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Evolution of the activation energy spectrum and defect concentration upon structural relaxation of a metallic glass determined using calorimetry and shear modulus data

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Abstract

We performed high precision shear modulus and calorimetry measurements on a model Pd-based glass subjected to different annealing treatments below the glass transition temperature T_g . Applying the Interstitialcy theory, we found that structural relaxation-induced heat release/absorption below and near T_g can be precisely described using shear modulus data confirming thus the generic relationship between the heat and elastic relaxation effects in metallic glasses. The same approach is used for the reconstruction of underlying activation energy spectra. It is found that the spectra derived from calorimetry and shear modulus data are very close. Integration of these spectra gives the change of the concentration of frozen-in defects and its absolute value for different stages of structural relaxation.

Keywords: metallic glasses, structural relaxation, activation energy spectrum, shear modulus, calorimetry, Interstitialcy theory

1. Introduction

The non-crystallinity of metallic glasses (MGs) is responsible for a gradual change of their structural state occurring upon annealing below the glass transition temperature T_g . This phenomenon was documented long ago is generally referred to as structural relaxation [1, 2]. Structural relaxation results in the alteration of many MGs' physical properties. Some properties change moderately (density [3], internal energy [4], elastic constants [5, 6], electrical resistivity [7, 8], etc) while the other alter drastically, by orders of magnitude (viscosity [9], diffusivity [10]). The studies of structural relaxation kinetics performed in the 70-s and 80-s of the past century as well as later investigations showed that elementary atomic rearrangements responsible for structural relaxation are distributed in activation energy [4, 11–13]. Several approaches were suggested to study distributed relaxations and some of them are being used hitherto. One of the simplest ways is to fit the relaxation kinetics by a two-relaxation time model or by a sum of exponential relaxations [14, 15]. A widely used approach applies the Kohlrausch-Williams-Watts function [16–19], which is supposed to reflect a distribution of relaxation times (activation energies) [6]. This distribution can be also modelled by a Gaussian spectrum [20, 21]. Another well-known approach to the understanding of distributed relaxations is employed by the activation energy spectrum model suggested in the 80-s [12] and gained much popularity since then [22–24].

A salient feature of all these approaches is that the number of atomic relaxing sites as a function of the activation energy (relaxation time), which constitutes the activation energy spectrum (AES), can be determined only in some relative units and, therefore, the integration of the AES cannot give the absolute number (concentration) of "defects" responsible for relaxation complicating thus the understanding of underlying physical mechanisms. The reason for this consists in the fact that these approaches are mostly phenomenological and do not relate the relaxation kinetics with the real "defect" structure of glass.

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