SPECIAL TOPIC — Amorphous physics and materials

Effect of icosahedral clusters on β-relaxations in metallic glasses*

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(Received 13 September 2016; revised manuscript received 12 October 2016; published online 29 November 2016)

The most pronounced β-relaxation was found in the Y-based binary metallic glasses (MGs). The correlation between β-relaxation and local atomic structure was studied. The dynamic mechanical measurements were performed for three chosen binary systems: Zr-, Ti-, and Y-based MGs. The experimental results show that, in each system, the larger negative enthalpy of mixing ($\Delta H_{\rm m}$) between the component elements makes β-relaxation become more pronounced. The less negative value of $\Delta H_{\rm m}$ facilitates the formation of icosahedral clusters, which have a pinning effect on the excitation of β-relaxations and correspondingly make the β-relaxation become less pronounced. These chemical effects on β-relaxations can only be compared in the same MG system, and it is not suitable for the comparison between different systems due to the different features of the major metallic elements.

Keywords: metallic glass, β-relaxation, enthalpy of mixing, icosahedral clusters

PACS: 61.25.Mv, 61.20.Ne, 64.70.pe, 62.40.+i **DOI:** 10.1088/1674-1056/26/1/016101

1. Introduction

Metallic glasses (MGs) show diverse relaxation dynamics, which has attracted considerable attention for the importance in understanding the nature of glasses.[1–5] At high frequencies or low temperatures in dynamic response spectra, β-relaxation splits out from the primary α-relaxation,[6] and plenty of work on β-relaxation has been done in MGs.[7–9] It is demonstrated that β-relaxation is the principal dynamics source in the glassy state and is of practical significance to many physical, chemical, and mechanical properties.[10–12] Although plenty of efforts have been made on β-relaxation, the correlation between the local structural feature and β-relaxation is not well understood, so that the structural origin of β-relaxation is still a controversial issue.[4] For most MGs, only a β-relaxation shoulder or an excess wing has been observed on the low-temperature flank of α-relaxation.[4,13–16] Until recently, La-based MGs have been found showing a pronounced β-relaxation peak in the dynamic mechanical spectrum.[7] Therefore, β-relaxation modes depend on the strength of β-relaxation and the coupling degree between α- and β-relaxations. It is still confusing why there are such significant different appearances of β-relaxations between different MG systems and even different component combinations in the same system, and this question fundamentally connects to the structural origin of β-relaxations. Recently, it was identified that pronounced β-relaxation is associated with systems in which all the atomic pairs have large similar negative values of enthalpy of mixing ($\Delta H_{\rm m}$) by Cu and Ni substitution in La–Ni/Cu–Al and Pd–Ni/Cu–P MGs, and a physical picture emerged that strong and comparable interactions among all component atoms maintain string-like atomic configurations for the excitation of β-events.[17] However, the universality of this chemical effect still needs to be verified.

In this work, we address an important question of β-relaxation in connection with local atomic structure, especially icosahedral clusters. Binary MGs, due to the relative simple structures, are selected as model systems to study the pinning effect on β-relaxation. We compare the dynamics of the β-relaxations in three typical binary systems which have different $\Delta H_{\rm m}$. Recently, a general rule of the chemical effect was proposed that more pronounced β-relaxations are correlated to stronger chemical interactions among component atoms.[17] However, according to the present study, the rule is only applicable for different compositions in one specific system but not applicable between different systems. Based on the chemical effect on local structure, we suggest that icosahedral clusters have a strong pinning effect on the motion of β-relaxation, which is similar to the effect of solute atoms on the motion of dislocations in crystalline alloys.

2. Experiments

Three binary systems, $Zr_{66.6}(Cu/Ni)_{33.4}$, $Ti_{60}(Cu/Ni)_{40}$, and $Y_{66.6}(Cu/Ni)_{33.4}$, were selected for the experiments by considering their good glass-forming ability and different $\Delta H_{\rm m}$. Amorphous ribbons of six compositions were obtained by the melt spinning technique, and the fully glassy nature

*Project supported by the National Basic Research Program of China (Grant No. 2010CB731603) and the National Natural Science Foundation of China (Grant Nos. 50921091 and 51071170).

