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Transient regimes during high-temperature deformation of a bulk metallic glass: A free volume approach

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Abstract

The homogeneous deformation of a zirconium-based bulk metallic glass is investigated in the glass transition range. Compression and stress-relaxation tests have been conducted. The stress–strain curves are modeled in the framework of the free volume theory, including transient phenomena (overshoot and undershoot). This approach allows several physical parameters (activation volume, flow defect creation and relaxation coefficient) to be determined from a mechanical experiment. This model is able to rationalize the dependency of stress overshoot on relaxation time. It is shown that, due to the relationship between flow defect concentration and free volume model, it is impossible to determine the equilibrium flow defect concentration. However, the relative variation of flow defect is always the same, and all the model parameters depend on the equilibrium flow defect concentration. The methodology presented in this paper should, in the future, allow the consistency of the free volume model to be assessed.

Keywords: Metallic glasses; Compression test; High temperature deformation; Plastic deformation; Free volume model

1. Introduction

It is now well established that at high temperatures (typically $T > 0.8T_g$, where T_g is the glass transition temperature) bulk metallic glasses (BMGs) deform homogeneously when undergoing mechanical tests such as compression and tension at a given strain-rate. In this temperature range, these materials can undergo tensile elongations as high as 10,000% [1]. In steady-state conditions, the relation between flow stress and applied strain-rate of the plastic deformation of BMGs can be successfully described by the free volume model [2–5]. In this framework, the relation between stress (σ) and plastic strain-rate ($\dot{\varepsilon}$) is given by [6]:

$$\begin{aligned}\dot{\varepsilon} &= 2c_f\nu_D\exp\left(-\frac{\Delta G^m}{kT}\right)\sinh\left(\frac{\sigma V}{2\sqrt{3}kT}\right)\\ &= c_f\dot{\varepsilon}_{0,c}\sinh\left(\frac{\sigma V}{2\sqrt{3}kT}\right)\end{aligned} \tag{1}$$

where c_f is the flow defect concentration deduced from the free volume, ν_D the Debye frequency, ΔG^m the migration free energy of the flow defects, V the activation volume and $\dot{\varepsilon}_{0,c}$ the frequency factor. De Hey et al. [3] have shown that under dynamic conditions, c_f is a balance between the creation of flow defects induced by plastic strain and a structural relaxation effect, which tends to annihilate the flow defects until the equilibrium concentration is reached. The rate equation governing c_f is given by:

$$\dot{c}_f = -k_r c_f(c_f - c_{f,eq}) + a_x\dot{\varepsilon}c_f\ln^2 c_f, \tag{2}$$

where k_r is the rate constant of the structural relaxation, $c_{f,eq}$ is the equilibrium concentration of flow defects at a given temperature (i.e. without any strain), which is thermally activated, and a_x is the creation factor giving the

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proportionality between plastic strain and flow defect concentration.

Another aspect of BMG behavior is the presence, during uniaxial tests at relatively high strain-rate (or stress), of transient states prior to stress plateauing (in the case of constant strain-rate experiments in compression or tension) or of a steady-state strain-rate (in creep conditions). For experiments performed at constant strain-rate, these transients take the form of stress overshoots, but in the case of strain-rate jump tests, undershoots can also be observed [7]. Within the framework of the free volume model, these changes in stress can be understood as the time necessary to reach a steady-state value of c_f according to Eq. (2) (i.e. $\dot{c}_f = 0$), which then corresponds to the stress plateau value at constant strain-rate.

In a previous paper [8], an internal variable model was proposed to rationalize the stress overshoot phenomenon, in which the total strain-rate $\dot{\varepsilon}$ was written:

$$\dot{\varepsilon} = c_f \dot{\varepsilon}_{0,c} \sinh\left(\frac{\sigma V}{2\sqrt{3}kT}\right) + \frac{\dot{\sigma}}{E} + \frac{\sigma}{\eta} \tag{3}$$

