ON THE MAGNETIC ANISOTROPY DISTRIBUTION IN THE SURFACE REGION OF THE CONVENTIONAL AMORPHOUS WIRES

BY

MUGUR B. RĂUŢ*

Abstract. In-water quenching technique, which is the preparation procedure of conventional amorphous wires (CAW), induces internal stresses in the material. These stresses, coupled with magnetostriction, give rise to large magnetoelastic anisotropies. Using the calculated distribution of internal stresses, the aim of this work is to evaluate the theoretical distribution of magnetoelastic anisotropies of CAW with positive, negative and nearly zero magnetostriction. The anisotropy constants were calculated as functions of wire dimensions, taking into account that the influence of wire length is neglected. Consequently, we elaborate two simple calculation programs which enable us to calculate the magnetoelastic distribution in every point of wire radius, for any magnetostriction, positive, negative or nearly zero, and any values of parameters and physical quantities involved.

Key words: magnetic anisotropy; conventional amorphous wires; internal stress.

1. Introduction

Conventional amorphous wires are prepared by the in-water quenching technique. This procedure has a very high cooling rate from the molten alloy and introduces internal stresses within the conventional amorphous wires (CAW). These stresses, which couple with material magnetostriction, give rise to large magnetoelastic anisotropies. The distribution of these anisotropies determines the domain structure and magnetization process of CAW. The aim of this paper is, first of all, to evaluate the distribution of magnetic anisotropies and compare the theoretical results with experimental results for CAW anisotropies. This could not be possible without the works (Velasquez *et al.*, 1991) which have established the distribution of internal stresses in CAW,

*Corresponding author; *e-mail*: m_b_raut@yahoo.com

(Severino *et al.*, 1992) and, last but not least, our own work for the experimental evaluations, (Răuţ *et al.*, 2006).

2. General Theory

The absence of long-range order, which is a property of magnetic amorphous materials, implies an absence of magnetocristaline anisotropy. Consequently, magnetoelastic anisotropy and the anisotropy induced by form are the main causes which the magnetization processes in these types of materials are based on. The form is a geometrical factor, thus it could be properly chosen in order to minimize the demagnetizing field or neglect it. So, the only one that exhibits some interest in this matter is the magnetoelastic anisotropy. This anisotropy originates in coupling between the internal stresses introduced by fabrication process and magnetostriction. Therefore it depends on value and distribution of internal stresses and on the intensity of magnetoelastic coupling with magnetostriction. By a proper choose of the composition we could have some control concerning the magnetostriction constant of the alloy we have working with, but in internal stresses case we don't have it at all. These stresses are a consequence of the rapid quenching process and not depend in most of cases on composition. Their dependence is on the cooling rate and on the temperature gradient within CAW during the rapid solidification process. The distribution of magnetoelastic anisotropy is the main factor which decides the configuration of magnetic domains; this is the reason why the magnetization processes are directly influenced by it.

In order to obtain the expression of the magnetoelastic anisotropy constant, let's consider a cylindrical sample.

If we apply this sample certain exterior mechanical stresses, the additional volumetric energy density due to sample deformation is:

$$W_\sigma = \sum_{i,j=1}^{3} e_{ij}\sigma_{ij} \tag{1}$$

where: e_{ij}, i,j = 1,2,3 is the deformation tensor and σ_{ij}, i,j = 1,2,3 the stress tensor.

In order to simplify the future calculations we will consider the mechanical stresses uniformly distributed within sample.: $\sigma_{ij} = \sigma$. The direction σ has the director cosines γ_i cu i = 1,2,3, relative to the crystal principal axes. For simplicity reasons we assume also that the crystal has cubic simmetry. Under these conditions:

$$\sigma_{ij} = \sigma\gamma_i\gamma_j , \tag{2}$$

the elastic deformations will be:

$$e_{ii} = \sigma[s_{11}\gamma_i^2 + s_{12}(\gamma_j^2 + \gamma_k^2)]$$
$$e_{ij} = \sigma s_{44}\gamma_i\gamma_j \text{, with } i\neq j\neq k=1,2,3 \tag{3}$$

where s_{ij} are the elastic constants of the crystal. Equations (3) are valid under the hypothessis that deformations and stresses are proportional.