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was identified by x-ray diffraction and differential scanning calorimetry. Dynamic mechanical measurements were performed using a TA dynamical mechanical analyzer (DMA). The storage modulus E' and loss modulus E'' were measured by the temperature ramp mode and various testing frequencies. The molecular dynamics (MD) simulations were performed with the LAMMPS code and a realistic embedded-atom method potential.[18] The Zr-based system was chosen, and it contains 10000 atoms with periodic boundary conditions. In the process of sample preparation, it was first melted and equilibrated at $T = 2000$ K for 1 ns, then cooled down to 300 K with a cooling rate of 10×10^{12} K/s. The samples were further relaxed at 300 K for 2 ns. The configurations in the last 1 ns were collected for structural and dynamical analysis.

3. Results and discussion

To provide an intuitionist comparison of the β-relaxations in the same and different systems, we fit the whole relaxation curves by coupling two Kohlrausch–Williams–Watts (KWW) equations in the Fourier transforms of $d\{\Delta E \cdot \exp[-(t/\tau)^{\beta_{\rm KWW}}]\}$.[19] Here, $\Delta E = E(f=\infty) - E(f=0)$, where $E(f=\infty)$ and $E(f=0)$ represent instant elastic modulus and static elastic modulus, and f and τ are frequency and average relaxation time. $\beta_{\rm KWW}$ is a nonexponential parameter related to the dynamic heterogeneity. The T dependent τ_β for β-relaxation has the form of the Arrhenius process $\tau_\beta = \tau_{\beta0}\exp(E_\beta/RT)$, and τ_α is described by the Vogel–Fulcher–Tammann (VFT) equation $\tau_\alpha = \tau_{\alpha0}\exp(DT_0/(T-T_0))$. The pre-factors for the six metallic supercooled liquids are $\tau_{\alpha0} \sim 10^{-13}$ s and $\tau_{\beta0} \sim 10^{-15}$ s, respectively, which are consistent with the previous works.[2]

Figure 1 shows the T dependent E'' of the three systems measured by DMA at 1 Hz, where T and E'' are scaled by the temperature of the α-relaxation peak $T_{\rm p\alpha}$ and the loss modulus $E''_{\rm p\alpha}$ at $T_{\rm p\alpha}$, respectively. $T_{\rm p\alpha}$ and $E''_{\rm p\alpha}$ are both obtained from fittings. The circles represent the experimental data and the red and blue regions represent the scaled KWW fits of the β-relaxations for M–Cu and M–Ni (M = Zr, Ti, and Y), respectively. Considering the scaled fits of the α-relaxations nearly coincide for M–Cu/Ni, they are represented by a single grey region. The fitting of storage modulus data, which is not presented here, was carried on as an adjunct to the fitting position and intensity of the α-relaxation peaks. The β-relaxation activation energy E_β and $T_{\rm p\alpha}$ obtained from the fitting are listed in Table 1. As shown in Fig. 1(a), a β-relaxation hump can be observed at the low temperature flank of the α-relaxation peak for $Zr_{66.6}Ni_{33.4}$, while $Zr_{66.6}Cu_{33.4}$ has only an excess wing, indicating that the substitution of Cu for Ni influences the β-relaxation behavior significantly. Similar results are observed in Ti-based MGs as shown in Fig. 1(b), however, the weak hump is further suppressed into an excess wing by the substitution of Cu for Ni. Figure 1(c) shows the effect of Cu/Ni substitution for Y-based MGs, which has the same tendency as that for Zr- and Ti-based MGs. Meanwhile, in sharp contrast to Zr- and Ti-based MGs, Y-based MGs exhibit the significant pronounced β-relaxation peaks at the temperature about $0.7T_{\rm g}$ (the glass transition temperature) in the deep glassy state. It is so far the most obvious β-relaxation behavior found in MGs. The β-relaxation in Y-based MG is much stronger than that of La-based MG which was the strongest one in the previous works. Another characteristic is that the β- and α-relaxations of $Y_{66.6}Ni_{33.4}$ MG are more separated compared with those of the other systems. It is also noteworthy that $T_{\rm p\beta}/T_{\rm g}$ for $Y_{66.6}Ni_{33.4}$ (about 0.69) is even lower than that for La-based MGs (about 0.78) which is well known for the obvious β-relaxation.[7,12]

