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Published in:
Central European Journal of Physics

DOI:
[10.2478/s11534-007-0006-9](https://doi.org/10.2478/s11534-007-0006-9)

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2007

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Jurikova, A., Csach, K., Miskuf, J., & Ocelik, V. (2007). Creep strain recovery of Fe-Ni-B amorphous metallic ribbon. *Central European Journal of Physics*, 5(2), 177-187. <https://doi.org/10.2478/s11534-007-0006-9>

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Creep strain recovery of Fe–Ni–B amorphous metallic ribbon

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Received 30 November 2006; accepted 1 February 2007

Abstract: Creep strain recovery and structural relaxation of the amorphous metallic glass $\text{Fe}_{40}\text{Ni}_{41}\text{B}_{19}$ after longtime loading at different annealing temperatures below the glass transition temperature have been studied using anisothermal differential scanning calorimetry (DSC) and dilatometry (TMA). It has been demonstrated that structural relaxation effects depend on the stress-annealing temperature of the amorphous ribbon. The structural relaxation states of the amorphous ribbon annealed at different temperatures under and without applied stress have been compared. The activation energy spectra were calculated from the anisothermal dilatometric measurements using the modern method based on the Fourier transformation technique. The influence of the annealing temperature on the shape of creep strain recovery spectra has been analyzed.

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Keywords: Metallic glass, anelastic strain, creep recovery, structural relaxation

PACS (2006): 61.43.Dg, 62.40.+i

1 Introduction

Amorphous metallic materials—metallic glasses—are very interesting both from a scientific and practical point of view because they present an amorphous form of condensed matter with very high deviation from thermodynamic equilibrium. During heat treatment, they undergo structural relaxation consisting of subtle rearrangements of their atomic structure connected with some densification. Structural relaxation phenomena are associated with changes in the short-range ordering of both chemical and structural

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types [1], and there is a decrease in enthalpy associated with these processes. Changes in topological short-range order largely involve redistribution of free volume within the glass, and changes in chemical short-range order involve rearrangements of the atomic chemical species at the sites. These changes produce significant variations in many physical-mechanical and magnetic properties [2–9].

In addition, there is a hierarchy of internal stresses of different ranges in amorphous metallic materials obtained by the rapid quenching process. This hierarchy consists of: (i) macroscopic quenching stresses (acting on the scale of the whole sample) [10], (ii) submacroscopic quenching stresses (acting on the scale of several hundredths of micrometers) [11] and (iii) local stresses of intercluster boundaries [12] or atomic level stresses [13]. At elevated temperatures, these stresses disappear during the structural relaxation process. When external mechanical stress is applied, the structural relaxation process is influenced.

At temperatures much lower than the glass transition temperature T_g , the metallic glasses 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 anelastic strain recovery [14, 15]. Anelasticity in metallic glasses is a process distributed over a range of activation energies [16–18]. Taub and Spaepen [19] first noticed that the anelastic deformation response of metallic glasses 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. This concept has been proposed for amorphous metallic materials in [20]. A study of the activation energy spectra is a possible help in understanding the atomic processes which take place during the relaxation in metastable systems. The strain recovery rate directly reflects the internal atomic rearrangement and the analysis of strain rate kinetics can provide important information about the glassy state.

It has been shown that three different creep stages in strain rate-time dependences in metallic glasses can be distinguished quite unambiguously [21]. The origin of these stages is quantitatively explained within the framework of the model of structural relaxation in an external stress field [22], which takes into account reversible (for the first stage) and irreversible (for the second and third stages) atomic rearrangements in ‘relaxation centers’. Creep rate kinetics is determined not only by temperature and applied stress, but also depends on the thermal prehistory of the metallic glass. Whereas the first and second creep stages are considerably influenced by the structural state, the last stage is characterized by the absence of thermal prehistory memory [21].

The aim of this work is to report some results on creep strain recovery and structural relaxation processes in Fe–Ni–B metallic glass after longtime loading derived from DSC and TMA studies, and to demonstrate how the activation energy spectra model is appreciated for the description of creep strain recovery process in the material.

