Dynamic mechanical relaxation behavior of binary metallic glasses

M.N. Liu[a], Q. Hao[a], J. Dong[b], B.A. Sun[b,c], S.D. Feng[d], D. Crespo[e], J.C. Qiao[a,*]

[a]*School of Mechanics, Civil Engineering and Architecture, Northwestern Polytechnical University, Xi'an 710072, China*

[b]*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

[c]*Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China*

[d]*State Key Laboratory of Metastable Materials Science and Technology, Yanshan University, Qinhuangdao 066004, China*

[e]*Departament de Física, Barcelona Research Center in Multiscale Science and Technology & Institut de Tècniques Energètiques, Universitat Politècnica de Catalunya, 08930-Barcelona, Spain*

*Corresponding author:

Dr. J.C. Qiao E-mail address: qjczy@nwpu.edu.cn

Abstract: Secondary β relaxation is a fundamental issue to understand the diffusion, plasticity and glass transition behavior of metallic glasses. Binary metallic glasses provide a simple glassy system to probe the dynamic mechanical processes. In the current research, dynamic mechanical behavior of the typical binary metallic glasses was investigated by dynamic mechanical analysis (DMA). The dynamic mechanical relaxation behavior of $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) and Cu_xZr_{100-x} (x= 30, 50, 56, 61.8 and 64) binary metallic glass system was studied. It is found that the β relaxation becomes pronounced with the increase of copper, which is analyzed in the framework of the principle of mixing enthalpy. However, the β relaxation of CuZr or CuTi binary metallic glasses is much more modest than that of $Y_{65}Co_{35}$, $Dy_{65}Co_{35}$ and $Y_{67}Cu_{33}$.

Keywords: Binary metallic glass; Mechanical relaxation; Secondary relaxation; Structural heterogeneity

1. Introduction and background

Metallic glasses (MGs) have drawn exhaustive interests in the past decades due to their unique physical and mechanical properties, such as high strength [1], excellent corrosion resistance [2], super thermo-plastic ability [3, 4] and superior catalytic capabilities [5]. These physical or mechanical properties are closely related to their heterogeneous microstructure. Different than conventional crystalline solids, there are no recognizable defects (i.e., dislocations and grain boundaries) in metallic glasses. It is very important to establish the correlation between the microstructure and mechanical/physical properties of MGs [6, 7]. The link between mechanical relaxation processes, in particular, the secondary β relaxation and mechanical properties (i.e. plasticity) has been established [8-10]. Therefore, MGs are considered to be the model system for probing the β relaxation of glassy materials.

It is believed that glassy materials experience two relaxation processes. The main α relaxation emerges at high temperature and becomes "frozen" when the temperature lies below the glass transition temperature T_g. On the other hand, the secondary β relaxation remains active below the T_g, decoupled from the main α relaxation. The primary α relaxation relies on the cooperative movement of atoms, while the secondary β relaxation is associated to the local motion of atoms. In essence, the main α relaxation of non-crystalline solids is linked to the dynamic glass transition process. According to the recent investigations [11-15], this is well in accordance with the cooperative shear model based on the concept of a potential energy landscape. While the slow β process corresponds to individual shear transformation zone (STZ) transitions, the α relaxation can be related to their percolation, leading to macroscopic plastic flow. Nevertheless, of more significant importance is the identification of the universal fast β′ secondary relaxation in addition to the slow β and α relaxation for MGs [15, 16]. Mechanical spectroscopy data shows that the two secondary relaxations in MGs have quite different dynamic characteristics [13, 17]. The peak of the loss modulus of fast β′ relaxation has much lower magnitude but larger broadness than that of the slow β relaxation. In the past two decades, a large corpus of experimental data has disclosed that β relaxation is key to describe the properties of glass-forming materials [18]. While there is only one

relaxation process at high temperature for liquid, the secondary relaxation process in the glass is commonly associated with the breakdown of the Stokes-Einstein relation. A correlation between atomic mobility and the local structure has been proved in supercooled liquids [19].

