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# Activation volume of microscopic processes in amorphous Pd<sub>77.5</sub>Cu<sub>6.0</sub>Si<sub>16.5</sub> due to stress and temperature

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#### ARTICLE INFO

#### ABSTRACT

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#### 1. Introduction

The transition from an amorphous solid state to the supercooled liquid is described by different microscopic activation processes, which are caused by local heterogeneities [1–3]. The length scale of relaxation processes in metallic glasses reaches from the distance of nearest neighbors (hard spheres model) to clusters of atoms (shear transformation zones – STZ's) which show a cooperative behavior [4–7]. However, the change in viscosity near Tg is not fully explained with these heterogeneities. Already in 1965 G. Adam and J. H. Gibbs predicted cooperative rearranging regions (CRRs) to be an essential feature of relaxation processes in metallic glasses [8]. To get more insight of these processes, one has to study the atomic configurations and its change with temperature or shear stress. Molecular dynamic simulations lead to the conclusion that an arrangement of atoms in the order of 20 Å is characteristic. Thereby a configuration of a string or quadrupolar feature (13–55 atoms) is particularly favourable [7].

Teichler et al. could identify strings of atoms as microscopic reasons for fast relaxation processes, whereby the involved atoms are moving cooperatively and are identified as secondary relaxation processes [9]. These processes can lead to reversible relaxations. [10] With reduction of a dimension of bulk like material to a thin film, Bedorf et al. argue that the string like processes are reduced or disappeared at a film thickness of ca. 30 nm [11]. This gives a first impression for a comparison with simulated length scales.

A. Argon extended the idea of CRRs to STZs, which describe spherical areas with a small number of atoms in diameter. In these regions shear stress is reduced by undergoing plastic deformation

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Creep/recovery measurements of amorphous PdCuSi ribbons have been performed. From these data we calculated the activation volume of relaxation processes in the temperature range from the glass transition temperature (Tg) to crystallization. The results indicate a correlation between applied stress and the activation volume of clusters in temperature regime close to Tg. With these results it is possible to compare molecular dynamic simulations for microscopic loss processes in metallic glasses with experimental values for mobile clusters.

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[12]. A further development of this model is demonstrated as cooperative shear model (CSM) by W. L. Johnson and K. Samwer [13]. Here STZs were predicted to consist of approximately 100 atoms, which are integrated into the picture of a potential energy landscape (PEL) [14]. In molecular dynamic simulations a diameter of STZs of 1.5 nm up to 100 nm was determined [15–17]. Recent publications of Chen et al. using a rate jump nanoidentation method give a first experimental atomic number of STZ-size in the order of 2.5 nm<sup>3</sup>–6.5 nm<sup>3</sup> depending on the type of metallic glass [18]. In a similar approach the authors relax the sample by thermal annealing and find a surprisingly large reduction of STZ-size of 50% [19].

Here we present creep/recovery experiments to get an estimate of the activation volume and thus to the length scale of heterogeneities. Thereby, the activation volume  $\Omega$  is calculated by the following:

$$\Omega = kT \left(\frac{\partial \ln \dot{\varepsilon}}{\partial \sigma}\right)_{p,T} \tag{1}$$

at which the relation of strain rate and stress is called the strain rate sensitivity (a),

$$a = \left(\frac{\partial \ln \sigma}{\partial \ln \dot{\epsilon}}\right)_{p,T} \tag{2}$$

(with k, the Boltzmann constant, T the absolute temperature, p the pressure,  $\dot{\epsilon}$  the strain rate and  $\sigma$  the external stress) [18–20]. To calculate an activation volume it is necessary to be in the anelastic area of the material. If the strain rate sensitivity is constant over a broad temperature range, the material is believed to be in the linear, elastic part of the stress strain curve, so that Eq. (1) is not applicable.

Because of this, the main focus is on the transition from the amorphous solid state (glass) into the supercooled liquid. Therefore

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changes in the activation volume at different temperatures and different external stresses are studied.