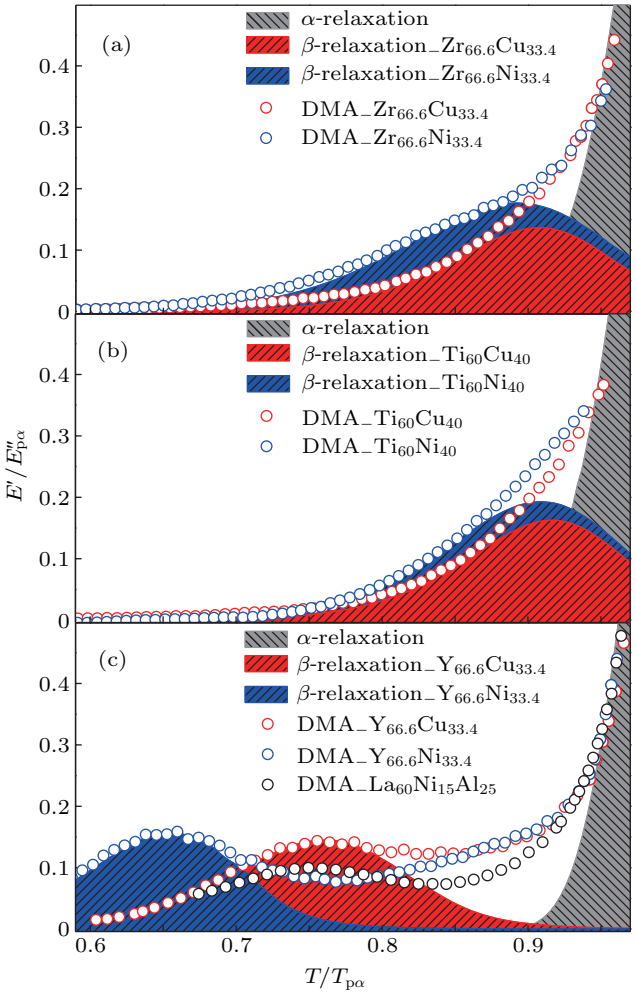


Fig. 1. (color online) $E''/E''_{\rm p\alpha}$ as a function of $T/T_{\rm p\alpha}$ for (a) $Zr_{66.6}(Cu/Ni)_{33.4}$, (b) $Ti_{60}(Cu/Ni)_{40}$, and (c) $Y_{66.6}(Cu/Ni)_{33.4}$. The circles are the experimental data. The red and blue regions represent the scaled KWW fits of the β-relaxations for M–Cu and M–Ni (M = Zr, Ti, and Y), respectively. The grey regions are fits of the α-relaxations.