and Eq. (3) was coupled to Eq. (2). In Eq. (3), η_a is the inelastic modulus of the glass at temperature T. However, it turns out that this supplementary degree of freedom is not necessary from a phenomenological point of view: it is possible to rationalize the stress overshoot phenomenon with a purely elastic–viscoplastic model based on the free volume theory, which is the object of the present paper. Moreover, in Ref. [8] we had to use a low value for the Young's modulus E, and we argued that it was due to the high value of the temperature; however, measurements conducted since then at frequencies higher than 2 kHz on a free–free bar apparatus (unpublished) have shown that at the temperature at which the experiments were conducted, the Young's modulus is approximately 77 GPa (vs. 81 GPa, measured at room temperature). This result suggests that the dependency of this modulus on temperature remains very limited, and hence, it is difficult to give a physical meaning to the parameter η as it is generally believed that the low apparent value of the Young's modulus is due to inelastic phenomena. However, in the current work, η does not allow the use of the actual value of the Young's modulus and hence does not really play the role of an inelastic contribution. Moreover, it has been established, thanks to internal friction measurements (e.g. [9]), that the high-temperature inelasticity of BMGs can only be described in terms of the distribution of relaxation times: an unique parameter η is then unable to rationalize such phenomena. Hence, this term σ/η is not satisfying from a physical point of view either. We propose here to examine another model, based solely on the free volume approach proposed by Spaepen [6], which should be easier to interpret. We propose an original procedure to determine solely from mechanical tests the parameters of a model proposed in a previous study [4] of the steady state, in which the dependency of the plateau stress on strain-rate for a Zr-based BMG ($Zr_{52.5}Al_{10}Cu_{22}Ti_{2.5}Ni_{13}$) was successfully described in terms of the free volume model. The relative variation of the flow defect concentration was predicted, along with the value of the key parameters controlling the steady-state regime, i.e. the ratio a_x/k_r and the frequency factor $\dot{\varepsilon}_{0,c}$. However, this study showed that it was not possible to deduce an absolute value of the concentration of flow defects from mechanical testing. Indeed, due to the exponential law linking free volume and flow defect concentration, several triplets ($a_x/k_r, \dot{\varepsilon}_{0,c}$, $c_{f,eq}$) can fit satisfactorily the experimental data, and the flow defect concentration cannot be determined from a study of only the steady state. It is important to note that these calculations were carried out after an estimation of the activation volume V. In order to minimize any change in the flow defect concentration, V was measured in order to fit Eq. (4) with the maximum stress when overshoots were present:

$$\dot{\varepsilon} = \dot{\varepsilon}_0 \sinh\left(\frac{\sigma V}{2\sqrt{3}kT}\right). \tag{4}$$

In this procedure, the overshoot was thus reduced to the values of both the maximum stress and the plateau stress. In other words, no strain data were taken into account.

The aim of the present paper is to extend the methodology initially suggested in Ref. [4] to the modeling of transient regimes. It will be shown that such regimes allow various parameters of the model to be determined, namely the free volume creation parameter a_x, the structural relaxation k_r and the activation volume V, all of which cannot be determined with the procedure proposed in Ref. [4]. (Indeed, in Ref. [4] it was hypothesized that V could be determined from the highest values of the stress of a strain-rate jump experiment; the present study of the transient phenomena does not use this hypothesis and gives a more accurate value of the activation volume.)

2. Materials and methods

A glass of composition $Zr_{52.5}Al_{10}Cu_{27}Ti_{2.5}Ni_8$ was made for this investigation. The pure metals were melted by induction in a cold crucible in an argon atmosphere and injected under pressure into a water-cooled copper mold. The amorphous character of the alloy was confirmed by X-ray diffraction, neutron diffraction and differential scanning calorimetry (DSC) [8]. DSC scans at a heating rate of 10 K min^{-1} under argon atmosphere yielded the characteristic temperatures (glass transition temperature T_g and crystallization temperature T_x): $T_{g2} = 663$ K (T_{g2} corresponds to the inflexion point of the glass transition) and $T_x = 745$ K, leading to a $\Delta T = 82$ K ($\Delta T = T_x - T_g$), which indicates successful formation of a BMG. Cylindrical samples 5 mm in diameter and 8 mm in height were used for the compression tests, which were carried out at 683 K, after heating at a rate of 10 K min^{-1}. The strain-rates were between 5×10^{-4} and $2.5 \times 10^{-3}\ \text{s}^{-1}$. Both strain-rate jumps and deformation-relaxation tests were performed. Samples were characterized post-mortem by X-ray diffraction (copper wavelength, using a $\theta - 2\theta$ goni-