According to the above considerations and the fact that the energy (1) is, after all, a magnetostriction energy, we can write it as:

$$\begin{aligned} W_\sigma = B_1\sigma(s_{11} - s_{12})[\alpha_1^2\gamma_1^2 + \alpha_2^2\gamma_2^2 + \alpha_3^2\gamma_3^2 - 1/3] + \\ B_2\sigma s_{44}(\alpha_1\alpha_2\gamma_1\gamma_2 + \alpha_2\alpha_3\gamma_2\gamma_3 + \alpha_3\alpha_1\gamma_3\gamma_1) \end{aligned} \tag{4}$$

where α_i, i = 1,2,3 are the director cosines of the vector M_s (saturation magnetization), *i.e.* apparent anisotropy direction, and $B_{1,2}$ are two constants with different expressions depending on the type of the crystalline lattice with cubic simmetry.

The form (4) is obtained from magnetostriction energy general expression customizing:

$$\begin{aligned} W_{me} = B_1[e_{11}(\alpha_1^2 - 1/3) + e_{22}(\alpha_2^2 - 1/3) + e_{33}(\alpha_3^2 - 1/3)] + \\ B_2(e_{12}\alpha_1\alpha_2 + e_{23}\alpha_2\alpha_3 + e_{31}\alpha_3\alpha_1) \end{aligned} \tag{5}$$

Consider that the sample is uniformly magnetized after two directions, (100) and (111), the most general case. Under these conditions, the relative elongations related to the two directions will be:

$$\begin{aligned} \lambda_{100} = -\frac{2}{3}\frac{B_1}{c_{11} - c_{12}} \\ \lambda_{111} = -\frac{1}{3}\frac{B_2}{c_{44}} \end{aligned} \tag{6}$$

where λ_{100} and λ_{111} are the magnetostriction constants and c_{ij} are elasticity moduli from tensorial expression.

Using the relations between elastic constants and elasticity modulii:

$$s_{11} + 2s_{12} = \frac{1}{c_{11} + 2c_{12}}$$
$$s_{11} - s_{12} = \frac{1}{c_{11} - c_{12}} \qquad (7)$$
$$s_{44} = \frac{1}{c_{44}}$$

and the relations (6), the expression (4) takes the form:

$$W_\sigma = -\frac{3}{2}\lambda_{100}\sigma\alpha_i^2\gamma_i^2 - \frac{3}{2}\lambda_{111}\sigma\alpha_i\alpha_j\gamma_i\gamma_j \qquad (8)$$

with sum after i, j = 1,2,3 and i ≠ j.

If the material has a positive anisotropy constant then the magnetization M_s is orientated after (100) direction. Under these circumstances $\alpha_i = 1$ and $\alpha_j = 0$, also the expression (8) becomes:

$$W_\sigma = -\frac{3}{2}\lambda_{100}\sigma\gamma_i^2 \qquad (9)$$

with i = 1,2,3 .

But if the material has a negative anisotropy constant then the direction of the magnetization vector coincides with (111). From the fact that $\alpha_i = \alpha_j = 1/\sqrt{3}$ it rezults:

$$W_\sigma = -\frac{1}{2}\lambda_{111}\sigma\gamma_i\gamma_j \qquad (10)$$

with i,j = 1,2,3 si i ≠ j.

If φ is the angle between the magnetization direction and the stress apllication direction then we have:

$$\cos\varphi = \alpha_i\gamma_i \qquad (11)$$

with sum after i = 1,2,3. From equations (10) and (11) we obtain:

$$W_\sigma = -\frac{3}{2}\lambda_{111}\sigma \cdot \cos^2\varphi \qquad (12)$$

Consider now the general expression (8) and costumize it for the amorphous materials case. We suppose that magnetostriction is isotropic, $\lambda_{100} = \lambda_{111} = \lambda$. It rezults:

$$W_\sigma = -\frac{3}{2}\lambda\sigma\cos^2\varphi \tag{13}$$

which is the expression of the volumetric energy density due to deformation.