Binary metallic glasses are excellent model alloys to study the mechanical relaxations. Theoretical research [20] and simulations [21] have proven that the β relaxation is closely linked to the diffusion of atoms in MGs. It has been proved that the activation energy of the β relaxation E_β is related to the diffusion activation energy in the Fe/Co/Ni MG system [22]. In addition, it was further confirmed that the self-diffusion of Fe/Co/Ni plays an important role in the β relaxation process. Previous investigations have proved that the glass transition (α relaxation) behavior is mainly dominated by the slowing down of diffusion of large particles, while the diffusion of small particles remains in a deep glassy state [23-25]. These results rise one unsolved question: What is the link between the diffusion and the relaxation modes in glassy materials?

In the current work, the mechanical relaxation behavior of the binary MGs, i.e., Cu_xZr_{100-x} (x= 30, 50, 56, 61.8 and 64), $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) and rare earth based (i.e. $Y_{65}Co_{35}$, $Dy_{65}Co_{35}$ and $Y_{67}Cu_{33}$) was probed by dynamic mechanical analysis (DMA). The results suggested that the slow β relaxation . The current research provides a new way to understand the origin of the slow β relaxation.

2. Experimental procedure

The ingots of the model alloys were prepared by arc melting in a Ti-gettered Argon atmosphere. The ingots were re-melted at least 5 times to keep the chemical homogeneity. Ribbons were obtained by single-roll melt-spinning in an Argon atmosphere with a tangential velocity of about 40 m/s. The thickness of the ribbons is around 30-40 μm. Dynamical mechanical behavior of the MGs was carried out on a commercial DMA (TA Q800) in film tensile mode in a nitrogen-flushed atmosphere.

3. Results and discussions

Fig.1 (a) shows the temperature dependence of the normalized storage modulus (E′/Eu) and the loss modulus (E″/Eu) of a $Cu_{50}Zr_{50}$ binary MG (driving frequency is 1 Hz, the heating rate is 3 K/min). E_u corresponds to the value of unrelaxed modulus at ambient temperature. It can be seen that in the low temperature range the model alloy is staying in amorphous state. The storage modulus E′ remains approximately constant with a high value, while the loss modulus E″ is very low. Consequently, elastic deformation dominates the mechanical behavior in this temperature domain. By increasing the temperature, the storage modulus decreases drastically while the loss modulus reaches its maximum value around 695 K, which is associated with the primary α relaxation. It is well recognized that the main α relaxation corresponds to the evolution from the out-of-equilibrium state to the supercooled liquid in glassy materials [17]. When the temperature is above 700 K, both E′ and E″ increase exceedingly as temperature increases, as a consequence of crystallization. It should be stressed that pronounced secondary β relaxation has not been observed in the $Cu_{50}Zr_{50}$ binary MG.

It is well known that the secondary β relaxation is linked to the localized movement of atoms while *α* relaxation is related to cooperative motion of atoms [26-28], and appears in form of shoulder or excess wing in the dynamic mechanical-loss spectra [29]. **Fig.1 (b)** presents the evolution of the normalized loss modulus with temperature in typical Cu_xZr_{100-x} (x=30, 50, 56, 61.8 and 64) binary MGs. The data are normalized to the values of temperature and loss modulus at the peak of α relaxation, namely T_α and E''_{max}. As already found in the literature, the secondary β relaxation in CuZr allows displays as an excess wing rather than a discernible shoulder or peak. Interestingly, the secondary relaxation process is sensitive to the chemical composition. By increasing the content of zirconium, the excess wing due to β relaxation of the binary MGs was almost removed.

