T_g annealing is an effective route to relax the as quenched glass to a more stable state. Zhang et al. [31] showed that by sub-T_g annealing the effective cooling rates in MD simulations reduces effectively. Experimental investigations on sub-T_g annealing of MGs [36–39] have demonstrated that, other than temperature, hydrostatic pressure (HP) is another variable that could be used to tune the structure of MGs toward a more relaxed state. Ruitenberg et al. [37] attributed the structural relaxation during pressurized annealing of the MGs to

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annihilation of structural defects such as free volume. Because MGs are meta-stable material, various rearrangement processes at elevated temperature give rises to the configuration corresponding to lower internal energies.

In the present study, we introduce long-term pressurized sub-T_g annealing as an effective tool to relax the $Cu_{64}Zr_{36}$ bulk glass former alloy [40,41] in MD simulation and produce samples with equivalent cooling rates closer to experimental condition. Such a model could catch the main structure features of real MGs. Furthermore, the correlation between structural relaxation and the MRO structures formed by interpenetrating icosahedral cluster is studied. By using network analysis, it will be shown that the size and morphology of MRO structures strongly depend on degree of relaxation hence on cooling rate.

2. Simulation method

Classical MD simulation of $Cu_{64}Zr_{36}$ MG was performed using LAMMPS package [42]. The interatomic interactions of the atoms were described by embedded atom model (EAM) potential developed by Sheng et al. [43] that satisfactory describes the glassy structure of the Cu-Zr system. Initially, 16,000 atoms with desired composition were randomly distributed in a cubic box with periodic boundary condition in all dimensions. After equilibrating at 2000 K for 2 ns (well above the liquidus temperature, 1324 K [44]) under the isobaric-isothermal (NPT) ensemble, the samples were cooled down to 700 K (below glass transition temperature) at a rate of 10^{10} K/s. The time step for integration of Newtonian equations of motion was 2 fs. Then, the models were isothermally annealed at 700 K for 1 μs at different hydrostatic pressures (0, 1, 2, 5 GPa). After sub-T_g annealing, the samples were cooled down to 300 K at a constant cooling rate of 10^{10} K/s while keeping the external pressure. Finally, at 300 K, the hydrostatic pressures were removed and the samples were further relaxed for 2 ns. Each Isothermal annealing was performed on 10 CPU cores (Intel Xeon 5680 3.3Ghz) and takes 10 × 840 CPU Hours. For comparison, four additional models of the same size and composition were created by uniformly cooling the liquid state from 2000 K to 300 K, using four different cooling rates (10^{13}, 10^{12}, 10^{11} and 10^{10} K/s). The structures of the glassy alloys at atomic scale were analyzed by voronoi tessellation method [45].

3. Results and discussion

Fig. 1a shows the instantaneous potential energies (PEs) during sub-T_g annealing at different hydrostatic pressures as a function of annealing time. In all cases studied, the PEs decrease monotonically with time demonstrating that the MGs undergo relaxation during long-term annealing. Below 2 GPa pressure, PEs decrease much faster and beyond this range, the decrease starts to slow down (supplementary materials Fig. S1). Reduction of the PE is more pronounced in the sample annealed at 2 GPa hydrostatic pressure demonstrating that the sample visited lower-energy sub-basin during annealing. The similar trend has also been observed experimentally [36,38] and has been attributed to activation volume for relaxation events. At low pressures, the relaxation is associated with topological rearrangement of atoms leading to annihilation of internal defects such as free volumes, whereas at high pressures hard-core repulsive forces gradually become dominant [46]. Because temperature is near to glass transition temperature, both thermal activated and stress driven mechanisms are active, therefore the rearrangement process could take place almost homogeneously (supplementary materials Fig. S2, S3 and S4). Recent investigation by Ding et al. [47] have demonstrated that pressurized quenching at very high hydrostatic pressure (up to 20 GPa) results in rejuvenation of MGs. It is known that glass transition temperature increases with hydrostatic pressure when metallic liquids are quenched to glassy states [38]. Therefore, during quenching, liquids with higher internal energy turn into glasses leading to formation of high energy MGs. In contrast,