The most general expression for the anisotropy energy (obtained by neglecting high order terms), is:

$$W = -k\cos^2\varphi \tag{14}$$

In the amorphous materials case the anisotropy energy is related to magnetostriction values. For this reason, comparing the expressions (13) and (14) we find for the amorphous materials anisotropy constant:

$$k = \frac{3}{2}\lambda\sigma \tag{15}$$

This is a very important result because underlies the anisotropy constant calculus. Except this fact, the form (15) successfully applies to materials with relative low magnetostriction, (Squire & Atkinson, 1995).

In concrete situations the anisotropy direction is determined by the following reasoning: if $\lambda\sigma > 0$, the minimum energy orientation of magnetization vector coincides with stress direction; if $\lambda\sigma < 0$, then the magnetization vector is perpendicular to stress direction.

3. Particular Case

We present in the following a model for anisotropy distribution of the conventional amorphous wires. A general model, for all types of amorphous wires, is impossible yet, because the anisotropy distribution is calculated different, depending on the type of the wire. Under these conditions, a general algorithm for anisotropy distribution calculation can't exist. The anisotropy distribution is established from case to case, according to (15), where λ is the magnetostriction constant and σ is the mechanical stress within material. The magnetostriction constant is usually known, being a material characteristic, the magnetic anisotropy distribution is therefore given by the mechanical stresses distribution, which originates in the fabrication process of these wires.

We go forwards and evaluate the stress distribution in amorphous wires.

Consider that these wires have the reduced radius ε and the temperature gradient, which is the physical cause of the stress, it propagates only in radial direction. For simplicity we will use the cylindrical coordinates to solve this problem. So, in accordance with elasticity theory, we have three component of stresses: an axial component $\sigma_z^*(x,\varepsilon)$, a radial component $\sigma_r^*(x,\varepsilon)$ and an angular one $\sigma_\theta^*(x,\varepsilon)$. Here x is the reduced coordinate $\left(\mathrm{x}=\frac{\mathrm{r}}{\mathrm{R}}\right)$, R being the wire radius and r the radial cylindrical coordinate.

The evaluation of the stress expression is made by considering that the solidification process is carried out in successive steps from the surface to the interior, after radial direction and uniformly. Heat conduction equation is solved according to the initial conditions, which are different from case to case. What we obtain is the temperature distribution within the wire. Considering this distribution, we obtain the stress distribution as a result of preparation process.

Taking into consideration the stress distribution we can obtain the magnetic anisotropy distribution as a result of stress-magnetostriction coupling.

According to the above considerations this coupling is made differently from case to case.

On this basis we treat in the following the conventional amorphous wires case, according to (Velasquez *et al*., 1991). Starting with equation (15) we will calculate the mechanical stress distribution. Taking this into account we have:

$$\sigma_z^*(x,\varepsilon) = kE/(1-\upsilon)[2/(1-\varepsilon^2)\int_\varepsilon^1 xT(x)dx - T(x)]$$

$$\sigma_r^*(x,\varepsilon) = kE/(1-\upsilon)(1/x^2)[(x^2-\varepsilon^2)/(1-\varepsilon^2)\int_\varepsilon^1 xT(x)dx - \int_\varepsilon^x xT(x)dx] \quad (16)$$

$$\sigma_\varphi^*(x,\varepsilon) = kE/(1-\upsilon)(1/x^2)[(x^2+\varepsilon^2)/(1-\varepsilon^2)\int_\varepsilon^1 xT(x)dx + \int_\varepsilon^x xT(x)dx - x^2T(x)]$$

By integrating the expressions (16) we will get the stress distributions corresponding the three cylindrical coordinates, exactly which is what we are interested. This integration must be done as mentioned above, according how the solidification of the melt is taking place. In other words, depending on how heat propagates in the material. This fact is taking into account by solving the heat conduction equation:

$$\frac{\partial T}{\partial t} = \frac{D}{r}\frac{\partial}{\partial x}\left(r\frac{\partial T}{\partial x}\right) \quad (17)$$

where $T = T(x,t)$ is the temperature in a random point x = r/R (r being the cylindrical coordinate and R the wire radius) corresponding to a time t, D is thermal diffusivity $D = k/\rho c$ (with k thermal conductivity, ρ mass density of the material and c is the material specific heat).

