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Relaxation studies of amorphous alloys with creep induced magnetic and structural anisotropy

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Amorphous ferromagnetic (Fe,Co,Ni) ribbons of various compositions have been prepared by rapid solidification and annealed with applied tensile stress. This process yields both creep-induced magnetic anisotropy and structural anisotropy. Post-annealing has been done to investigate the relaxation process. X-ray diffraction and thermomechanical analysis measurement of post-annealed samples reveal a clear underlying relaxation process in the material, proving that structural anisotropy corresponds to the elastic strain induced by creep annealing.

Keywords: Annealing; Residual elastic strain; Heterogeneity in amorphous structures; Thermomechanical analysis

Creep-induced anisotropy of amorphous magnetic ribbons has been known since the first use of such ribbons [1–3]. It is used, for example, to fine-tune the magnetic properties for sensor applications [4–7]. However, the origin of creep-induced anisotropy has not been fully understood because of insufficient structural information. Recently, anisotropy of halo positions in amorphous metals has been detected by X-ray diffraction (XRD) in transmission geometry [8] by measuring the difference between the halo top positions of samples aligned parallel and perpendicular to the ribbon's rolling direction. The origin of this anisotropy has been interpreted in terms of the constrained strain induced during heating under tensile stress. In addition, a difference in shrinkage was observed by post-heating samples annealed under different stress levels, which suggests the existence of the strain. These observations suggest that the constrained strain has an elastic origin. However, these processes can also be linked to the intrinsic structural relaxation related with the original amorphous structure. To separate the contribution of creep-induced anisotropy from the intrinsic relaxation process, it is necessary to investigate the mechanism of the relaxation process and creep-induced anisotropy in greater detail. The purpose of this work is thus to analyze in detail the relaxation processes in amorphous materials with structural anisotropy and to confirm whether the constrained strain is truly of elastic origin.

Amorphous, ferromagnetic (Fe,Co,Ni)-based ribbons of various compositions (see Table 1) have been produced by rapid solidification from the melt. The ribbons were annealed at 350 °C for 30 s under a tensile stress σ_a along the ribbon axis with a magnetic field of 1 kOe applied simultaneously across the ribbon width [8]. Samples representative of each composition were annealed at two different stresses: low σ^{small} and high σ^{large}. Table 1 shows the values of the applied stress as well as the crystallization temperature (T_x). The latter was determined from thermomechanical analysis (TMA) measurements with a heating rate of 20 K min^{-1}.

The X-ray measurements were performed using a conventional XRD system in transmission geometry (t-XRD), with an Mo Kα line (18 kW; 60 kV–300 mA) in $\theta/2\theta$ scan. The incident X-ray beam was aligned at an angle θ from the normal vector of the ribbons and

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Table 1. Compositions in at.% (σ, applied stress during annealing at 350 °C for 30 s; T_x, crystallization temperature).

Alloy	Composition	σ^{small} (MPa)	σ^{large} (MPa)	T_x (°C)
I	$Fe_{40}Ni_{40}Mo_4B_{16}$	37	328	422
II	$Fe_{24}Ni_{58}Mo_2B_{16}$	34	308	373
III	$Fe_{24}Co_{12}Ni_{46}Si_2B_{16}$	32	293	409
IV	$Co_{56.5}Fe_6Mn_1Ni_{16}Si_4B_{16.5}$	43	364	437
V	$Co_{72}Fe_1Mn_4Mo_1Si_{13}B_9$	30	254	505
VI	$Co_{72.5}Fe_5Si_{5.5}B_{17}$	39	353	489
VII	$Co_{69.5}Fe_{3.5}Mo_3Nb_1Si_{16}B_7$	30	249	547
VIII	$Co_{72.5}Fe_{1.5}Mn_4Si_5B_{17}$	38	332	476
IX	$Fe_{65}Co_{18}Si_1B_{16}$	37	336	427

the diffracted beam was also aligned at θ from the normal vector of the opposite side of the ribbons. This way, the diffraction vector q is in the ribbon plane. Two measurements were conducted for each sample: one for the measurement in which the diffraction vector q was parallel to the ribbon direction (in the same direction as the applied tensile stress) and the other for q parallel to the ribbon width direction (perpendicular to the stress direction). The structural anisotropy was evaluated from the difference in the halo top position, q_p, between the directions perpendicular and parallel to the ribbon using the following equation:

$$\Delta q = q_{p\perp} - q_{p//} \tag{1}$$

In order to study the relaxation processes, TMA was employed to evaluate the linear thermal expansion (LTE) of the annealed ribbons. For simplicity, the elongation $\frac{\Delta l}{l}$ is denoted as ϵ. The measurements were performed using a Rigaku TMA with a heating rate of 20 K min^{-1} under an Ar flow. In addition, the t-XRD profiles of post-annealed ribbons were measured. Post-annealing was performed using TMA with the same heating rate as in the LTE measurements.