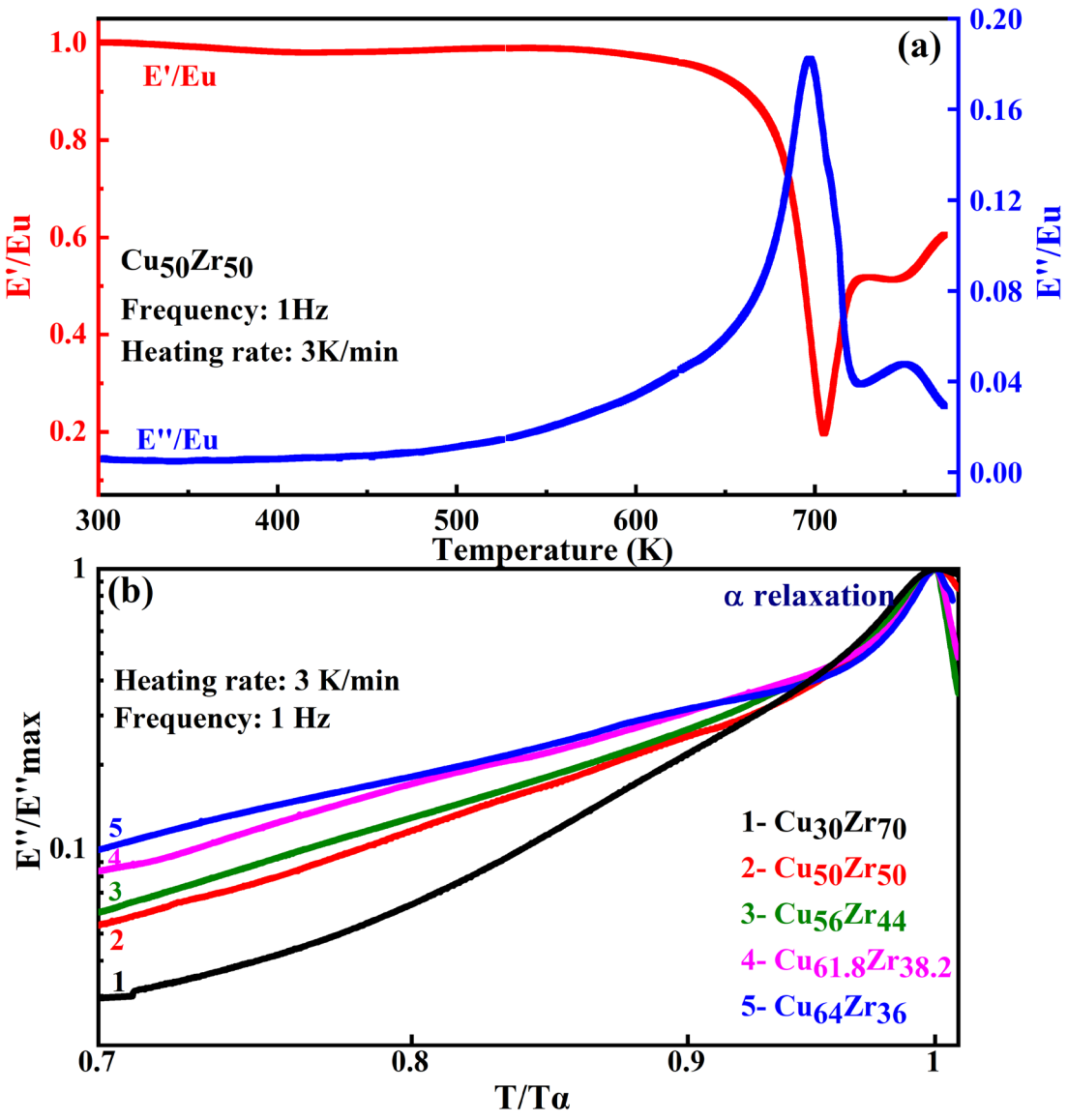

Fig.1 (a) Evolution of normalized storage modulus and loss modulus with the temperature of $Cu_{50}Zr_{50}$ metallic glass (heating rate is 3 K/min; driving frequency is 1 Hz). **(b)** The normalized loss modulus E''/E''_{max} as a function of temperature in Cu_xZr_{100-x} (x= 30, 50, 56, 61.8 and 64) MGs. Measurement frequency is 1 Hz and heating rate is 3 K/min. E''_{max} is the peak temperature of the main α relaxation.

In order to further understand the effect of the chemical composition in binary

MGs, mechanical relaxation of $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) binary MGs was investigated. **Fig.2** shows the temperature dependence of the normalized loss modulus with temperature of $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) binary MGs. According to the mechanical-loss spectra, the slow β relaxation was suppressed by increasing the content of titanium.