Under the hypothesis that the solidification of the material is producing in successive concentric shells, from the exterior to interior and the heat flow is stationary during this solidification process, equation (17) has the solution:

$$T(x) = T_2 + (T_1 - T_2)\log x / \log \varepsilon \tag{18}$$

where T_1 is the temperature of the melt and T_2 − the environmental temperature reached by the melt after solidification and cooling.

By entering the eq. (18) in (16) and solve the integral we obtain the mechanical stress expressions within wire after solidification:

$$\begin{aligned} \sigma_z(x) &= \int_0^x \sigma_z^*(x,\varepsilon)d\varepsilon \\ \sigma_r(x) &= \int_0^x \sigma_r^*(x,\varepsilon)d\varepsilon \\ \sigma_\varphi(x) &= \int_0^x \sigma_\varphi^*(x,\varepsilon)d\varepsilon \end{aligned} \tag{19}$$

Because these integrals have not exact solutions, we solve them numerically. The values of parameters and physical quantities involved are representative for CAW. Thus for T_1=1400 K, T_2 = 300 K, E = 100000 MPa, k = 0.0000004, 1/K υ = 0.33, which are typical for CAW, we obtain a maximum stress of hundreds of MPa, as we can observe in fig. 1.

Based on the stresses distribution, the next logical step is to establish the magnetic anisotropy distribution. Considering the above remarks we have performed two calculation programs which enable us to establish the anisotropy distribution for CAW corresponding to every kind of situation. Thus we can calculate the magnetoelastic anisotropy constant in every point of wire's radius, with any magnetostriction, positive, negative or nearly zero and any values of parameters and physical quantities involved.

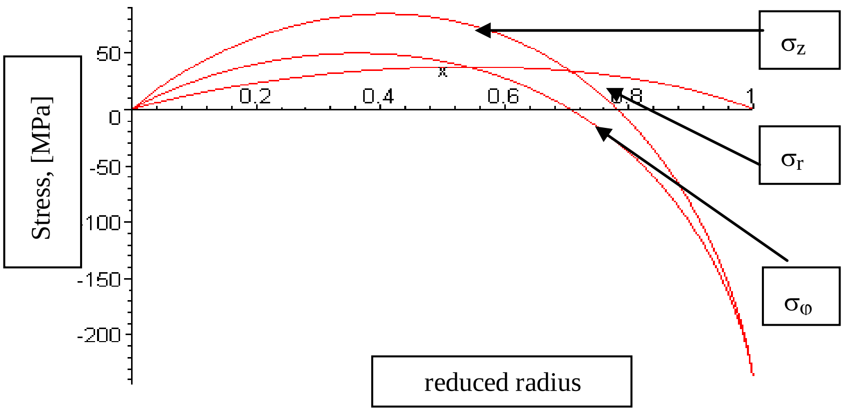

Fig. 1 − Stress distribution depending on wire radius under following conditions: T_1 = 1400 K, T_2 = 300 K,ν = 0.33, k = 4x10^{-6} K^{-1}, typical for CAW.

