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Viscosity anomaly of a metallic glass-forming liquid under high pressure

Qifan Wang,[a,b] Hongbo Lou,[b] Yoshio Kono,[c,d] Daijo Ikuta,[d,e] Zhidan Zeng,[b] Qiaoshi Zeng[a,b,f,*]

[a.] *Jiangsu Key Laboratory of Advanced Metallic Materials, School of Materials Science and Engineering, Southeast University, Nanjing 211189, People's Republic of China.*

[b.] *Center for High Pressure Science and Technology Advanced Research, Shanghai 201203, People's Republic of China.*

[c.] *Geodynamics Research Center, Ehime University, 2-5 Bunkyo-cho, Matsuyama, 790-8577, Japan.*

[d.] *Geophysical Laboratory, Carnegie Institution of Washington, 9700 South Cass Avenue, Argonne, Illinois 60439, USA*

[e.] *Institute for Planetary Materials, Okayama University, 827 Yamada, Misasa, Tottori 682-0193, Japan.*

[f.] *Shanghai Key Laboratory of Material Frontiers Research in Extreme Environments (MFree), Shanghai Advanced Research in Physical Sciences (SHARPS), Pudong, Shanghai 201203, P.R. China*

**Correspondence and requests for materials should be addressed to Q.S.Z. (email: zengqs@sharps.ac.cn)*

Abstract

Viscosity, as a critical property closely associated with the glass-forming ability of a liquid, has been extensively studied with varying temperatures. However, its pressure dependence has not been well explored yet due to experimental difficulties. Here, we measured the viscosity of a metallic glass-forming liquid, $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$, at pressures up to 6.1 GPa above the melting points by falling-sphere viscometry with ultrafast synchrotron x-ray imaging. Overall, the viscosity increases with pressure, while surprisingly, there is an abrupt drop between 3.2 GPa and 3.7 GPa, indicating the possible existence of a pressure-induced liquid-to-liquid transition. Pressure could change the short- and medium-range orders in the multicomponent glass-forming liquid, as suggested by the different crystalline outcomes after cooling to room temperature at high- and low-pressure ranges. Our work extended the viscosity investigation of metallic glass-forming liquids to the high-pressure regime, which will expand our understanding of liquid-liquid transitions and metallic glass formation.

Keywords: metallic glass-forming liquid; viscosity anomaly; high pressure; liquid polymorphism

1. Introduction

Metallic glasses (MGs) usually form if crystallization is avoided by rapidly quenching metallic melts through a glass transition [1-3]. The critical cooling rate required for glass formation limits the sample size of glasses that can be synthesized via melt quenching, which is a critical issue especially for MGs due to their relatively low glass-forming ability (GFA) [3]. Owing to the superior mechanical, magnetic, and chemical properties of MGs compared to conventional metallic materials, the development of various MGs with high GFA has been attracting extensive enduring research efforts. However, it remains a time-consuming work of trial and error in practice, even though some combinatorial methods and machine-learning-based prediction has been recently developed [4, 5]. Therefore, uncovering the fundamental mechanism underlying the GFA of the metallic alloy system is crucial.

Viscosity is the key parameter that affects the nucleation and growth processes of crystals in their supercooled liquids during cooling, therefore, dictating the GFA [6-10]. The glass-forming liquids showing an Arrhenius variation of the viscosity are classified as ‘strong’ liquids and the liquids showing non-Arrhenius behavior are classified as ‘fragile’ liquids when the liquids are cooling down from high temperatures approaching their glass transition temperatures [6-10]. The ‘strong’ liquids usually require a relatively low critical cooling rate to form glasses, showing high GFA. It has been found that some glass-forming liquids undergo a fragile-to-strong liquid transition during cooling towards a glassy state such as water [11], SiO_2 [12, 13], and BeF_2 [14], etc. Many studies have confirmed that this

fragile-to-strong liquid transition can also exist in many metallic glass-forming liquid systems [15-17], which were presumably to be closely related to glass transitions and GFA. Most of the fragile-to-strong liquid transitions occur in the supercooled liquid states and are believed to result from the short- and medium-range order changes in the liquids [18-20]. In addition, liquid-to-liquid transitions have been found in metallic glass-forming liquids [21-25], providing a vital opportunity for investigating the relationship between glass and liquid. However, so far, the studies on the viscosity of metallic glass-forming liquid are all limited to ambient pressure measurements. It is well known that pressure is another effective tool to tune the structure and properties of glasses and liquids and even can induce unusual polyamorphism in some MGs at room temperature [26, 27]. Therefore, it is intriguing to investigate how the viscosity of metallic glass-forming melts evolve at high pressures, which could shed new light on the metallic glass-forming liquids by providing a complete physical description of them.