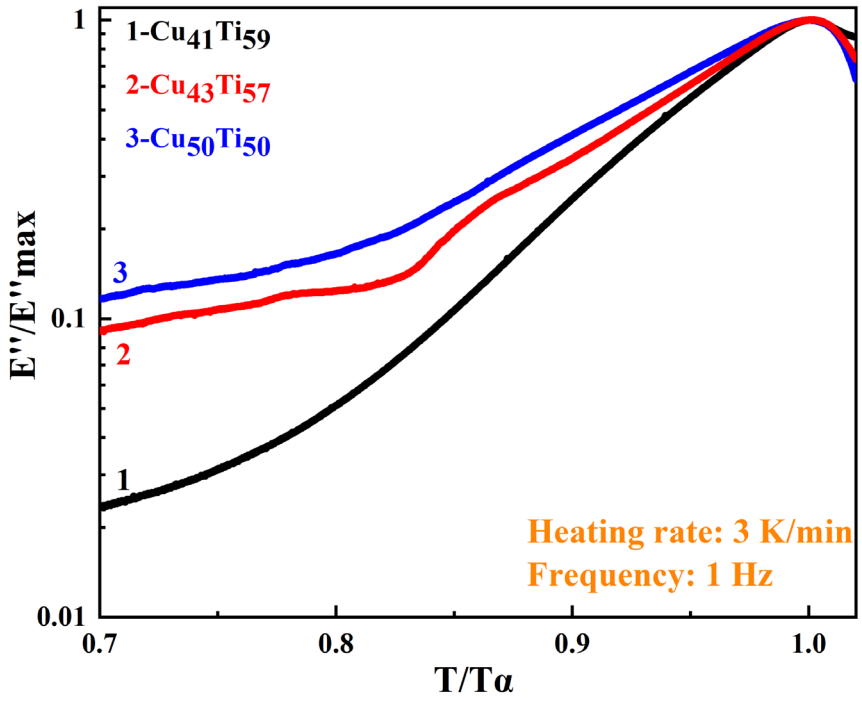

Fig.2 Normalized loss modulus E''/E''_{max} as a function of temperature in $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) MGs. Testing frequency is 1 Hz and heating rate is 3 K/min.

The behavior of β relaxation in MGs can be explained in the framework of mixing enthalpy principle [30]. A negative enthalpy of mixing for a pair of atoms exhibits an evident β relaxation, while a positive or a significantly different mixing enthalpy will suppress β relaxation. The average mixing heat ΔH^{chem} is given by

$$\Delta H_{mix} = 4\sum \Delta H_{ABmix} c_A c_B \quad (1)$$

where ΔH_{ABmix} represents the enthalpy of mixing between elements A and B. c_A, c_B indicate the molar fractions of elements A and B in the alloy, respectively [31]. The data of mixing enthalpy of constituent atomic pairs are derived from Ref. [32]. **Fig.3** plots

the mean chemical affinity ΔH_{mix} of the $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) MGs. Cu-Ti atomic pair has a relatively negative enthalpy of mixing of -9 kJ/mol. In the case of the $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) MGs, $Cu_{50}Ti_{50}$ has the largest negative enthalpy of mixing, which corresponds to the most prominent β relaxation peak. The experiments in the current research are in good agreement with the prediction of mixing enthalpy principle.