Magnetic anisotropy is the result of coupling between stresses and magnetostriction. The stresses have the dominant role in this coupling and in accordance with the sign and the value of magnetostriction we distinguish two cases:

A) Materials with positive magnetostriction (for example Fe-Si-B alloys), $\lambda = 10^{-6}$. In fig. 1 we observe two zones of interest from the dominant stress point of view. One region corresponding to x = 0-0.7, here the dominant stresses are the axial stresses and one corresponding to x = 0.7-1, where the dominant stresses are the radial stresses. In the first one the coupling between axial stresses and positive magnetostriction give rise to an axial magnetic anisotropy. In the second one the magnetic anisotropy is radial, following the same considerations. In conclusion, in this case we have two regions: a central core with axial anisotropy and an outer shell with radial anisotropy (fig.2).

The existence of magnetic domain structure given by this distribution was reported in (Severino *et al.*, 1992) experimental work. Except that, some new magnetization particularities were observed in both regions. Because of Matteuci and Wiedeman inverse effects and the magnetoelastic behavior of material, they have found a circular component of local magnetization.

B) Materials with negative magnetostriction and nearly zero magnetostriction (Co-Si-B and Fe-Co-Si-B alloys), $\lambda = 10^{-6}, \lambda = 10^{-7}$ (fig.3). We have been included in this category the materials with nearly zero magnetostriction because these materials have also negative magnetostriction but with one order of magnitude smaller than the others, the reason why the distribution of magnetic anisotropy is the same for both categories.

In this case we have a different situation. Because we have negative magnetostriction and for more than 0.7 of wire's radius we have positive stresses, the anisotropy distribution is not established the same way as in case

A). In a case like this we must choose the dominant stress that is the smallest one. The coupling with magnetostriction will establish the value of anisotropy but not its direction. The magnetization easy axis it will be a perpendicular direction to the smaller stress direction.

Considering the above remarks, in this case we have three interest regions. The first one corresponding to x = 0-0.54, with the smallest positive stress the radial stress, the second one corresponding to x = 0.54-0.7, with the smallest positive stress the azimuthal stress and the last one corresponding to x = 0.7-1, in which the stress and magnetostriction have the same sign. In this region the direction of anisotropy will be the azimuthal direction because of direct coupling between stresses and magnetostriction. In the other two regions the coupling between the smallest stresses and magnetostriction will give the absolute value of anisotropy. But its direction will be perpendicular to the smallest stresses directions. This direction will be axial or azimuthal (radial in the second zone). We will choose the axial direction corresponding to energy minimization. Otherwise, in these regions will be large magnetostatic and exchange energies. The magnetic domain forming it has to be the result of minimal energy and only an axial anisotropy will assure this.

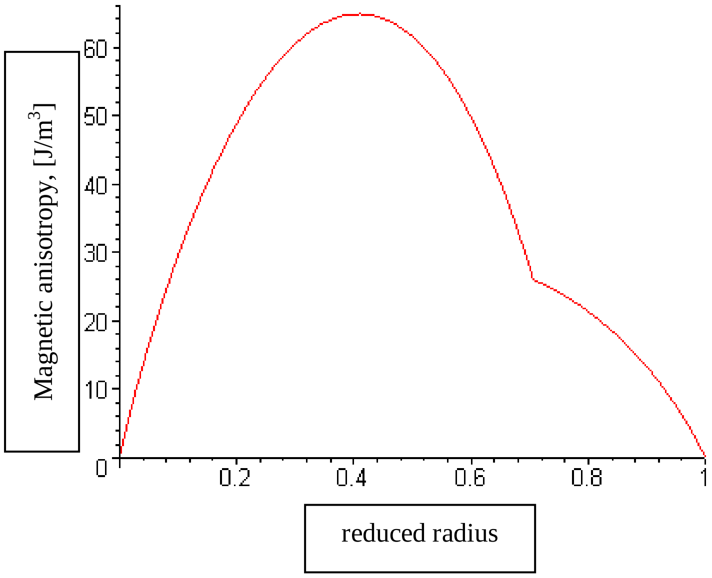

Fig. 2 – The magnetic anisotropy distribution in the general case $\lambda > 0$ under the conditions: $T_1 = 1400$ K, $T_2 = 300$ K, $E = 10^6$ MPa, $k = 4 \times 10^{-6} K^{-1}$, $\nu = 0.33$, $\lambda = 10^{-6}$.