All three systems have large negative $\Delta H_{\rm m}$[20,21] in a wide range as shown in Table 1. Here we define the reduced activation energy of β-relaxation $E_{r\beta} = E_\beta/RT_{\rm p\alpha}$, and the smaller $E_{r\beta}$ indicates the separation between α- and β-peaks is more

significant, which means the β-relaxation peak is more pronounced. The β-relaxation changes from an excess wing to a hump, finally to a peak with decreasing $E_{r\beta}$. $E_{r\beta}$ was evaluated and listed in Table 1. For each system, the more negative ΔH_m between two elements is, the less $E_{r\beta}$ will be, shown in Table 1 and Fig. 2. The $E_{r\beta}$ does not change significantly for Zr- and Ti-based systems, but the β-relaxation strength changes about 20% for both systems, which leads to the β-relaxation changing from excess wings to humps. It indicates that $E_{r\beta}$ is a sensitive parameter to the appearances of β-relaxations. This is further verified by Y–based MGs. With the smallest $E_{r\beta}$ for Y–Ni, the β-relaxation is farthest from the α-relaxation and the most pronounced.

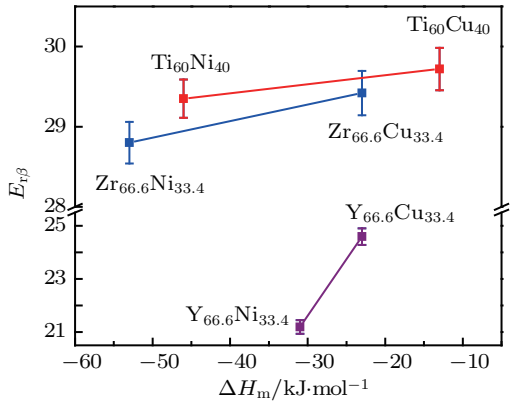


Fig. 2. (color online) The reduced activation energy $E_{r\beta}$ versus ΔH_m.

Table 1. Enthalpy of mixing ΔH_m, β-relaxation activation energy E_β, glass transition temperature T_g, temperatures corresponding to β- and α-relaxation peaks $T_{p\beta}$ and $T_{p\alpha}$, and the reduced activation energy $E_{r\beta}$.

Systems	ΔH_m/kJ·mol^{-1}	E_β/kJ·mol^{-1}	T_g/K	$T_{p\beta}$/K	$T_{p\alpha}$/K	$T_{p\beta}/T_g$	$E_{r\beta}$
$Zr_{66.6}Cu_{33.4}$	–25	159±1	622	588±2	648±2	0.95	29.4
$Zr_{66.6}Ni_{33.4}$	–53	165±1	671	612±2	687±2	0.91	28.8
$Ti_{60}Cu_{40}$	–12	168±1	632	622±2	678±2	0.98	29.7
$Ti_{60}Ni_{40}$	–46	184±1	712	682±2	752±2	0.96	29.3
$Y_{66.6}Cu_{33.4}$	–22	110±1	507	396±2	538±2	0.78	24.6
$Y_{66.6}Ni_{33.4}$	–31	111±1	598	412±2	630±2	0.69	21.2

Now the questions arise what the connections between ΔH_m, $E_{r\beta}$ and local structures are and how these factors affect β-relaxation. As we known, mainly three factors affect local atomic structures in binary MGs, which are atomic radius, electronic configuration, and chemical interaction between atomic pairs. In order to study the chemical effect without interference of other factors, we carried out the substitution of Cu and Ni which have similar atomic radii and electronic configurations in each system. When considering different systems, the observation and analysis of the chemical effect are interfered by different principal elements. For instance, ΔH_m of Y-based MG are only about 20–30 kJ/mol, which are not the most negative ones in the three systems, however, the β-relaxations are significantly pronounced, consistent with their small $E_{r\beta}$. Such results further indicate that β-relaxations depend not only on the chemical effect between elements, but also strongly on the characteristics of major elements which might lead to different local structures. It is noticed that compared with transition elements, rare earth elements have some special properties so that the pronounced β-relaxations have only been observed in rare earth based MGs, such as La-based,[7] specially Y-based found in this work. Despite the complexity of the three factors, some regularity still could be figured out. Comparing M–Ni with M–Cu in each system, the MGs with larger negative ΔH_m have smaller $E_{r\beta}$. Thus, the chemical effect rule is valid for each system, but not applicable for the comparison between different systems. To compare different systems, besides the chemical effect, atomic radius and electronic configuration should also be considered.