Falling-sphere viscosity measurement using x-ray radiography in a Paris-Edinburgh (PE) cell has been developed to measure liquid viscosities at high pressure and high temperature [28, 29]. Using ultrafast x-ray imaging at frame rates up to $\sim 10^4$ frames per second (fps) in the PE cell, we could determine the viscosity of liquids at high pressures with high accuracy [30-32]. The falling-sphere viscometry has measured the viscosity of many geoscience-related high-pressure liquids such as FeS [33, 34], Fe-FeS [33], FeNiC [35], carbonate [30], and silicate melts [30, 32], which enables us to obtain viscosity data with typical uncertainties of ~0.5 decades in

units of Pa s.

In the present study, the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ bulk metallic glass (BMG), as a typical system with relatively simple composition and good GFA, was chosen to measure the viscosity of a metallic glass-forming liquid at high pressure above melting points using a PE cell. The structure of samples quenched from melts at high pressures was measured by *in situ* white beam energy dispersive x-ray diffraction (XRD). We found an abnormal viscosity drop, which is most likely linked to a liquid-liquid transition in MG melts induced by high pressure. This finding might shed new light on the liquid-to-liquid transitions in metallic glass-forming liquids.

2. Material and methods

The master ingots of the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ BMG were prepared by arc melting in a Ti-gettered Ar atmosphere and then cast into rods with a diameter of ~1 mm. High-pressure viscosity measurements were conducted using a PE cell installed at the beamline 16-BM-B, Advanced Photon Source (APS), Argonne national laboratory (ANL). The samples of ~1.5 mm in height were assembled in the high-pressure cell as shown in Fig. 1. Pressures were determined by the equation of state of MgO [36] and temperatures were estimated by power-temperature relation curves that were calibrated in a separate experiment using an identical cell configuration [28]. Falling-sphere viscosity measurements were conducted by using a tungsten carbide (WC) ball as a probing sphere. X-ray imaging was used to measure the falling velocity of the probing WC sphere using unfocused white x-rays with a beam size of 1.0 mm (horizontal) × 1.0-1.3 mm (vertical), depending on the height of the PE cell at

high pressures. The imaging rate was 250 fps, which enables us to precisely determine the terminal velocity

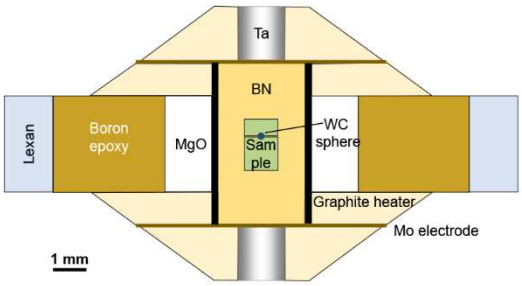


Fig. 1. A schematics illustration of the high-pressure Paris-Edinburgh cell assembly. Different colors represent different materials used in the cell. All the materials of the cell are quite transparent to the x-ray beam. The scale bar represents 1 mm.

of samples with a calibrated pixel resolution of 2.498 μm/pixel using a high-speed camera (Photron FASTCAM SA3, pixel size is 0.95 μm). The position of the WC sphere in each frame was analyzed by using the Tracker plugin in the ImageJ software package. The diameter of the WC sphere was measured by x-ray radiography and the density of the WC sphere was calculated by its equation of state. The viscosity (η) was calculated with the Stokes equation including the correction factors for the effect of the wall (F)[37] and for the end effect (E) [38]:

$$\eta = \frac{g d_s^2 (\rho_s - \rho_l)}{18 v} \frac{F}{E} \quad (1),$$

$$F = 1 - 2.104\left(\frac{d_s}{d_l}\right) + 2.09\left(\frac{d_s}{d_l}\right)^3 - 0.95\left(\frac{d_s}{d_l}\right)^5 \quad (2),$$

