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PECULIARITIES OF CURIE POINT RELAXATION IN A METALLIC GLASS

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Résumé - La relaxation structurale d'un verre métallique trempé a été étudiée par mesure de la température de Curie. L'augmentation de la température de Curie, due à la relaxation isotherme, varie de façon approximativement linéaire, avec cependant des ondulations, avec le logarithme du temps de recuit, pour des temps qui ne soient pas trop longs. Cependant, la dérivée de ces courbes présente trois pics qui correspondent à trois processus différents. Les énergies d'activation de ces processus ont été obtenues.

Abstract - The structural relaxation of an as-quenched metallic glass was investigated by Curie point measurement. The increase in Curie temperature due to isothermal relaxation of as-quenched samples exhibits approximately linear - though with slight waviness - dependence on the logarithm of annealing time for medium length of annealing. However the derivatives of these curves exhibit three peaks which correspond to three distinct processes. The activation energies of these processes were obtained.

I - INTRODUCTION

Structural relaxation of metallic glasses can be followed up by Curie point measurement. In the last few years many publications have appeared on this topic [1-9]. It was pointed out in a previous work [10] that suitable heat treatment is able to remove the non-inherent defects in an as-quenched metallic glass. The annealing out of these defects is observed as an irreversible process in structural relaxation. In this paper we intend to investigate how this irreversible process affects the kinetics of reversible relaxation.

II - EXPERIMENTAL

Relaxation processes were investigated by Curie point measurement during quasi-isothermal annealing of an as-spun metallic glass ribbon of composition $\text{Fe}_{16}\text{Ni}_{65}\text{B}_{19}$ with low Curie temperature. Quasi-isothermal annealing means that after measuring the Curie temperature the sample was heated to the annealing temperature as rapidly as possible, it was kept at that temperature for a given time, it was then cooled to room temperature as rapidly as possible. The Curie point was measured and after that annealing was performed again at the same temperature as before; and so on until the Curie temperature approached its quasi-equilibrium value corresponding to this annealing temperature.

Heating the sample from Curie temperature to the annealing temperature took about 2 minutes; about 3 minutes were necessary for the cooling to 140 °C. This means that it was not possible to apply very short annealing times which would be desirable at the starting part of the kinetics, where rapid change was observed.

Curie temperatures were obtained from temperature dependence of magnetization measured in a magnetic field of induction $B = 8 \cdot 10^{-3}$ T. Magnetization measurement was

performed using a force magnetometer made by means of a Sartorius 4434 type vacuum microbalance. The apparatus and the method of measurement are described in more detail in /10/.

III - RESULTS AND DISCUSSION

Kinetics of increase in Curie temperature of as-quenched samples were obtained at 433 K, 468 K, 493 K and 519 K. As was suggested in one of our earlier works /11/ a function with one constant and three exponential terms was fitted to the experimental points:

$$\Delta T_c(t_a) = A_0 + A_1 e^{-\alpha_1 t_a} + A_2 e^{-\alpha_2 t_a} + A_3 e^{-\alpha_3 t_a}, \quad (1)$$

where ΔT_c is the change in Curie temperature, t_a the annealing time, A_i and α_i are the maximum increase in Curie temperature and the inverse time constant respectively for the i -th process ($i=1,2,3$), $A_0 = -(A_1+A_2+A_3)$.

This choice was also motivated by the fact that according to Tewari's method /12/, i.e. deriving the change in property as a function of the logarithm of the annealing time, three peaks were found which correspond to three discrete processes (see also Fig. 2 in /10/).

Isotherms obtained at four different temperatures are plotted against the logarithm of annealing time in Figs. 1 and 2, together with their derivatives. The $\Delta T_c(\ln t_a)$ curves were calculated by the method of least squares and the points represent the experimental values. Derivatives were calculated from the parameters obtained by Eq. (1). The separation of these curves into two figures is not solely a practical matter: it has a deeper meaning, viz. the two isotherms of lower temperature exhibit qualitatively quite different behaviour than those of the two higher temperatures. This is more obvious if one looks at the derivatives. It seems that there are two temperature intervals in which the course of relaxation differs qualitatively. This observation is in agreement with the results of Chen et al. /13,14/ who were the first to observe two temperature intervals with different relaxation behaviour.

Fig. 1 - Change in Curie temperature due to isothermal annealing of as-spun sample vs logarithm of annealing time. The derivatives of the isotherms are also presented. The annealing temperature were 433 K and 468 K.

It is seen from Figs. 1 and 2 that each derivative curve consists of three peaks which indicate the presence of three processes. The places of maxima at the derivatives are obviously connected with the time constants of different relaxation processes but these correlations are influenced by the extent of maximum change for different processes, and so the place of maxima do not correctly characterize the

relaxation. Instead, the amplitudes and the time constants of the three processes (playing a role in Eq. (1)) are more convenient for this purpose.

Fig. 2 - As for Fig. 1 but with annealing temperatures of 493 K and 519 K.

Fig. 3 - Amplitudes of the three processes vs annealing temperature.
• first process, x second process,
Δ third process

Fig. 4 - Logarithms of inverse time constants for the three processes vs inverse temperature

The temperature dependence of the amplitudes A_i of the relaxation processes is shown in Fig. 3. It is seen that the extent of increase in Curie temperature increases with annealing temperature for the first process (A_1) and decreases for the other two (A_2 and A_3). The first point in the second process deviates from

this tendency (see dashed line), this is attributable to the fact that at the lowest temperature the kinetics curve would have been measured for a longer time than was actually done.

In Fig. 4 the logarithms of the inverse time constants are plotted against the inverse annealing temperature. It seems reasonable to fit linear functions to each process for the whole temperature range. Similarly to the case of amplitudes there is also a deviation from the general tendency at the lowest annealing temperature, but in this case for the first process (see dashed line). The scatter of the points indicated in the figure was calculated from the fit of Eq. (1) to the increase in Curie temperature measured with an accuracy of ± 0.05 K. The solid lines in Fig. 4 were calculated by the method of least squares and from the slope of these lines activation energies were obtained for the three processes: $E_1 = (37.6 \pm 3)$ kJ/mol; $E_2 = (41.5 \pm 1)$ kJ/mol; $E_3 = (29 \pm 2)$ kJ/mol. One of them must be activation energy of the irreversible process. If we take this to be E_1 , then E_2 and E_3 correspond to the reversible processes. Comparing these values with those obtained for preannealed samples /10/ ($E_1 = 41$ kJ/mol, $E_2 = 65$ kJ/mol) it is seen that, due to the irreversible process, at least one of the activation energies for reversible relaxation decreases.

IV - CONCLUSIONS

The derivative of the increase in Curie temperature vs the logarithm of annealing time during quasi-isothermal annealing of an as-spun metallic glass consists of three peaks which indicate the existence of three distinct relaxation processes.

From the plot of $\ln \alpha_i$ vs T^{-1} for the three processes three activation energies could be obtained. One of them was attributed to the irreversible process.

Comparing the activation energies of the reversible processes with those obtained on measuring the pre-annealed samples /10/ we can conclude that the presence of an irreversible process makes the reversible relaxation easier.

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