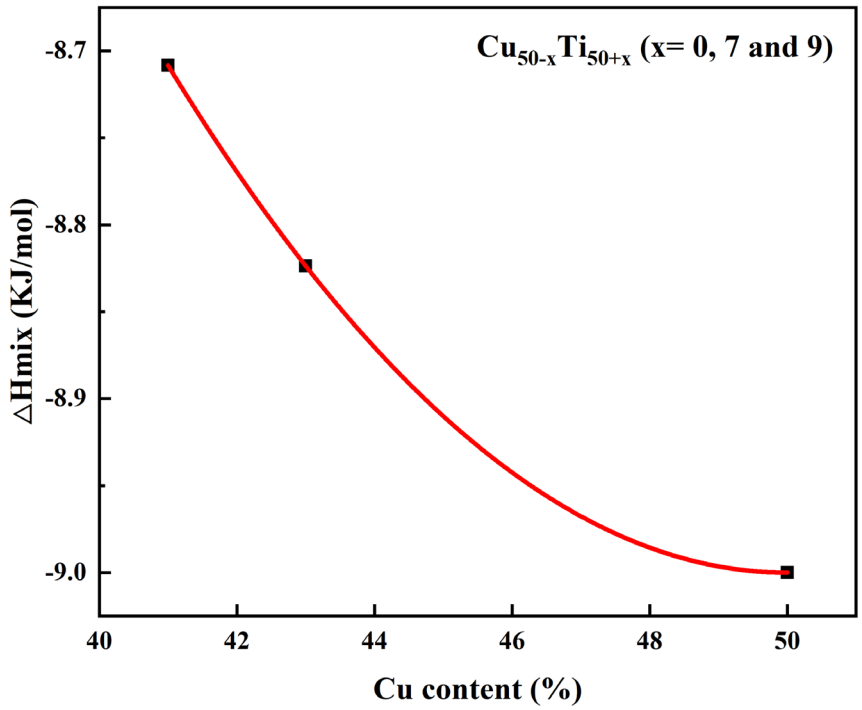

Fig.3 Mixing enthalpy ΔH_{mix} for the $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) MGs.

Recent investigation on a LaNiAl MG suggests that the slow β relaxation is sensitive to the chemical compositions [33]. It has been found that binary La-Ni MGs shows an discting slow β relaxation peak while La-Al and La-Cu binary MGs show excess wing process. Since both La and Y are rare earth (RE) elements, whether the unusual dynamics are general to similar MGs based on other RE elements. The question which remains open is whether the pronounced β relaxation peak is general for RE-Ni/Co binary MGs. The binary RE-Ni/Co MGs may serve as model materials to investigate the β relaxation in MGs due to its simple composition. In general, only a

few types of MGs exhibit significant β relaxation revealed by DMA measurements, such as La- [34] and Nd-based [28] MGs. For other MGs, i.e. Zr-[35], Cu-[36], Ti-[37], Mg-[38] and Ce-based [39] MGs show an excess wing in their loss modulus. Most binary MGs exhibit weak evidence of β relaxation in their dynamic mechanical spectroscopy. In contrast to other MGs, $Co_{35}Dy_{65}$ and $Co_{35}Y_{65}$ show a distinct β relaxation peak in the mechanical relaxation measurements.

As shown in **Fig.4**, the temperature dependence loss modulus E″ of a $Y_{67}Cu_{33}$ binary MG exhibits two distinct peaks : an α relaxation peak of E″ located at about 600±5 K and a pronounced β relaxation peak situated at about 420±5 K. The β relaxation peak of the loss modulus is associated to the heterogeneity of microstructure or the "defects" of MGs. The inset of **Fig.4** compares the β relaxation behavior of $Dy_{65}Co_{35}$, $Y_{65}Co_{35}$ and $Y_{67}Cu_{33}$ binary MGs. Only the curves with the testing frequency of 1Hz are exposed. The loss modulus E″ curves have been scaled for comparison. The temperature is reduced by the peak temperature of α relaxation T_α and E″ is normalized by E''_α. From the normalized plot, it can be realized that the β relaxation of $Y_{65}Co_{35}$ is stronger than that of $Y_{67}Cu_{33}$. In addition, the intensity β relaxation of $Dy_{65}Co_{35}$ is the largest, which further confirms that the magnetic element Co can facilitate pronounced β relaxation.