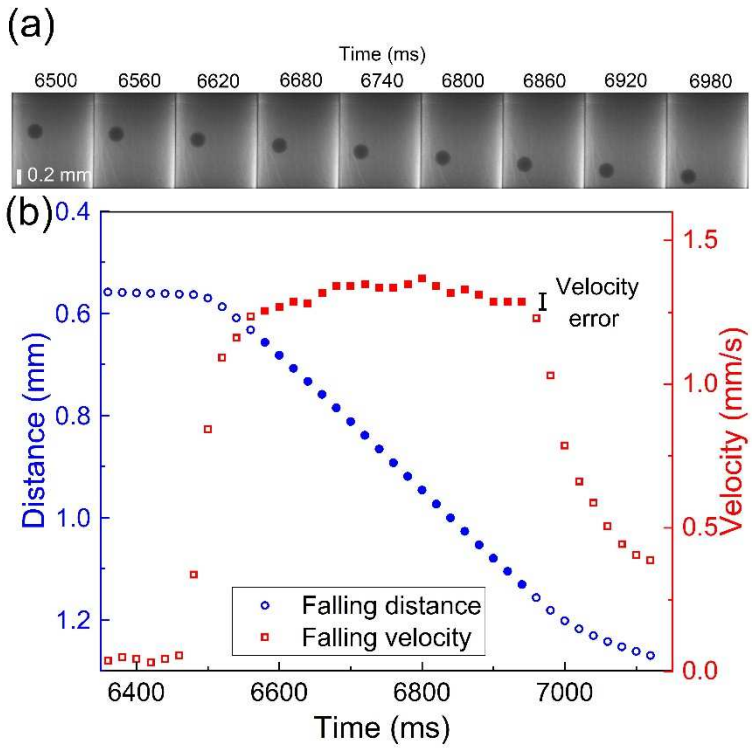
$$E = 1 + \frac{9}{8}\frac{d_s}{2Z} + \left(\frac{9}{8}\frac{d_s}{2Z}\right)^2 \quad (3),$$

where v, ρ, and d are the terminal velocity of the probing sphere, density, and diameter, respectively. The subscripts s and l denote properties of the probing sphere and liquid, respectively. Z is the total height of the molten sample. g is the gravitational acceleration. The uncertainties in terminal velocity dominate the precision of the

viscosity determination. High-speed camera of 250 fps used in our research with the identified constant velocity region at the given pressure, enables us to obtain accurate viscosity values with the relative error of ~1%.

3. Results and discussion

High-pressure viscosity measurements were carried out up to 6.1 GPa at temperatures just above melting. Fig. 2a shows selected images of the WC sphere in liquid $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ during falling at 1.7 GPa. The WC sphere is falling in a straight line, indicating that the sample is completely molten and there is no residual solid to interfere with the falling of the sphere. Thus, the terminal velocity can be obtained reliably from the falling trajectory of the WC sphere. Fig. 2b shows the



distance and velocity of the falling WC sphere as a function of time at 1.7 GPa. The

Fig. 2. Falling-sphere viscosity measurement using ultrafast x-ray imaging. (a) A time series of x-ray radiography images of the falling WC sphere (0.2 mm in diameter) in the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ glass-forming melt at 1.7 GPa and ~ 950 °C. (b) The results of the falling distance and the falling velocity for each frame (20 ms interval). Note that terminal velocity was achieved only at limited

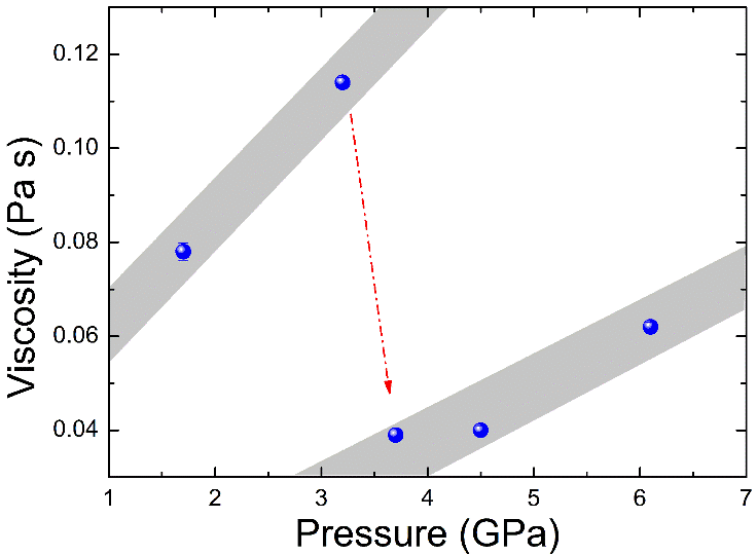
regions of falling distance (0.65-1.13 mm distance, indicated by filled symbols). The data clearly show that monitoring the motion of a falling sphere with substantial oversampling by using ultrafast imaging is essential to accurately determine terminal velocity and the resultant viscosity.