#### 2. Sample preparation and measuring setup

Samples of Pd<sub>77.5</sub>Cu<sub>6.0</sub>Si<sub>16.5</sub> are produced by melt spinning [21] and used as cast. The amorphous state was validated by X-ray measurements (data not shown). For the measurements ribbons with a length of 11 mm-13 mm, a width approx. 1.8 mm and a thickness of 0.025 mm-0.033 mm were used. Measurements were done in a creep/recovery mode using a DMA 7 from PerkinElmer [22]. To observe the creep behavior an upper force of 450 mN-950 mN (approx. 10 MPa-32 MPa) and at the recovery part a lower force of 50 mN-450 mN (approx. 1 MPa-10 MPa) was applied. At the higher stress, the force is held for 1.1 min and after jumping back to the lower force for 0.5 min. Therefore the material has time to relax the stress. During the measurement it was heated continuously from 280 K to 680 K ( $T_x \approx 640$  K) with a heating rate of 0.78 K/min and measured in steps of 0.2 s. The sample length *l* is detected via a linear variable differential transformer (LVDT), so the relative strain  $\varepsilon$  could be calculated by the following:

$$\varepsilon = \frac{\Delta l}{l}.$$
(3)

To determine the activation volume with Eq. (1), four approximations are assumed:

1. The Eq. (1) can only be appropriated by using a constant strain rate. In the creep/recovery measurements this is not possible, because the force is changing by jumps. However, Eq. (1) is only valid in the linear part after the force jump. A linear connection between the applied stress and a fictitious strain rate  $\dot{\varepsilon}$  is given by the following:

$$\dot{\varepsilon} = \frac{\varepsilon}{t} \tag{4}$$

(with t the time). Therefore we can only use the part of 15 data points (equivalent to 3 s) after the force jumps to make sure only the linear part is considered.

- 2. Eq. (1) is also only to be used at a constant temperature. However, with a heating rate of below 1 K/min, the temperature can be regarded as constant, because in the observed time interval of 3 s the temperature increase is extremely low.
- 3. The absolute value of the activation volume was taken.
- To calculate the activation length (the diameter of the activation volume), it was assumed, that the activation volume is spherical. [10,11].

#### 3. Results

To investigate the sample behavior during the experiment the true sample strain is plotted versus the temperature.

Fig. 1 shows the dependence of true strain on sample temperature. The inset shows a magnification of the graph where the behavior at the force jumps can be observed.

With rising temperature a linear increase of the strain up to 620 K (Tg) is recognized, where the material becomes soft. This is shown by the dramatic change in the slope of the data. The beginning of the crystallization is noticed by flattening out of the strain around 640 K.

With this measurement the activation volume over the temperature range from Tg to the supercooled liquid and up to the crystallization could be calculated (see Eq. 1).

At temperatures near to Tg the activation volume for the higher forces starts to turn over at around 618 K and decreases after 623 K, for lower forces this turning is observed at 626 K. The maximum of the data for 450 mN is at 618 K ( $\Omega$ =8.48 nm<sup>3</sup>) and for 50 mN 626 K

**Fig. 1.** Dependance of true strain on sample temperature. The inset shows the creep behavior at force jumps (450 mN to 50 mN and back) followed by the linear part.

 $(\Omega = 8.59 \text{ nm}^3)$ . Consequently the maximum of the activation volume differs in temperature but barely in its value. In the range of the supercooled liquid the difference becomes smaller and with reaching the crystallization the curves of different stresses overlap again. In the inset of Fig. 2 one can see the strain rate sensitivity coefficient versus the temperature. A nearly constant behavior from room temperature up to Tg is recognized followed by a drastic increase at 610 K and a drop down due to crystallization. This shows that a calculation of the activation volume in the area from 610 K to 670 K is possible because the material becomes strain rate sensitive.

Fig. 3 shows the behavior of the activation volume at different external stresses and the corresponding temperature onset (turn at Tg). With higher stress the activation volume decreases and the temperature onset shifts to lower values.

One can recognize a change in the activation volume of about  $4.6 \text{ nm}^3$  ( $9.95 \text{ nm}^3$  to  $5.32 \text{ nm}^3$ ) by increasing the external stress from 1 MPa to 32 MPa. Also a big difference in the temperature onset of more than 26 K (626 K to 599 K) is observed.

To verify this new analysis method, the strain rate in the plastic regime (at approx. Tg) is observed at fixed strains for three different stresses (9.5 MPa, 13.9 MPa and 15.9 MPa). Therefore the fictitious strain rate (using Eq. 4) is calculated for five different strains (2%-4%) and plotted logarithmically versus the true stress (see Fig. 4) to calculate the lead of the five strain curves. By using Eq. (1)

**Fig 2.** Connection between activation volume and temperature from 610 K–670 K. Two different stresses are compared, 10 MPa (black squares) and 1 MPa (red dots). The inset reflects the strain rate sensitivity in the whole temperature range.





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