STRUCTURAL RELAXATION OF STRESS-ANNEALED
Fe–Ni–B AMORPHOUS ALLOY[1]

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Structural relaxation during strain recovery of the metallic glass $Fe_{40}Ni_{41}B_{19}$ after longtime stress-annealing at different temperatures below the crystallization temperature was investigated by anisothermal DSC and dilatometric experiments. The structural relaxation effects depend on the stress-annealing temperature of the amorphous metallic alloy. The activation energy spectra were calculated from the anisothermal experimental data. The influence of the annealing temperature on the shape of creep recovery spectra was analyzed.

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1   Introduction

As a result of high cooling rate which does not allow sufficient time for atomic rearrangement to reach the structure of so called ideal glass, metallic glasses (MGs) are not in their internal equilibrium. Structural relaxation phenomena are associated to changes in the short-range ordering of both chemical and structural types and there is a decrease in enthalpy associated to these processes. Changes in topological short-range order involve largely the redistribution of free volume within the glass and changes in the chemical short-range order is the rearrangement of the atomic species on the sites. These changes produce significant variations in many physical mechanical and magnetic mechanical properties [1–3].

MGs under certain conditions exhibit deformation that is macroscopically reversible but delayed in time. In particular, pre-deformed samples can partially restore their shape after stress removal. Such behaviour is known as strain recovery. It was noticed that the anelastic deformation of MGs could not be described by a single relaxation process. A sum of exponential decays, spanning a spectrum of time constants, is required to describe the anelastic component of the homogeneous strain response of amorphous alloys to applied stress [4,5].

The strain recovery rate directly reflects of internal atomic rearrangement and the analysis of the strain-rate kinetics can provide important information about the glassy state. The aim of the presented work is to study the influence of the longtime loading at different annealing temperatures on the structural relaxation and on the shape of creep recovery spectra of the metallic glass based on Fe–Ni–B.

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2 Experimental

An amorphous ribbon of the nominal composition $Fe_{40}Ni_{41}B_{19}$ with the thickness of 17.3 μm was used in the experiment. The specimens of 4 mm width were cut from as-received ribbon, heated up to the different annealing temperatures T_a from 150 to 300 °C and annealed for 20 hours under the external tensile stress of 383 MPa inside a tube furnace with a flowing nitrogen atmosphere. After finishing the stress-annealing the samples were cooled down to the room temperature and then unloaded. Samples annealed under the same conditions but without an applied stress were used as reference specimens.

After that the thermal analysis measurements were carried out using a Perkin Elmer DSC 7 differential scanning calorimeter and a Setaram TMA 92 thermomechanical analyser in the tension arrangement with the absolute resolution of 10 nm. The changes of the enthalpy and of the length Δl were measured under linear heating with the constant heating rate β of 20 and 10 Kmin^{-1}, respectively, on both the stress-annealed and the reference samples. A flow of pure nitrogen was used to protect samples. Temperature stability was better than 0.2 K.

Obtained experimental data were analyzed using the simple method for calculating the spectrum of activation energy $N(E)$ from a linear heating experiment: $T = T_o + \beta t$, where T_o is the starting temperature of annealing and t is the time. The activation energy spectrum (AES) model is based on the equation proposed in [5] and developed in [6]:

$$\gamma_A(T) = \int\limits_0^{+\infty} N(E)\, \frac{(\alpha_1 T + \alpha_2 T^2) - E}{k\overline{T}}\, dE \tag{1}$$

with parameters: $\alpha_1 = (k/b)(-a - 1 + \ln(\nu_o \overline{T}/\beta))$, $\alpha_2 = k/b\overline{T}$, where a, b are constants ($a = 2.49663$, $b = 1.03197$), $\overline{T}$ is the average temperature during heating, k is the Boltzmann constant and ν_o denotes the attempt frequency, $\gamma_A(T) = \sqrt{3}\Delta l/l_o$ ($l_o = 15$ mm is an effective length of the sample) is the shear anelastic deformation measured at a temperature T. This modern method without prior assumptions about the shape or other properties of the spectrum uses the fast Fourier transformation to determine the activation energy spectrum from the experimental data. The method has been described and tested in more detail in paper [7].

3 Results and data analysis

DSC traces recorded for the stress-annealed samples are shown in Fig. 1. All measured curves obtained at a constant heating rate clearly show some exothermic effects. The glass forming temperature and the onset of crystallization is denoted as T_g and T_x, respectively. The wide exothermic decreases are present for all samples. They tend to shift towards high temperatures as the stress-annealed temperature is increases.