The findings above demonstrate that the chemical effect on β-relaxations can only be compared in the same system. Meanwhile a question arises around how to understand this chemical effect of ΔH_m. According to the coupling model proposed by Ngai,[15] an equation could be obtained as follows: $(\log\tau_\alpha - \log\tau_\beta) \approx n(\log\tau_\alpha - \log t_c)$, where τ_α and τ_β are α- and β-relaxation time respectively, t_c is a constant about 2×10^{-12}s, and the coupling parameter n is associated with the interaction between particles and increases with stronger interaction. Larger negative ΔH_m means that n and therefore $(\log\tau_\alpha - \log\tau_\beta)$ are larger, thus the separation between α- and β-peaks is more significant. Furthermore, ΔH_m provides an effective way to understand many chemical-related issues, such as the local structure. Historically, hard sphere dense random packing has been widely cited as a structural model for MGs.[22] It is now understood that the model fails to describe many binary MGs with significant chemical short-range order (CSRO) which is associated with atomic size ratio and ΔH_m.[23] The short-range order (SRO) in $Zr_{60}Cu_{20}Pd_{10}Al_{10}$ liquid was investigated by the small-angle x-ray scattering,[24] and revealed that larger negative ΔH_m is related to the stronger attractive interaction between solute and solvent. The coordination number (CN) of Zr–Pd pairs increases significantly with lowering liquid temperature, while that of Zr–Cu pairs increases slightly, indicating that the large negative ΔH_m between Zr and Pd (−91 kJ/mol) leads to the formation of (Zr, Pd)-rich domains of SRO in the supercooled liquid.

To obtain a quantitative estimation for the CSRO in binary supercooled liquids, the CSRO degree was estimated using the Warren parameter[25]

$$\alpha_{\rm W} = 1 - \frac{N_{ij}}{c_j N} = 1 - \frac{N_{ji}}{c_i N},$$

where c_j is the concentration of species j, $N_i = N_{ii} + N_{ij}$ the total CN around species i, and $N = c_j N_i + c_i N_j$. Negative values of $\alpha_{\rm W}$ indicate a preference for unlike-neighbor bonds, otherwise homopolar pairs are preferred.[25,26] Honeycutt and Anderson pair analysis is able to characterize the local environment surrounding each atomic pair, and has been widely used to characterize the local cluster structure.[27] In this method, a set of four integers i, j, k, j is designed to describe different local configurations. For instance, 1551 index is the characteristic of icosahedral short range order (ISRO), while 1541 and 1431 indices are the characteristic of distorted ISRO.[26,28] Usually, 1551, 1541, and 1431 pairs are referred to the common local atomic configuration characteristic of supercooled liquid. Table 2 summarizes the data of $\Delta H_{\rm m}$, $\alpha_{\rm W}$, and the relative abundance w of the selected main bonded pairs $(1551 + 1541 + 1431)$ for different MGs and liquid alloys. Figure 3 shows the dependence of $\alpha_{\rm W}$ and w on $\Delta H_{\rm m}$. Both $\alpha_{\rm W}$ and w decrease with increasing negative $\Delta H_{\rm m}$, which indicates the chemical effect on the local structure in MGs. Especially for Zr–Cu (~ -25 kJ/mol) and Zr–Ni (~ -53 kJ/mol) systems, it has been widely accepted that the dominant SRO in Zr–Cu is of topology with icosahedral symmetry (Voronoi index $\langle 0,0,12,0\rangle$),[26,31,32] in contrast, the ISRO degree is much lower in Zr–Ni MG.[27,33–35] The interaction is much stronger for Zr–Ni with large negative $\Delta H_{\rm m}$, and the polyhedra around Ni have lower CNs less than 10,[34] which increases the energy of Ni-centered icosahedral clusters with CN of 12. Therefore, large negative $\Delta H_{\rm m}$ leads to a tendency for unlike-neighbor bonds and relatively fewer icosahedral clusters. Nevertheless, the population of icosahedral clusters is also controlled by the radius ratio of component elements.[23,28] The center atom of a perfect icosahedral cluster is slightly smaller, and the radius ratio is 0.902. When the atomic radius ratio deviates from 0.902, the population of other polyhedra types increases, such as the Frank–Kasper type, the BSAP type, and the TTP type.[23] Furthermore, the electronic configuration of component elements has an important influence on the local structure, such as the covalent-like bonding in the systems containing metalloids[36] and bond shortening due to the special interaction between the outer electrons of the component elements.[28,37] Thus to predict the relative population of icosahedral clusters, the multiple factors should be considered but not the single factor of $\Delta H_{\rm m}$ for different MG systems.