falling velocity first increased between 6460 ms and 6560 ms and reached an almost constant value (terminal velocity) after 6580 ms. Then, the falling velocity rapidly decreased after 6940 ms. Note that the terminal velocity can be achieved only at limited regions of the falling distance (filled symbols in Fig. 2b). The data demonstrate that

using ultrafast imaging to monitor the trajectory of a falling sphere with substantial oversampling is essential to accurately determine the terminal velocity.

Fig. 3 shows the viscosities of the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ melts as a function of pressure. The experimental results as well as the experimental conditions are summarized in Table 1. The viscosity at 1.7 GPa is 0.078 Pa s, which is consistent with the typical values of viscosity of Zr-based MG liquids in other studies measured at the similar temperatures [15, 39]. With the pressure increased from 1.7 GPa to 3.2 GPa, the viscosity increased from 0.078 Pa s to 0.114 Pa s. Interestingly, with pressure slightly increased by 0.5 GPa to 3.7 GPa, the viscosity sharply drops to 0.039 Pa s. With pressure further increased from 3.7 GPa to 6.1 GPa, the viscosity increases again with increasing pressure.

Crystalline phases were observed after cooling to room temperature, likely due to the limited cooling rate available in the PE cell. Fig. 4 shows the XRD patterns of the



crystalline phases quenched at different pressures in the PE cell. The major crystalline

Fig. 3. Viscosity of the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ metallic glass-forming melts at high pressures. Viscosity measurements were carried out up to 6.1 GPa. The gray zones are guides to the eye to show the pressure dependence of the viscosity below 3.2 GPa and above 3.7 GPa.

Table 1. Experimental parameters and the viscosity results. T_{melt} is the melt temperature (not the melting temperature) at which the measurement was carried out.

Pressure (GPa)	T_{melt} (°C)	Viscosity (Pa s)
1.7	950	0.078 ± 0.002
3.2	1000	0.114 ± 0.001
3.7	1100	0.039 ± 0.001
4.5	1090	0.041 ± 0.001
6.1	1090	0.062 ± 0.001

phase at 4.5 and 6.1 GPa were $Cu_{10}Zr_7$ (space group: *Cmce*) and $CuZr_2$ (space group: *I4/mmm*) phases, possibly together with minor $AlCu_2Zr$ (space group: *Fm-3m*) and some unidentified phases. Surprisingly, only the $CuZr_2$ phase was observed when the melt was cooled down at 3.2 GPa. The difference in the crystalline phases indicates that the potential energy of the system, therefore, the melts at low- and high- pressures

may be different, which rationalizes the abnormal viscosity drop.

This study measured the viscosities of a metallic glass-forming liquid at high pressures for the first time. Compared with the viscosity value at 1.7 and 3.2 GPa, the viscosity of the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ liquid shows positive pressure dependence, which is

similar to those of liquid metals [40]. On the other hand, our data clearly show an