2 Experimental method and method of analysis

An amorphous ribbon of the nominal composition $\text{Fe}_{40}\text{Ni}_{41}\text{B}_{19}$ with a thickness of $17.3\ \mu\text{m}$ was used in the experiments. The crystallization temperature of the amorphous ribbon was $T_x = 415\ ^\circ\text{C}$, estimated by differential scanning calorimetry for the as-quenched sample at the temperature of onset of the first crystallization peak. The specimens of 4 mm width were cut from as-received ribbon, heated up to the different annealing temperatures T_a from 150 to $300\ ^\circ\text{C}$ and annealed for 20 hours under an 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 room temperature and then unloaded. Samples annealed under the same conditions but without the 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 an absolute resolution of 10 nm. The enthalpy and length changes were measured under linear heating with a constant heating rate β of 20 and $10\ \text{Kmin}^{-1}$, respectively, for both the stress-annealed and the reference samples. A small tensile stress of about 1 MPa was applied to the sample during the constant heating rate experiment. A flow of pure nitrogen was used to protect the samples. Temperature stability was better than 0.2 K.

The dilatometric data obtained were analyzed using the simple method for calculating the spectrum of activation energies $N(E)$ from a tempering experiment that is a linear time-temperature dependence: $T = T_o + \beta t$, where T_o is the starting temperature of annealing and t is the time. The activation energy spectrum (AES) model introduced by Gibbs et al. [20] and developed by Stulens et al. [23] is based on the equation:

$$\gamma_A(T) = \int_0^{+\infty} N(E) \frac{(\alpha_1 T + \alpha_2 T^2) - E}{k\bar{T}} dE, \quad (1)$$

where $\bar{T}$ is the average temperature during heating, k is the Boltzmann constant and $\gamma_A(T)$ is the anelastic shear strain:

$$\gamma_A(T) = \sqrt{3} \frac{\Delta l}{l_o}. \quad (2)$$

In this relation, Δl is the length change measured at a temperature T in our experiment, and l_o is an effective length of the sample ($l_o = 15\ \text{mm}$). Eq. 1 and the parameters α_1 and α_2 were chosen in such a way as lead to the exact activation energy spectrum AES according to [23]. This fact results in the following values for α_1 and α_2 :

$$\alpha_1 = (k/b)(-a - 1 + \ln(\nu_o \bar{T}/\beta)), \quad (3)$$

$$\alpha_2 = k/b\bar{T}, \quad (4)$$

where ν_o denotes the attempt frequency, and a and b are constants ($a = 2.49663, b = 1.03197$). This modern method without prior assumptions about the shape or other

properties of the spectrum uses fast Fourier transformation to determine the activation energy spectrum from the non-isothermal experimental data. The method has been described and tested in more detail in our earlier paper [24].

3 Results and data analysis

Metallic glasses prepared from melting by a rapid quenching method are in a metastable state and at elevated temperatures, structural relaxation takes place where the atoms rearrange to a more stable amorphous state. During longtime annealing, structural relaxation proceeds and in the case of a sample subjected to stress-annealing, creep also occurs. The creep strain is more intensive at a higher annealing temperature and therefore after the same time of annealing at different temperatures, states with different final creep deformation are produced. During a subsequent linear heating when creep strain

Fig. 1 DSC traces recorded for the samples preannealed at the indicated temperatures under and without stress, and the differences between them.

Fig. 2 DSC traces recorded for the samples stress-annealed at the indicated temperatures.

recovery occurs, DSC and TMA traces can reveal a difference in energy release between the samples preannealed at the same temperature T_a with and without applied stress.

Typical DSC curves measured for samples annealed at some temperatures T_a (175, 200, 250, 275 °C) are shown in Fig. 1. The curves recorded for samples subjected to stress-annealing were compared with the curves measured for the reference samples. The difference between them is also depicted. The onset of crystallization is denoted as T_x in accordance with DSC measurements. The DSC traces measured for samples subjected to stress-annealing and for the reference samples have a similar shape at a given annealing temperature. Thus the structural relaxation that takes place during heating is qualitatively the same for all the samples—preannealed under stress and without stress. DSC traces only start to have a different deviation at a temperature near 200 °C at a given heating rate for all the temperatures of annealing T_a . The more significant changes associated with structural relaxation and so with the rearrangement of the initial structure can be seen at the temperatures about $T_a + 100$ °C at a given heating rate when the energy accumulated during the creep strain starts to release. At temperatures above 400 °C, the crystallization process begins, which has generally more stages and is accompanied by much more extensive release of energy.

DSC traces recorded for the stress-annealed samples at different temperatures from 150 to 300 °C are shown in Fig. 2. Each of the measured curves obtained at the constant heating rate clearly shows an exothermic effect. The wide exothermic decreases are present for all annealing temperatures. The starts of the decreases tend to shift towards high temperatures as the stress-annealed temperature increases. Fig. 3 shows the differences of measured DSC data between the samples preannealed at indicated temperatures

Fig. 3 The differences of measured DSC data between the reference sample and the sample stress-annealed at the indicated annealing temperatures.

under and without stress. There is no sequence with the temperature of annealing. However it can be said that annealing under stress causes in general more intensive structural relaxation and so a closer structure arrangement.