Table 2. Warren parameter $\alpha_{\rm W}$ and relative abundance w of the selected main bonded pairs $(1551+1541+1431)$ for systems with different $\Delta H_{\rm m}$.

System	$\Delta H_{\rm m}$	$\alpha_{\rm W}$	w	Reference
$Zr_{60}Al_{40}$	–75	–0.106	58%	[26]
Zr_2Ni	–53	–0.089	57%	[28]
Zr_2Cu	–25	–0.053	63%	[28]
Zr_2Ag	–24	0.022	64%	[28]
$Ca_{70}Mg_{30}$	–6	0.084	76%	[29]
$Mg_{50}Al_{50}$	–2	0.106	74%	[30]

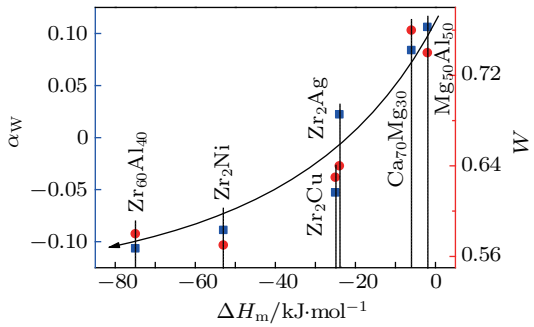


Fig. 3. (color online) The trend of $\alpha_{\rm W}$ and w for systems with different $\Delta H_{\rm m}$.

β-relaxation and SRO in MGs both have a close relation with crystallization and mechanical properties,[11,12,38,39] which implies that β-relaxation may be related with SRO. Plenty of researches confirm the correlation between the non-affine displacement and the local structures. It was indicated that in Cu–Zr MG the non-pentagonal regions, which are sources for the generation of high free volume structures, preferentially undergo irreversible atomic rearrangements in the early stage of the deformation process. Non-pentagonal regions are easily deformed plastically because of high energy and low dynamic stability.[40] The degree of local fivefold symmetry (LFFS) was used as the structural indicator to predict plastic deformation of local structures and the plastic events propagate in regions with a lower degree of LFFS.[41] Therefore, the perfect and distorted icosahedral clusters with high fivefold symmetry have relative high dynamic stability. Recently, it was found that β-relaxation is related to the cooperative motion of particles chains by the MD simulations.[42] In the simulations, they pinned randomly a small fraction of the particles to the coordinate frame, as a result the β-relaxation was suppressed more sufficiently with increasing the pinned particle fraction. These pinned particles were only allowed to participate in the affine transformation, which is similar to the icosahedral clusters with high dynamic stability in MGs. These findings indicate that icosahedral clusters could suppress the motion of β-relaxation. Our MD simulations have revealed the atomic fraction associated with icosahedral configuration for the Zr-based MGs. The fraction of the 20 most abundant Voronoi indices that appear in the Zr-based MGs is presented in Fig. 4(a), where the Voronoi index $\langle 0,0,12,0\rangle$ associated with icosahedral clusters is 1.09% and 0.76% for $Zr_{66.6}Cu_{33.4}$ and $Zr_{66.6}Ni_{33.4}$, respectively. The population of the Voronoi indices related to distorted icosahedral structures, such as $\langle 0,1,10,2\rangle$ and $\langle 0,2,8,2\rangle$, for $Zr_{66.6}Cu_{33.4}$ are also larger than those for $Zr_{66.6}Ni_{33.4}$, which