To compare the recovery, the relaxation fraction for t-XRD measurement of Δq was defined as

$$\frac{\Delta q_{\text{without post-annealing}} - \Delta q_{\text{at given temperature}}}{\Delta q_{\text{without post-annealing}}} \tag{2}$$

and for TMA measurement as

$$\frac{\epsilon_{\sigma}^{small} - \epsilon_{\sigma}^{large}}{\epsilon_{\sigma_{T_x}}^{small} - \epsilon_{\sigma_{T_x}}^{large}} \tag{3}$$

where $\epsilon_{\sigma_{T_x}}^{small}$ and $\epsilon_{\sigma_{T_x}}^{large}$ are the values of ϵ at crystallization temperature T_x for the samples annealed at σ^{small} and σ^{large}, respectively.

As mentioned above, the relaxation phenomena were checked using two different approaches. The first approach was to evaluate constrained stress by measuring the LTE of ribbons annealed under different conditions. The second was to measure the structural anisotropy of the ribbon after post-annealing using t-XRD. Figure 1a shows the LTE curve of alloy VI annealed under σ^{small} and σ^{large} (the graph also appears in Fig. 6c of Ref. [8]). Both samples exhibit typical thermal expansion up to 200 °C, and there is no difference in their TMA curves. However, after exceeding this temperature, the samples show different behaviors. The sample annealed under σ^{small} still follows the linear trend, whereas the

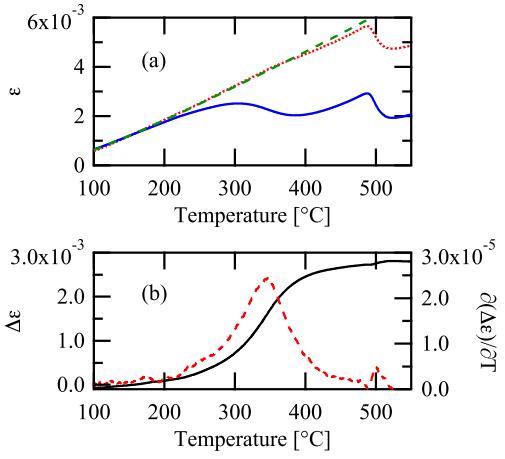

Figure 1. (a) The LTE curve for $Co_{72.5}Fe_5Si_{5.5}B_{17}$ (alloy VI) [8]. Solid line: alloy annealed under σ^{large}; dotted line under σ^{small}. The broken line is the second run after heating to 600 °C, representing the LTE of the crystalline phase. (b) Solid line: $\Delta\epsilon$; broken line: derivative of $\Delta\epsilon$.

one annealed under σ^{large} exhibits shrinkage. After reaching ~400 °C the process is stopped and the sample again exhibits linear thermal expansion. At 489 °C one can see crystallization in both samples. Comparing these results to the TMA of an already crystallized sample, one can see that sample annealed under σ^{small} shows only a small amount of intrinsic relaxation. On the other hand, the sample annealed under σ^{large} undergoes simultaneous expansion (due to the thermal expansion) and shrinkage due to the relaxation processes. Using the TMA data from Figure 1a, when we subtract the LTE curve of the sample annealed under σ^{small} from the one annealed under σ^{large}, we can extract the relaxation of creep-induced anisotropy from the sample σ^{large}. Figure 1b shows $\Delta\epsilon$, defined as ϵ of σ^{small} – ϵ of σ^{large}. It illustrates the relaxation of elastic strain, and we can distinguish three phases of annealing. In the first phase, from room temperature to 250 °C, the sample undergoes thermal expansion. In the second, between 250 and 450 °C, we observe relaxation process. In the third and final phase, from 450 °C to the end of the measurements, the sample again undergoes thermal expansion. From the derivative of temperature we can calculate its peak (342 °C), which corresponds to the maximum rate of the relaxation process.

Figure 2a shows curves for all of the data of $\Delta\epsilon$ (with the exception of sample VIII, which did not show any