Nanocrystallization of Finemet Alloy after Intensive Plastic Deformation

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Abstract. The Finemet alloy prepared by the crystallization of an amorphous precursor is a basic model material for study of magnetic nanoparticle structures. It was shown that intensive plastic deformation localized into adiabatic shear bands causes intensive heating and structural changes in the amorphous alloy. These changes influence the magnetic properties of the amorphous precursor and the Finemet crystallization. Direct creep measurements during the crystallization of Finemet alloys were performed and the creep properties of the residual amorphous phase formed during the nanocrystallization were described. It was shown that due to relatively high temperatures the residual amorphous phase undergoes intensive structural relaxation resulting in the obvious embrittlement of these materials.

1. Introduction
In recent years the range of available soft magnetic materials has been significantly increased by the development of nanocrystalline magnetic materials beginning with Yoshizawa et al. [1]. The most widely investigated Finemet alloy is Fe\textsubscript{73.5}Nb\textsubscript{3}Cu\textsubscript{13.5}Si\textsubscript{13.5}B\textsubscript{9} which is produced by rapid solidification and then annealed above the crystallization temperature to produce the nanocrystalline structure [2]. The disadvantage of preparation procedure of amorphous ribbons is their form. The plastic deformation of metallic glasses is carried out via the shear band creation and the localized shear in these bands. It was shown that catastrophic shear band creation is done in nearly adiabatic conditions. The stored elastic energy releases by local heating of material in the thin layer of the shear band. At the final stage of deformation the manifestations of local heating are observed as dimples or needles at fracture surface on the failed metallic glasses [3]. It was observed that high amount of plastic deformation energy introduced by intensive milling leads to partial or full crystallization of amorphous alloys [4, 5].

The well-known disadvantage of these materials is their brittleness. Because their structure consists of the hard Fe-rich nanoparticles enclosed in amorphous matrix it is possible to conclude that the macroscopic brittleness is caused by the properties of amorphous matrix. Amorphous alloys during heating can overcome the glass transition temperature above which the viscosity of metallic glass decreases. In this state there are good conditions for the homogeneous deformation. Khonik et al. [6] have observed that at heating rate about 10 K/min the glass transition temperature can be reached below the onset crystallization temperature of Finemet metallic glass.
2. Experimental

It is well known that during the crystallization of amorphous Fe$_{73.5}$Nb$_3$Cu$_1$Si$_{13.5}$B$_9$ alloys, the first crystallization is associated with the ferromagnetic Fe-Si $\alpha$-phase production. The influence of the structural changes on the crystallization was studied by means of thermal analysis equipment Setaram TGDTA92 adapted to faraday type balance. Structural changes were performed by repeated impacting of small hummer during 30 s (def1) for the low deformation state, 10 min (def2) and 30 min (def3) for the high deformed structure. The length changes of Finemet type amorphous ribbons Fe$_{73.5}$Nb$_3$Cu$_1$Si$_{13.5}$B$_9$ with dimensions of 20 $\times$ 1.3 $\times$ 0.027 mm$^3$ were measured under uniaxial tensions of 2.5 MPa and 87.5 MPa during linear heating at the rate of 10 K/min in flowing Ar using thermomechanical analyser Setaram TMA92 with the resolution of 0.01 $\mu$m. The viscosity was determined using the method described in [7]. This method is based on measuring of the sample elongation $\Delta l(t, T, \sigma)$ during linear heating under different stresses $\sigma$:

$$\Delta l(t, T, \sigma) = l_0 \int_{T_0}^{T} \alpha(T) dT + l_0 \sigma \int_{T_0}^{T} \frac{\alpha(T)}{E(T)} dT + l(T) K \sigma \int_{0}^{\ell(T)} \frac{1}{\eta(T)} dt. \quad (1)$$

The first term corresponds to the thermal dilatation of the sample with the length $l_0$ and the dilatation coefficient $\alpha(T)$. The second term is given by the elasticity under applied load $\sigma$. The last term reflects the flow of the sample under applied load, $K$ being a numerical constant. Considering the small temperature dependence of elastic properties it is possible to eliminate the first and the second terms in eq. (1) by repeating the identical experiment under high and low applied stresses.