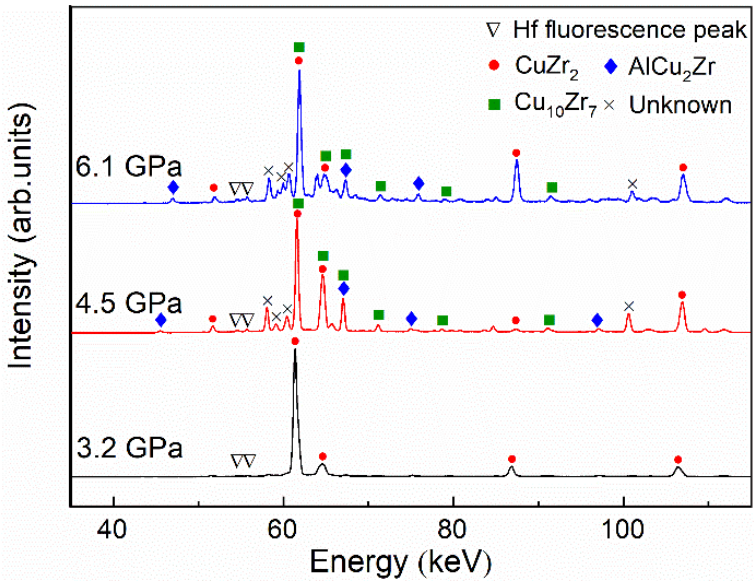


Fig. 4. In situ high-pressure XRD patterns of the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ samples at room temperature cooled down from melts at different pressures.

anomalous drop of viscosity at ~3.7 GPa, which may imply the existence of pressure-induced liquid-liquid transition.

Similarly to our observed viscosity anomaly at high pressures, anomalous viscosity drops were also reported in CuZr-based MG melts with decreasing temperature at ambient pressure conditions [22]. The viscosity anomaly coincides with an exothermic signal in the differential scanning calorimetry (DSC) curve, which is considered to be the existence of a liquid-to-liquid transition with varying temperatures in the CuZr-based melts. This liquid-liquid transition is a structural

transformation from strongly ordered high-density liquid to weak-local low-density liquids at low-temperature conditions [23, 41]. In these studies of the Cu-Zr based glass-forming liquids, the viscosity change is ranging from 1% to 8% at ambient pressure while in our experiment the viscosity changed from 0.114 to 0.039 Pa s by ~66%. Our observed viscosity anomaly at high-pressure conditions implies the existence of a similar liquid-liquid transition at ~3.7 GPa.

It is widely believed that the dynamic behavior is linked to the atomic structure of liquids such as the short-range and medium-range order in metallic melts [39, 42]. For instance, in the $Zr_{66.7}Cu_{33.3}$ MG, the short-range order is dominated by Cu-centered clusters and the fraction of highly-coordinated Cu clusters increases with pressure at the expense of lowly-coordinated ones, while the pressure effect on Zr-centered clusters is not significant [43, 44]. Similar results are also found in $Zr_{50}Cu_{44}Al_6$ metallic glass-forming liquids. Pressure increases the coordination of Cu with Al remarkably, but only moderately for Zr-Cu and Zr-Al pairs [45]. The medium-range order has been proposed to be closely related to liquid-liquid transitions in many studies[46-48]. A recent fluctuation electron microscopy study showed that the medium-range order clusters become more diverse in the Zr-Cu-Al bulk metallic glass after adding 5 at. % of Ag, suggesting that the Ag and its cluster may play an essential role in the formation of metastable phases [49, 50]. Our experimental observation of the sudden drop of viscosity of the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ liquid upon compression would be related to the liquid-liquid transition and could be induced by the different responses of various atomic pairs upon compression in the

system [51].

4. Conclusion

In conclusion, our falling-sphere viscosity measurement of the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$ glass-forming liquid up to 6.1 GPa revealed an abrupt drop of viscosity between 3.2 and 3.7 GPa, indicating the possible existence of a pressure-induced liquid-to-liquid phase transition in the $Zr_{46}Cu_{37.6}Ag_{8.4}Al_8$. Pressure could change the short- and medium-range orders in the multicomponent glass-forming liquid, resulting in different crystalline outcomes after cooling to room temperature at high- and low-pressure ranges. Our work extended the viscosity investigation of metallic glass-forming liquids to the high-pressure regime, which should expand our understanding of the glass transition and liquid-liquid transitions. If the distinct liquids at different pressures can be quenched as glasses, novel MGs may be obtained, and it opens a new way to explore the space for MGs development by alternative high-pressure means.

Credit authorship contribution statement

Qifan Wang: Investigation, Methodology, Writing – original draft. **Hongbo Lou:** Conceptualization, Investigation, Methodology, Writing – review & editing. **Yoshio Kono:** Investigation, Methodology, Writing – review & editing. **Dajio Ikuta:** Investigation, Methodology, Writing – review & editing. **Zhidan Zeng:** Writing – review & editing. **Qiaoshi Zeng:** Supervision, Conceptualization, Investigation, Methodology, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

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