

# A Diagram for Glass Transition and Plastic Deformation in Model Metallic Glasses



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In this paper, we report a strain rate related glass transition in model SrCaYbMg(Li)Zn(Cu) metallic glasses at room temperature. A critical strain rate, equivalent to glass transition temperature, is found for the strain rate induced glassy state to liquid-like viscoplastic state translation. The results show that the observation time, equivalent to temperature and stress, is a key parameter for the transition between the glass and supercooled liquid states. A three-dimension glass transition diagram involved in time, temperature and stress in metallic glasses is established.

**KEY WORDS:** Homogeneous plastic flow; Glass transition; Metallic glass

## 1. Introduction

Metallic glasses (MGs) with atomic randomly packing structure, excellent glass-forming ability and wide supercooled liquid temperature region are model system for studying the fundamental issues in glass field<sup>[1–6]</sup>. Glasses are regarded as liquids that have lost their flow ability. Generally, it is recognized that the glass transitions between supercooled liquid and glassy states in metallic glasses (MGs) are induced by thermalization or by applied stress<sup>[7–13]</sup>. Through simulation, Guan et al.<sup>[12]</sup> quantified the intimate coupling of temperature and shear stress in glass transition. In fact, the glass transition is observation time dependent<sup>[9]</sup>. For polymer glasses, their responses to stress can be observed both within a short time at high temperature and a sufficient long time at low temperatures, which is time-temperature superposition principle<sup>[14]</sup>. The physical similarity among MGs, colloidal and polymer glasses suggests that the glass transition in MGs could have the similar response to the key parameters of temperature ( $T$ ), stress as well as the observation time.

However, little work has been done on the effects of observation time on glass transition in MGs. This is because the MGs normally have high flow energy barrier below glass transition temperature ( $T_g$ ), and the homogeneous deformation of MGs

generally is only observed at high  $T$  or in inaccessible long time scale under stress at low  $T$ <sup>[15,16]</sup>. For example, for Zr-based MG with a flow activation energy  $\sim 1.3$  eV, the room temperature (RT) uniaxial compression at 80% of its yield stress for 5 h only leads to homogeneous flow with irreversible strain of  $\sim 2.0 \times 10^{-4}$ <sup>[17]</sup>. The long time scale and very small homogeneous flow strain at low  $T$  make the study of the effects of strain rate and observation time on glass transition very difficult. Recently, some MGs such as Ce-, La- and Sr-based with low  $T_g$  have been developed<sup>[18–21]</sup>. Remarkable liquid-like homogeneous flow near room temperature and under relatively high strain rates can be realized in the Sr-based glasses due to their very low flow activation energy<sup>[18,22]</sup>. Importantly, the supercooled liquid state of the MGs can be realized solely by the homogeneous mechanical behavior, which provides a model system to study the roles of strain rate or observation time in glass transition, and to test the concept of glass transition diagram in MGs.

In this work, we report that the glass to supercooled liquid state transition can be solely induced by the strain rate  $\dot{\gamma}$  in the model SrCaYbMg(Li)Zn(Cu) MG systems. We examine the role of  $\dot{\gamma}$ , which is equivalent to the observation time, in the glass transition induced by stress of the MGs, and a time involved glass transition diagram is obtained. The results show that the observation time, temperature and stress can be incorporated into a generalized description of transition between glass and supercooled liquid states in MGs.

## 2. Experiments

Cylindrical specimens of Sr<sub>20</sub>Ca<sub>20</sub>Yb<sub>20</sub>Mg<sub>20</sub>Zn<sub>20</sub>, Sr<sub>20</sub>Ca<sub>20</sub>Yb<sub>20</sub>Mg<sub>20</sub>Zn<sub>10</sub>Cu<sub>10</sub>, Sr<sub>20</sub>Ca<sub>20</sub>Yb<sub>20</sub>(Li<sub>0.55</sub>Mg<sub>0.45</sub>)<sub>20</sub>Zn<sub>20</sub> MGs of

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**Table 1** Values of  $T_g$ , shear modulus  $G$ , compression yield strength  $\sigma_y$ , the maximum flow stress  $\sigma_{\max}$  and steady flow stress  $\sigma_{\text{flow}}$  at the critical strain rate, the experimental critical strain rate  $\dot{\gamma}_g$  and the calculated critical strain rate  $\dot{\gamma}_g^{\text{cal}}$  of MGs