3. Results and discussion

The thermomagnetic traces for the original as quenched and three different deformed states measured at linear heating rate of 20 K/min are shown in Fig. 1. The transitions to the paramagnetic state in amorphous structure are observed. The temperature interval of the Curie transition is independent on the used structural states. The different magnetic forces for samples with different structure at lower temperatures can be caused by the structural changes in shear bands and by the presence of the introduced stress field as well as by different structural relaxation of amorphous structure. In addition, the influence of the demagnetization factor by

![Figure 1](image1.png)

**Figure 1.** The temperature dependences of magnetization for undeformed sample and samples in different deformed states.

![Figure 2](image2.png)

**Figure 2.** The temperature dependence of the magnetization of high deformed sample at different heating rate.
sample shape changes is present. The production of ferromagnetic Fe-Si $\alpha$ phase above 800 K causes the increasing of magnetic force, meanwhile by next increasing the sample temperature the transition to paramagnetic state of the crystalline phase occurs. During next cooling the transition to the magnetic state manifested by increasing of the magnetic force is independent on the structural state. Figure 2 suggested the kinetics of crystallization at different heating rates. Lowering the heating rate decreases the temperature of crystallization, and crystallization peak becomes larger. The difference in thermomagnetic curves during cooling is caused by different degree of transformed volume and small differences in the structure of Fe-Si $\alpha$ phase.

Measured length changes of samples under low and high loading during heating and cooling are shown in Fig. 3. In the case of low applied stress the linear expansion of the sample during heating changes to the shrinkage at temperatures above the onset crystallization temperature $T_x$ due to higher specific density of crystals. Under high stress the deformation rate increases with the temperature (homogeneous creep deformation) up to the onset crystallization temperature $T_x$. During crystallization the decrease of deformation rate is observed. The dilatation curves measured during cooling are similar for low and high stresses, no remarkable creep is present. Figure 4 shows the crystalline fraction determined from the relative shrinking of the sample under low stress. The viscosity of amorphous ribbon during heating decreases through the classical Arrhenius expression. The presence of hard particles in Newtonian liquid causes the increase of the viscosity. Several formulae have been derived to the description of the influence of particle amount on the viscosity of the composite [8].

The measured temperature dependence of the strain rate for Finemet alloy in narrow temperature interval in the vicinity of the onset crystallization temperature $T_x$ can be seen in Fig. 5. This dependence was fitted using the power formula similar to Dryden [9]:

$$
\dot{\varepsilon} = 0.68 \dot{\varepsilon}_{\text{amorphous}}(1 - \Phi)^{2.5},
$$

(2)

where $\dot{\varepsilon}_{\text{amorphous}}(T)$ was expressed through Arrhenius law. We suppose that the nature of amorphous phase during the crystallization is unchanged and the contribution of crystalline particles to the deformation rate can be neglected due to their high viscosity.

The dependence of viscosity on the crystalline fraction shown in Fig. 6 is non linear. The lowest estimated value of viscosity $\approx 8 \times 10^{12}$ is the evidence that the glass transition temperature of Finemet glass was not reached. Observed dramatic increase of viscosity with the crystalline

**Figure 3.** The temperature dependences of sample length changes under indicated loading during heating and cooling.

**Figure 4.** The temperature dependence of the crystalline fraction in the sample during crystallization.
Figure 5. The temperature dependence of the deformation rate in the vicinity of the onset crystallization temperature.

Figure 6. The dependence of the viscosity on the crystalline fraction during heating of Finemet sample.

Fraction is due to the presence of crystalline particles and at the same time due to the structural relaxation of the amorphous matrix. At relatively high temperatures there are good conditions for structural relaxation of amorphous residuum, the structural relaxation is intensive and the brittleness of Finemet is observed.

4. Conclusion

We studied the influence of the plastic deformation via creation of shear bands on the crystallization of Finemet alloy. The thermomagnetic study has shown that even small deformation leads to the change in the magnetization. During heating the structural relaxation occurs and the contribution of the highly modified structure inside shear bands vanished and no differences in the crystallization products were observed. Viscosity measurements under isochronal conditions during the crystallization of Finemet type amorphous alloys have shown that no glass transition or softening of amorphous phase was observed at the heating rate of 10 K/min. The dependence of the viscosity on the crystalline fraction $\Phi$ was found to be as $\eta \approx (1 - \Phi)^{-2.5}$.

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