basically agrees with the previous results.[28,43] Figures 4(b) and 4(c) show the distribution of the icosahedral clusters for $Zr_{66.6}Cu_{33.4}$ and $Zr_{66.6}Ni_{33.4}$ respectively, which indicates that $Zr_{66.6}Cu_{33.4}$ has more icosahedral clusters and the distribution is homogenous without significant aggregation for both compositions. Besides, we also performed MD simulations for Ti-based MGs, and obtained similar results. Correspondingly, $Zr_{66.6}Cu_{33.4}$ and $Ti_{60}Cu_{40}$ MGs have less pronounced β-relaxations and higher $E_{r\beta}$ compared with that of $Zr_{66.6}Ni_{33.4}$ and $Ti_{60}Ni_{40}$, respectively. These experimental and simulation results suggest the pinning effect of dynamic stable clusters on the motion of β-relaxation.

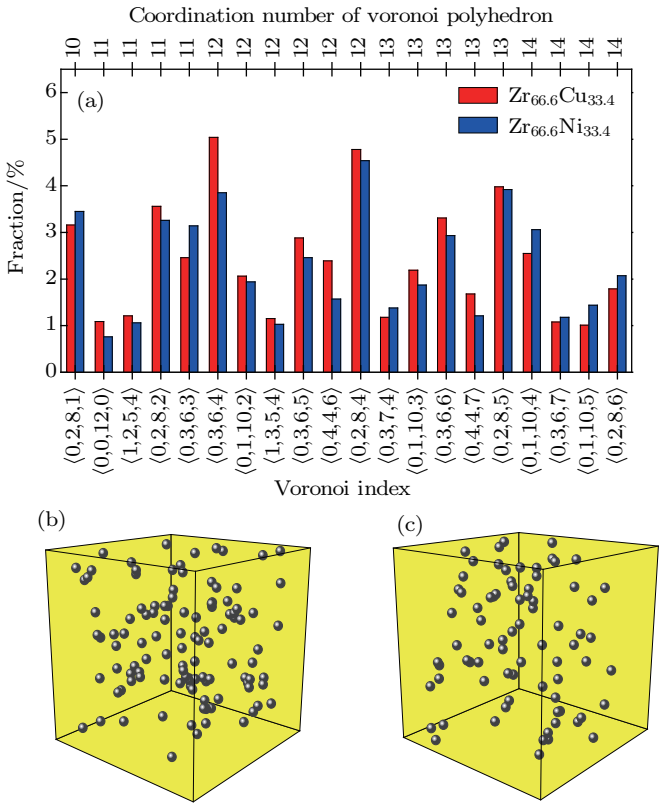


Fig. 4. (color online) (a) The occurrences of the 20 most popular Voronoi indices for $Zr_{66.6}(Cu/Ni)_{33.4}$ in a glassy state. The indices are sorted by the coordination number as shown on the top of the figure. In the icosahedral cluster distribution in $Zr_{66.6}Cu_{33.4}$ (b) and $Zr_{66.6}Ni_{33.4}$ (c), the dark gray balls represent the icosahedral clusters.

4. Conclusion

In summary, the chemical effect on β-relaxation is experimentally verified in three systems. The substitution of Ni and Cu shows that the MG with larger negative ΔH_m has more significant β-relaxation in each system. The MD simulations show that more icosahedral clusters exist in MGs with less negative ΔH_m. The icosahedral clusters make β-relaxation activation energy increase and pin up the atomic motion of β-relaxation.

Acknowledgement

Thanks to Prof. W. H. Wang for our discussion of the experiment results.

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