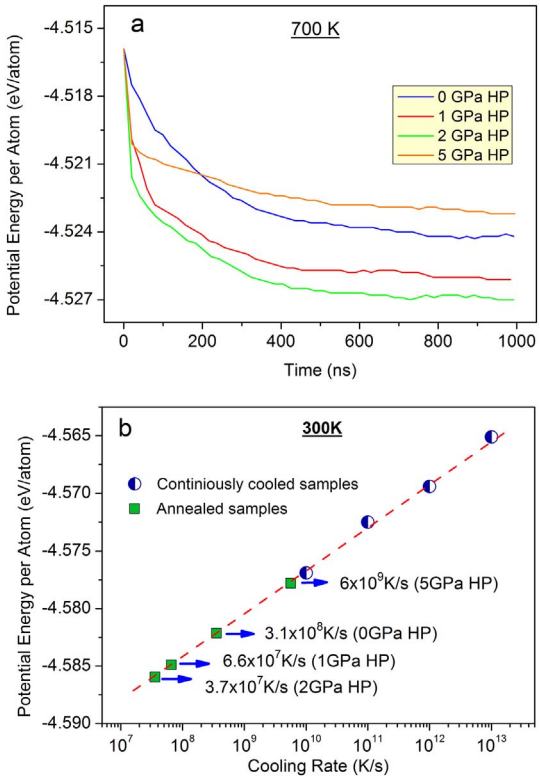

Fig. 1. a) PE variation as a function of time during pressurized sub-T_g annealing. b) PE of all the models after equilibration for 2 ns at 300 K as a function of cooling rates; the red dashed line indicates a linear fitting to a logarithmic dependence for all the uniformly cooled samples (circles). The green squares represent the potential energy of pressurized annealing samples. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

annealing below glass transition temperature under low hydrostatic pressures is accompanied by short range topological and chemical reordering leading to formation of more stable local structures as it is evident from continuous reduction of PE during annealing.

The PEs at 300 K for the continuously cooled models and annealed models as a function of cooling rate are plotted in Fig. 1b. The value of PEs are calculated by averaging over the last 1 ns of two-nanosecond isothermally relaxed samples at 300 K. As it can be seen in this figure, potential energies of continuously cooled samples vary linearly with logarithm of cooling rates (red dashed line in the figure). Such a correlation has also been reported for the same alloy by Zhang et al. [31] using different interatomic potential. Assuming this linear relationship, the samples annealed at $P = 0$, 1, 2 and 5 GPa have PEs that are equivalent to PEs of continuously cooled samples with rate of are 3.1×10^8 K/s, 6.6×10^7 K/s, 3.7×10^7 K/s, and 6×10^9 K/s respectively. Therefore, annealing at low hydrostatic pressure effectively accelerates relaxation phenomena and pushes the equivalent cooling rate toward realistic condition of MGs. Comparing the models annealed at $P = 0$ and 2 GPa reveals that applying hydrostatic pressure during sub-T_g annealing further reduces the equivalent cooling rate by an order of magnitude. The equivalent cooling rate in the sample annealed at 2 GPa is at least two orders of magnitude lower than conventional MD simulations and is near to the cooling rates in melt spinning experiments. The atomic density of this sample reaches 0.0632 atom/Å^3, which compares well with the value reported for melt spinning ribbons of the alloy with the same composition (0.0637 atom/Å^3 [48]).

The calculated pair distribution functions (PDF) of the MD models prepared at cooling rate of 10^{13} K/s (loosely-relaxed) and sub-T_g annealing at 2 GPa pressure (well-relaxed) are compared with experimental X-ray diffraction data reported in ref. [49] in Fig. 2. The PDFs of the both MD models agree with the experiment in term of positions and

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