The anelastic shear strain measured under linear heating as a relative length change of the samples after substracting the linear elongation of the samples due to the thermal expansion is given in Fig. 2. The creep strain recovery manifests itself in a relative shrinking of stress-annealed samples in regard of reference ones. The annealing under applied stress leads to different amounts of total deformation. The maximal recovered anelastic strain was found to be 5×10^{-3} which is in agreement with the results published elsewhere [7, 8].
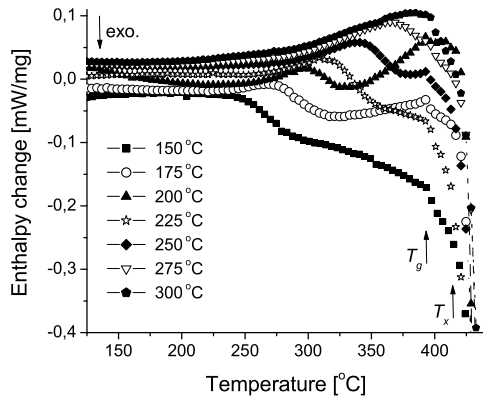
Fig. 1. DSC traces recorded for the samples stress-annealed at the indicated temperatures.
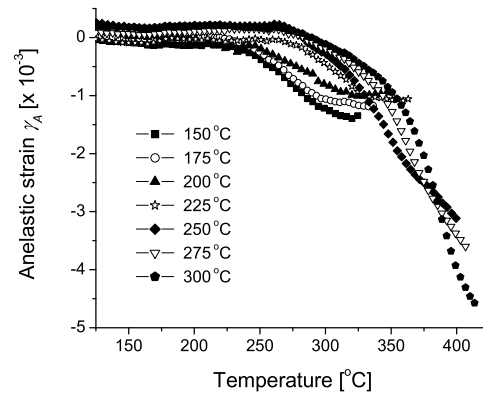

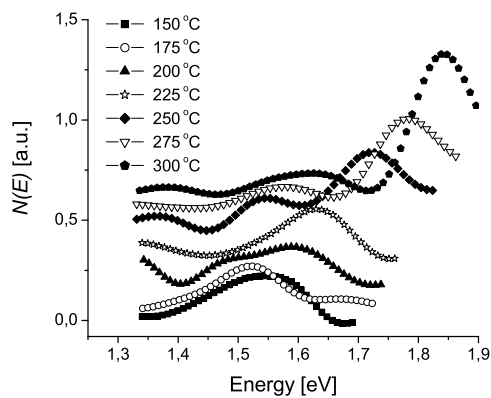Fig. 2. The anelastic shear strain for the samples stress-annealed at the indicated temperatures.

Fig. 3. Creep recovery spectra $N(E)$ calculated from the linear heating experiments in Fig. 2.
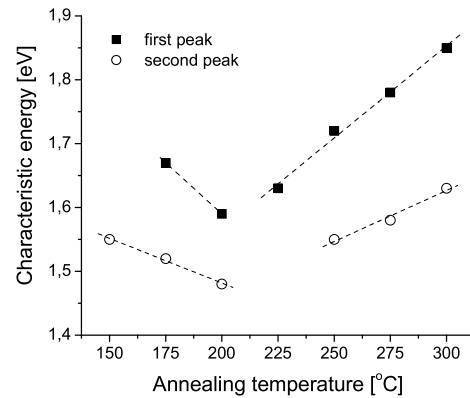

Fig. 4. Peak positions in depending on the annealing temperature. The lines are depicted only for better eye orientation.

From these non-isothermal creep strain recovery experiments the AES were calculated using a modern method based on Fourier technique. The results of our calculations can be seen in Fig. 3. Creep recovery spectra have a discrete character, they consist of a finite number of peaks with well defined characteristic activation energies. Each of peaks represents probably a distribution of different type of deformation defects in the amorphous structure. The height of peaks in calculated creep recovery spectra tends to increase with the increasing activation energy. The positions of two most significant peaks in depending on the stress-annealing temperature are shown in Fig. 4. Two tendencies of peak position dependence on the T_a are evident. At lower T_a the characteristic energy of peaks decreases as the stress-annealing temperature increases meanwhile at higher T_a the opposite tendency is observed. It can be connected with different structural states of the samples obtained during the stress-annealing at different temperatures.

MGs are prepared by rapid melt quenching and, therefore, are in non-equilibrium state. Irreversible local atomic rearrangements which arise as a results of any heat treatment are generally referred to as structural relaxation. The strain recovery kinetics can be well described using the directional structural relaxation model [9, 10] which treats homogeneous plastic flow of MGs as a result of structural relaxation oriented by the external stress. So the strain recovery can be interpreted as a set of irreversible atomic rearrangements in spatially separated regions of structure called relaxation centers (RCs) oriented favorably or unfavorably to the external stress.

As the structural relaxation depends on the stress-annealing temperature, it can be said that in the samples stress-annealed at lower temperatures both RCs, the paralell and the antiparalell in sign to the external stress, rearrange during strain recovery. As the annealing temperature increases the influence of antiparalelly oriented RCs decreases thus for higher temperatures only paralelly oriented RCs contribute to the strain recovery process. It influences also the total shrinking during the creep strain recovery. The rearrangements of both the favorably and the unfavorably oriented RCs are connected with lowering a free energy of amorphous structure towards an equilibrium glassy state and it manifests itself in a decrease on DSC traces. The energy accumulated during creep starts to release at the temperatures about T_a+100 °C. It is also evident from the calculated spectra that the creep strain recovery is determined by the temperature of stress-annealing. The opposite tendency of characteristic energies of creep recovery for the samples pre-annealed at lower temperatures can be associated with the cooperation both RCs, the paralell and the antiparalell in sign to the external stress.

4   Conclusion

Different creep strain accumulation is realized during the stress-annealing of Fe–Ni–B amorphous ribbon in dependence on the annealing temperature. This has an influence on the structural relaxation of the metallic glass. Both relaxation centers, the paralell and the antiparalell in sign to the external stress, rearrange during strain recovery in the samples stress-annealed at lower temperatures. In the samples stress-annealed at higher temperatures only the relaxation centers favorably oriented to the external stress contribute to the strain recovery process.

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