Fig. 4 The anelastic shear strain for the samples stress-annealed at the indicated temperatures.

Fig. 5 Creep recovery spectra $N(E)$ calculated from the linear heating experiments in Fig. 4.

The anelastic shear strain measured under linear heating as a relative length change of the samples stress-annealed at different temperatures after the subtraction of the linear elongation of the samples due to the thermal expansion is given in Fig. 4. Creep strain recovery manifests itself in a relative shrinking of the stress-annealed samples in comparison to the reference ones. This deflection from the conventional ‘linear expansion’ behaviour starts at a temperature near the annealing temperature and corresponds to the strain recovery onset [15, 25]. Annealing under applied stress leads to different amounts of total deformation. The recovered anelastic strain was found to be up to 5×10^{-3} which is in agreement with the results published elsewhere [8, 24].

The activation energy spectra were calculated from the non-isothermal anelastic shear strain experimental data shown in Fig. 4 using a modern method based on Fourier technique. The results of our calculations can be seen in Fig. 5. Creep recovery spectra have a discrete character consisting of a finite number of peaks with well defined characteristic activation energies. Each of the peaks probably represents a different type of deformation defect in the amorphous structure. These peaks can be considered as evidence of definite relaxation mechanisms that may be related to the different type of ‘flow defects’ in the amorphous structure. It is evident that the creep strain recovery is determined by the temperature of stress-annealing. The height of peaks in the calculated creep recovery spectrum tends to increase with increasing activation energy for a given stress-annealing temperature.

The positions of the two most significant peaks depending on the stress-annealing temperature are shown in Fig. 6. Two tendencies of peak position dependence on the an-

Fig. 6 Peak positions depending on the annealing temperature. The lines are depicted only to orient the eye.

nealing temperature are evident. For lower temperatures of annealing, the characteristic energy of peaks decreases as the stress-annealing temperature increases, while for higher stress-annealed temperatures, the opposite tendency is observed. This can be connected with the different structural states of the samples obtained during stress-annealing at different temperatures.

Metallic glasses are produced by rapid melt quenching and, therefore, are highly non-equilibrium structures. This is the origin of irreversible local atomic rearrangements that arise as the result of any heat treatment and are generally referred to as structural relaxation. Recently, a model treating the homogeneous plastic flow of metallic glasses as a result of structural relaxation oriented by external stress was proposed by Khonik et al. [22]. According to the model, called the Directional Structural Relaxation (DSR) model, non-isothermal strain recovery can be interpreted as a set of mainly irreversible local atomic rearrangements, with a distributed activation energies spectrum, in spatially separated regions of the structure called ‘relaxation centers’ oriented favorably or unfavorably to the external stress [15].

As structural relaxation depends on the stress-annealing temperature, it can be said that in the samples stress-annealed at lower temperatures, both relaxation centers, the parallel and the antiparallel in sign to the external stress, rearrange during the strain recovery process. As the annealing temperature increases, the influence of antiparallely oriented relaxation centers decreases; thus for higher temperatures, only parallelly oriented relaxation centers contribute to the creep strain recovery process. Therefore, the opposite tendencies of the characteristic energies of creep recovery for samples pre-annealed at lower and higher temperatures can be associated with cooperation between

both relaxation centers, the parallel and the antiparallel in sign to the external stress at lower annealing temperatures. This also influences total shrinkage during creep strain recovery. Rearrangements of both the favorably and unfavorably oriented relaxation centers are connected with lowering the free energy of the amorphous structure towards an equilibrium glassy state, and manifests itself in an exothermic decrease in the DSC traces. The energy accumulated during creep strain starts to release at temperatures around $T_a + 100$ °C. It is evident that creep strain recovery is determined by the temperature of stress-annealing.

4 Conclusion

Different creep strain accumulation is realized during the stress-annealing of Fe-Ni-B amorphous ribbon depending on the annealing temperature. This fact influences the structural relaxation and creep strain recovery processes in the metallic glass. Structural relaxation is qualitatively the same for samples preannealed under and/or without stress. Both relaxation centers, the parallel and the antiparallel in sign to the external stress, rearrange during creep 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 creep strain recovery process in the Fe-based amorphous ribbon.

Acknowledgment

The authors are grateful to Dr. P. Duhaj for providing the amorphous ribbon and to the Slovak Grant Agency for Science (grant No. 2/5045/25) for support of this work.

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