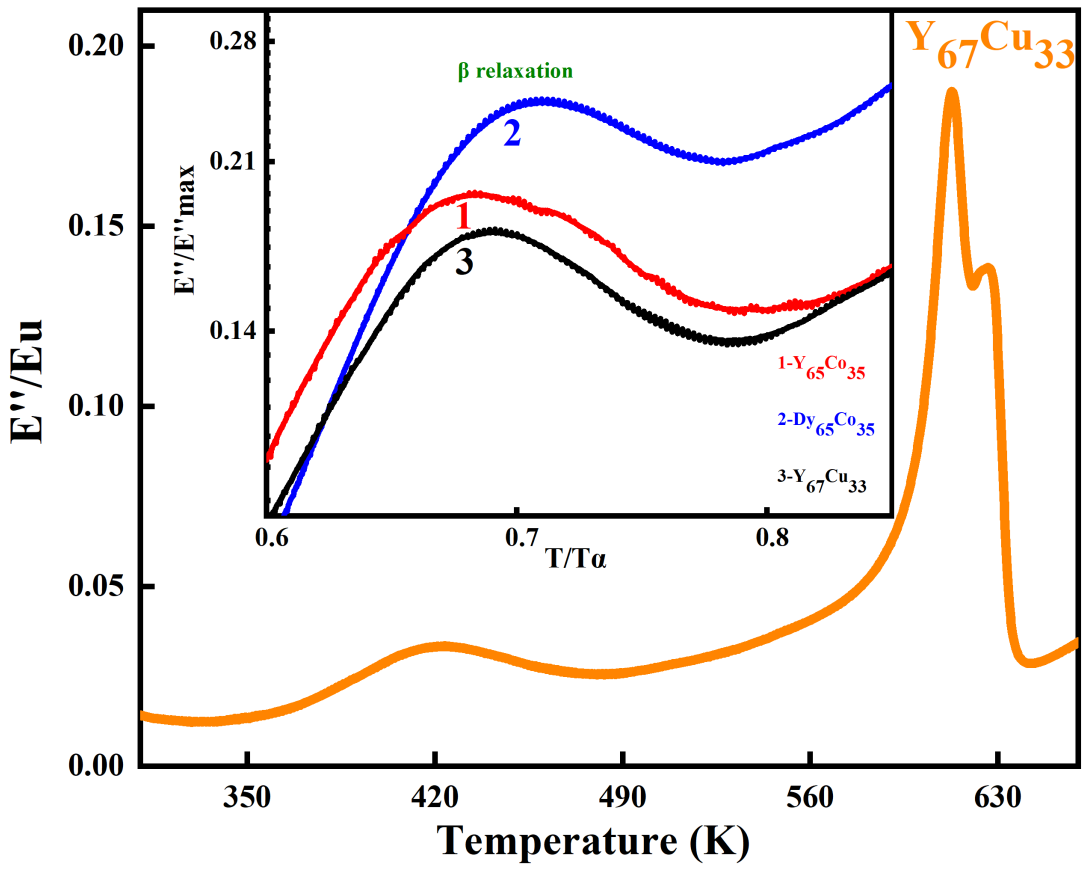

Fig.4 Temperature dependence of the loss modulus E″ of $Y_{67}Cu_{33}$ at 1 Hz and a heating rate is 3 K/min. The inset shows the temperature dependence of normalized loss modulus in $Y_{65}Co_{35}$, $Dy_{65}Co_{35}$ and $Y_{67}Cu_{33}$ binary MGs (heating rate is 3 K/min and driving frequency is 1 Hz).

Fig.5 illustrates the temperature dependence of E″ at different frequencies (0.5, 1, 2, 4 or 8 Hz) of $Dy_{65}Co_{35}$ and $Y_{65}Co_{35}$ binary MGs. The T_β shifts to higher temperatures by increasing the frequency. The activation energy for β relaxation (E_β) is determined by the $\ln(f)$ vs $1000/T_\beta$ plot. It can be fitted to an Arrhenius equation [40]:

$$f = f_\infty \exp\left(-\frac{E_\beta}{RT}\right)$$

where f_∞ is the Arrhenius pre-factor, E_β is the activation energy of the β relaxation and R is the gas constant.

The inset of **Fig.5 (a)** shows the Arrhenius fit of the measured data of $Dy_{65}Co_{35}$ MG. The fitted activation energy E_β is 110 kJ/mol, or $E_\beta = 23RT_g$ (R is the gas constant). In the same way, the inset of **Fig.5 (b)** shows the Arrhenius fit of the measured data of $Y_{65}Co_{35}$MGs. The fitted activation energy E_β is in this case 107 kJ/mol, or $E_\beta = 23RT_g$. Bot are is in good agreement with the relationship reported between the E_β and T_g in different MGs [9].

Compared to $Dy_{65}Co_{35}$ MG, it is worth noting that $Y_{65}Co_{35}$ MG presents an evident slow β relaxation at low temperature. Nevertheless, $Y_{67}Cu_{33}$ MG don't show an apparent slow β relaxation. In fact, whether β relaxation is obvious will affect the mechanical properties of metallic glass. At room temperature, due to MGs are staying in the metastable state, namely, atoms are in "frozen" state. When the temperature increases (but still below the glass transition temperature T_g), the "frozen" atoms soak up amounts of energy. The origin of β relaxation is not clear, but it is generally accepted that it is associated with string-like atomic rearrangements [41].