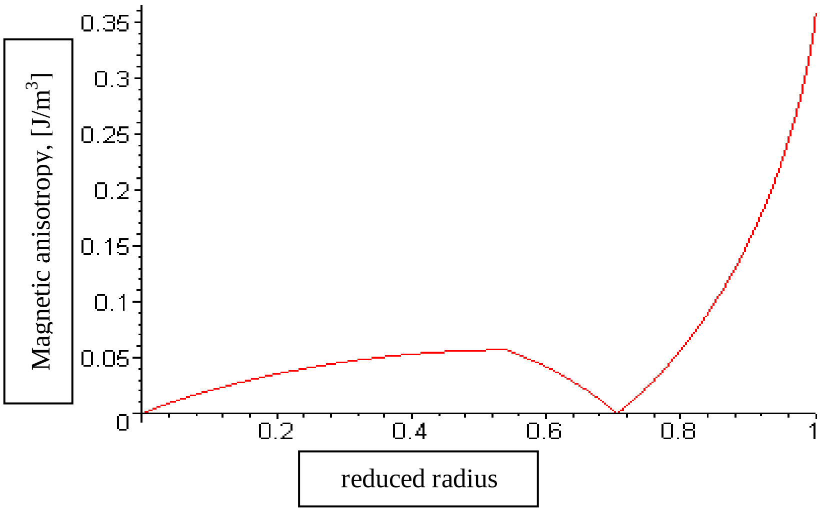

Fig. 3 – The magnetic anisotropy distribution in the general case $\lambda < 0$ under the conditions: $T_1 = 1400$ K, $T_2 = 300$ K, $E = 10^6$ MPa, $k = 4\text{x}10^{-6}$ K^{-1}, $\nu = 0.33$, $\lambda = 10^{-7}$.

By observing the magnetic domains in considered regions we concluded that the axial direction is the only one possible.

Our experimental data confirm these conclusions, (Răuţ *et al.*, 2006). We have been performed experimental measurements by GMI (giant magnetoimpedance effect) method in order to verify the theoretical results in the surface region of CAW. We have been tested some probes consisting of Co-Fe-Si-B CAW and we have found for anisotropy field H_K values near to 50 A/m. The anisotropy constant deduced from anisotropy field expression, written in uniform rotation of magnetization hypothesis

$$H_K = 2k / \mu_0 M_s$$

was estimated at 20.25 J/m^3. The calculated theoretical value for the same conditions is 22.6 J/m^3. There is a good agreement between theory and experiment, (Răuţ *et al.*, 2006).

4. Conclusions

Rapid solidification process gives rise in CAW of three kinds of stresses: axial, radial and azimuthal. These stresses coupled with magnetostriction are at the origin of the magnetoelastic anisotropy. In case corresponding to materials with positive magnetostriction we have shown the existence of two regions of interest: central core, where the magnetoelastic anisotropy is axial, and the outer shell, with radial anisotropy.

The case corresponding to materials with negative magnetostriction was studied too. We have shown the existence of three regions of interest. One is central, with axial anisotropy, the others with azimuthal anisotropy.

The experimental investigations confirm the theoretical results.

REFERENCES

Răuţ M.B., Ţibu M., Chiriac H., *Theoretical Model for the Calculation of the Anisotropy Distribution in the Surface Region of the Amorphous Wires*. Bul. Inst. Pol. Iaşi, **L(LV)**, 11−18, 2006.

Severino A.M., Gomez-Polo C., Marin P., Vasquez M., J. Magn. Magn. Mater., **103**, 117, 1992.

Squire P.T., Atkinson D., *Magnetostrictive and Magnetoelastic Properties of Rapidly Quenched Wire*. IEEE Trans. Mag., **25**, 24−27, 1995.

Velasquez J., Vasquez M., Hernando A., Savage H.T., Wun-Fogle M., J. Appl. Phys. **70**, 6525, 1991.