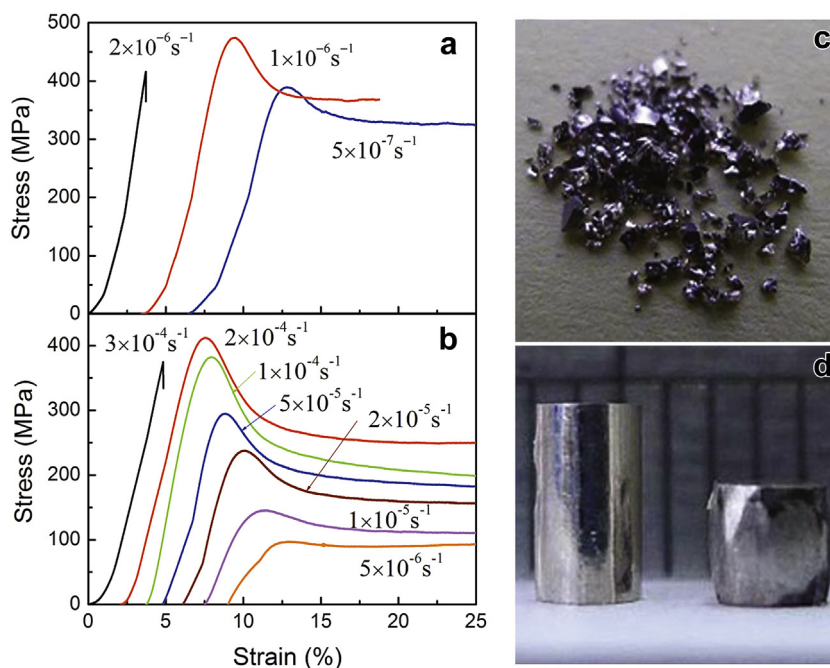
	$\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Mg}_{20}\text{Zn}_{20}$	$\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Mg}_{20}\text{Zn}_{10}\text{Cu}_{10}$	$\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}\text{Zn}_{20}$
$T_g$ (K)	353	351	319
$G$ (MPa)	8.89	9.47	6.28
$\sigma_y$ (MPa)	452.1	433.6	375.1
$\sigma_{\max}$ (MPa)	473.6	450.3	411.9
$\sigma_{\text{flow}}$ (MPa)	366.9	416.2	249.3
$\dot{\gamma}_g$ ( $\text{s}^{-1}$ )	$1 \times 10^{-6}$	$5 \times 10^{-7}$	$2 \times 10^{-4}$
$\dot{\gamma}_g^{\text{cal}}$ ( $\text{s}^{-1}$ )	$1.51 \times 10^{-6}$	$1.45 \times 10^{-6}$	$1.25 \times 10^{-4}$

2 mm in diameter were prepared by copper mold casting method<sup>[22]</sup>. The  $T_g$  and shear modulus  $G$  of the MGs were presented in Table 1. Compression tests at RT were conducted on these MGs with a length-to-diameter ratio of 2 at different applied strain rates on an Instron 3384 machine (Norwood, MA). The temperature of the compression tests was controlled by a thermocouple and the tests were performed in pure Ar atmosphere to avoid the oxidation. The X-ray diffraction results showed that the temperature in the range of 310–350 K for compression tests did not induce the crystallization. The scanning electron microscopy (SEM) was conducted in a Philips XL30 instrument. According to the shear cooperative model<sup>[23]</sup>, the barrier energy of plastic flow units  $W_{\text{STZ}}$  (also termed as shear transformation zones (STZs)) in MGs can be estimated by<sup>[24]</sup>  $W_{\text{STZ}} \propto 0.4GV_m$ , where  $V_m$  is average molar volume. These MGs have ultra low  $G$  (see Table 1), and their flow barrier energy  $W_{\text{STZ}}$  is estimated to be 0.79, 0.75, and 0.52 eV, respectively. Clearly, these MGs have much lower activation energy of flow units compared with that of conventional MGs<sup>[24]</sup> and the activation of flow units is then much easier. We then could realize the homogenous flow or glass to supercooled liquid transition near RT in the MGs by stress in reasonable observation

time scale, and characterize the effect of time on glass transition through their homogeneous mechanical behavior.

### 3. Results and Discussion

Fig. 1 shows the compressive stress–strain curves at different applied strain rates for as-cast rods of  $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Mg}_{20}\text{Zn}_{20}$  (Fig. 1(a)) and  $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}\text{Zn}_{20}$  (Fig. 1(b)). For both of the MGs, at RT and above a critical strain rate  $\dot{\gamma}_g$ , the true stress–strain curves exhibit an elastic loading region followed by catastrophic failure into small pieces without any plastic deformation (see Fig. 1(c)). The fracture approaches the ideal brittle behavior<sup>[25]</sup>, which indicates that at RT the MG is in glassy state. However, below a critical strain rate  $\dot{\gamma}_g$ , remarkably, the MGs displays a stress overshoot which is often observed in the supercooled liquid state of MGs at high  $T$  (e.g. for Zr-based MGs, the superplastic flow occurs above 600 K<sup>[15]</sup>). The stress overshoot is due to the structural evolution (i.e. accumulation of free volume) accompanied by softening<sup>[25,26]</sup>. Due to the intrinsic relaxation, the stress attains a steady state after the overshoot (Fig. 1(a and b)) which is a typical feature of homogeneous flow in glass-forming liquid. As shown in Fig. 1(d), the



**Fig. 1** Compressive stress–strain curves at different strain rates of (a)  $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Mg}_{20}\text{Zn}_{20}$  and (b)  $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}(\text{Li}_{0.55}\text{Mg}_{0.45})_{20}\text{Zn}_{20}$  MGs; (c) pictures of brittle behavior above  $\dot{\gamma}_g$  and (d) homogeneous deformation behaviors (about 50% deformed) below the critical strain rate for  $\text{Sr}_{20}\text{Ca}_{20}\text{Yb}_{20}\text{Mg}_{20}\text{Zn}_{20}$  MG.

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