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Consistency of the free-volume approach to the homogeneous deformation of metallic glasses

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ABSTRACT

One of the most widely used approaches to model metallic-glasses high-temperature homogeneous deformation is the free-volume theory, developed by Cohen and Turnbull and extended by Spaepen. A simple elastoviscoplastic formulation has been proposed that allows one to determine various parameters of such a model. This approach is applied here to the results obtained by de Hey et al. on a Pd-based metallic glass. In their study, de Hey et al. were able to determine some of the parameters used in the elastoviscoplastic formulation through DSC modeling coupled with mechanical tests, and the consistency of the two viewpoints was assessed.

1. Introduction

Understanding the physical basis of the homogeneous deformation of bulk metallic glasses is an important fundamental as well as practical subject. On a fundamental basis, there exists no consensus on the nature of the mechanism(s) responsible for the deformation of amorphous materials. In practice, the homogeneous mode is of interest, particularly for shaping operations on metallic glasses at high temperature. For the latter class of materials, one of the most widely used models to describe the homogeneous deformation is the free-volume model. It was proposed by Cohen and Turnbull [1–3] for liquids and applied to the glass-transition phenomenon, and was later adapted to the case of the deformation of metallic glasses by Spaepen [4]. In this framework, the carriers of plastic deformation are so-called flow defects, whose concentration c_f is given by:

$$c_f = \exp\left(-\frac{\gamma v^*}{v_f}\right) \tag{1}$$

where v^* is the critical free volume level, v_f is the average free volume per atom and γ is a geometrical factor allowing for the overlap of free-volume areas. The viscoplastic strain rate then writes:

$$\dot{\gamma} = c_f 2\nu \exp\left(-\frac{\Delta G^m}{kT}\right) \sinh\left(\frac{\tau \Omega}{kT}\right) \tag{2}$$

where ν is the atomic vibration frequency, ΔG^m is the migration free energy, τ is the shear stress and Ω is the activation volume. k and T have their usual meaning.

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This model leads to a satisfying rationalization of the dependency of glasses viscosity with temperature, by assuming that the average free volume writes:

$$v_f = \frac{T - T_0}{B} \gamma v^* \tag{3}$$

which allows us to write the Vogel–Fulcher–Tammann equation from the free-volume system, the VFT equation being commonly used to describe the temperature dependence the viscosity of undercooled liquids (see for example [5]). This equation is then interpreted as a temperature-dependent equilibrium concentration of defects, c_{eq}:

$$c_{eq} = \exp\left(-\frac{B}{T - T_0}\right) \tag{4}$$

where B and T_0 are material parameters. For a glass at sufficiently high temperature, if the free-volume level is higher than the equilibrium one, the so-called structural relaxation occurs. This phenomenon is notably accompanied by a density increase. The viscosity of the glass then increases linearly with time, which in the framework of the free-volume model can be understood as an annihilation of flow defects with a second-order kinetics, which writes [6–8]:

$$\dot{c}_f = k_r c_f (c_f - c_{eq}) \tag{5}$$

where k_r is the structural relaxation kinetic coefficient, which is thermally activated [8].

The stress–strain curve of a metallic glass deformed in the homogeneous mode exhibits stress overshoots whose amplitude increases with the strain rate. Various studies have investigated this phenomenon (see, for example, [9–11]), and from a free-volume perspective, it is due to an increase of the flow defect concentration. Several forms of the dependency of the flow defect concentration with strain have been studied, and based on their measurements coupling uniaxial tests and DSC results, de Hey et al. have shown [12] that the best description of the flow defect creation by strain is:

$$\dot{c}_f = a_x \dot{\varepsilon}_p c_f \ln^2(c_f) \tag{6}$$

where $\dot{\varepsilon}_p$ is the plastic strain-rate and a_x is the flow defect creation coefficient. This equation is derived from the free-volume model assuming $\mathrm{d}v_f \propto \mathrm{d}\varepsilon_p$. By coupling Eqs. (5) and (6), the kinetic of flow defect concentration, when a metallic glass is plastically deformed, is given by:

$$\dot{c}_f = a_x \dot{\varepsilon}_p c_f \ln^2(c_f) - k_r c_f (c_f - c_{eq}) \tag{7}$$

whose steady-state solution c_f^* writes:

$$c_f^* = c_{eq} + \frac{a_x}{k_r} \dot{\varepsilon}^p \ln^2 c_f^* \tag{8}$$

By coupling Eqs. (2) and (8), it is possible (see [13]) to deduce the ratio a_x/k_r and $\dot{\varepsilon}_{0,c}$, knowing the activation volume Ω and c_{eq}.

Assuming that the total strain is the sum of the elastic and viscoplastic contributions, it writes (in uniaxial conditions):

$$\dot{\varepsilon} = \frac{\dot{\sigma}}{E} + c_f \varepsilon_{0,c} \sinh\left(\frac{\sigma \Omega}{2\sqrt{3} kT}\right) \tag{9}$$

where E is the Young modulus. By coupling Eq. (9) with Eq. (7), it is then possible to model mechanical experiments [11]. By coupling this type of modeling with the approach based on the steady state [13], most of the parameters of the free-volume model—at a given temperature—can be deduced from mechanical results only, with the noticeable exception of the equilibrium flow defect concentration, c_{eq}.

The aim of the present paper is to examine the consistency of the elastovisoplastic approach, by comparing the values found by de Hey et al. [12] for the various parameters of the model thanks to DSC measurements with the ones that can be deduced from a purely mechanical analysis of their results, following the method depicted in [11].

2. Structural parameters determination and comparison

In their study [12] of the structural modifications induced by plastic deformation of a PdNiP metallic glass in the homogeneous domain correlating tensile tests and DSC measurements, de Hey et al. have conducted one experiment that is particularly favorable to apply the mechanical analysis proposed in [11]. Indeed, using the results of uniaxial strain-rate jump experiments, it is possible to determine almost all the parameters of Eqs. (7) and (9), which include the structural relaxation coefficient k_r and the free-volume creation coefficient a_x. These two parameters were also determined by de Hey et al. thanks to the modeling of DSC curves, *i.e.* a thermodynamical approach.

However, for the model to be consistent, the values found by an approach and the others must be the same. In Fig. 1 are presented the data published by de Hey et al. for a tensile test involving strain-rate jumps between $\dot{\varepsilon} = 8.3 \times 10^{-5}$ s^{-1} and 4.2×10^{-5} s^{-1}. By adjusting the parameters of Eqs. (7) and (9), it is possible to obtain a rather satisfying fit of the