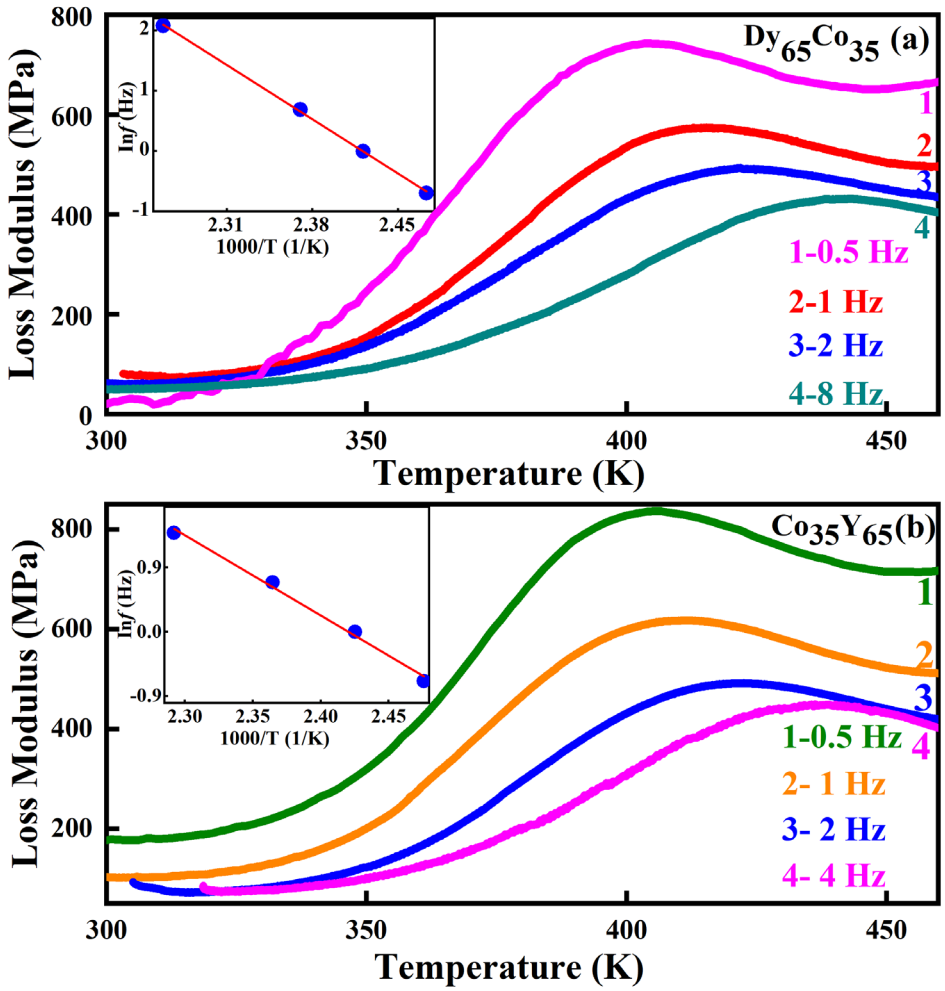

Fig.5 (a) Temperature dependence of loss modulus of $Dy_{65}Co_{35}$ metallic glass measured at various frequencies. Inset is the Arrhenius plot of the frequency *vs* peak temperature of the β relaxation. **(b)** Temperature dependence of loss modulus of $Y_{65}Co_{35}$ metallic glass measured at different frequencies. Inset is the Arrhenius plot of the frequency *vs* peak temperature of the β relaxation.

Fig.6 shows the correaltion between the activation energy of β relaxation and the glass transition temperature T_g in typical MGs. It has been proven that activation energy of the slow β relaxation in glassy materials obeys the empirical relation $E_\beta=(26\pm4)RT_g$ (R is gas constant)[9],. As T_g is the characteristic temperature of α relaxation, the relationship between E_β and T_g shows that β relaxation and α relaxation are tightly connected. In other words, α relaxation may consist of many percolating β relaxation

processes. In the case of $La_{30}Ce_{30}Al_{15}Co_{25}$ [42] metallic glass, the activation energy of α relaxation and β relaxation is 3.61 eV and 0.86 eV, respectively.It should be noted that the apparent activation energy of α relaxation is approximately four times that of β relaxation. The correlation between the activation energy of $Dy_{65}Co_{35}$/$Y_{65}Co_{35}$ metallic glasses and the glass transition temperature T_g obeys with the $24RT_g$ relationship.

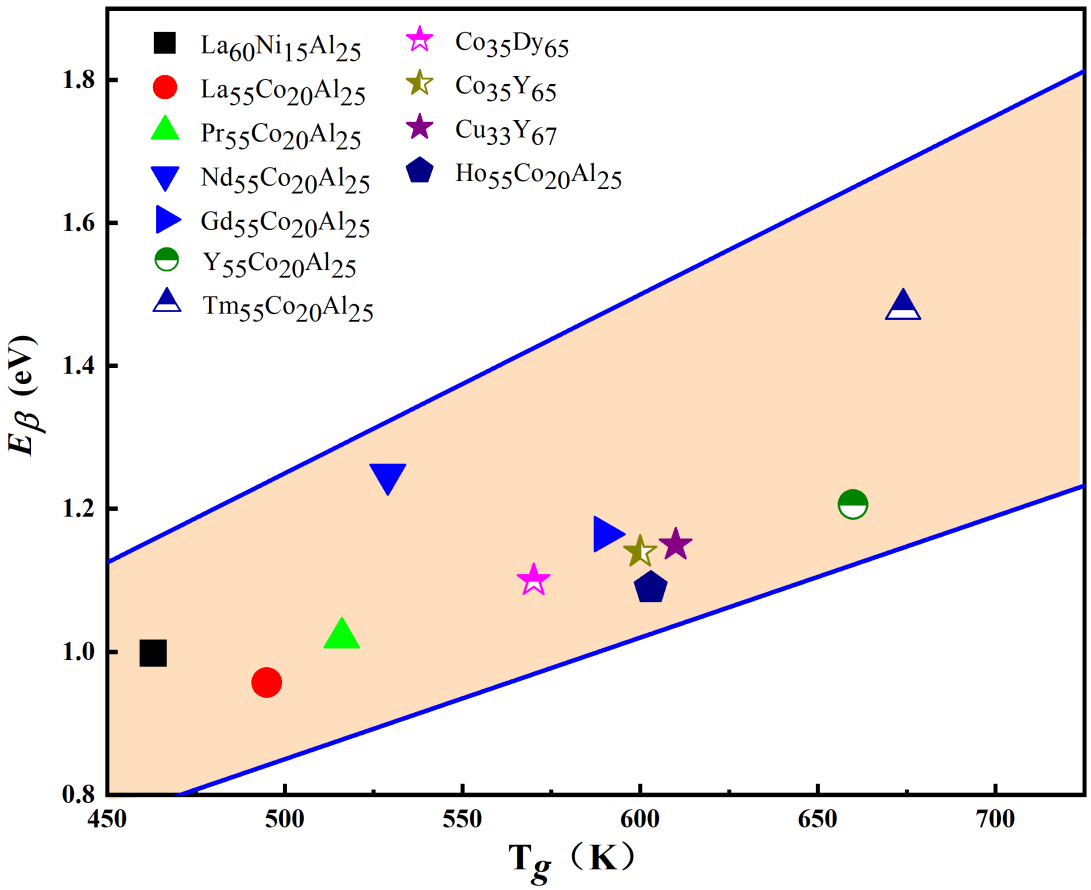

Fig.6 Correaltion between the activation energy of slow β relaxation $E_β$ with the glass transition temperature T_g in typical MGs. The data are obtained from Ref. [43].

4. Conclusion

In summary, mechanical relaxation processes of binary MGs were probed by mechanical spectroscopy. β relaxation of the Cu_xZr_{100-x} (x= 30, 50, 56, 61.8 and 64) binary MG systems was suspended by increasing the content of zirconium. In addition, in the case of the $Cu_{50-x}Ti_{50+x}$ (x= 0, 7 and 9) binary MGs, the slow β relaxation is

suppressed by increasing the content of titanium. Finally, rare earth-based binary MGs, i.e, $Dy_{65}Co_{35}$, $Y_{65}Co_{35}$ ad $Y_{67}Cu_{33}$, show an evident slow β relaxation process. The activation energy of the β relaxation obeys the empirical relation: $E_{\beta}\sim 26RT_g$. The β relaxation process is sensitive to the chemical composition of MGs. These binary MGs provide an ideal model alloy to sheld light on the fundamental issues in MGs, i.e. relaxation processes, diffusiton behavior and mechanical properties.

Acknowledgements

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