Journal of Alloys and Compounds 622 (2015) 635–639

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jalcom

Improving soft magnetic properties in FINEMET-like alloys. A study

J.A. Moya

4. GIM – IESIING, Universidad Católica de Salta, INTECIN (UBA-CONICET), Argentina

ARTICLE INFO

Article history:
Received 10 September 2014
Received in revised form 11 October 2014
Accepted 24 October 2014
Available online 4 November 2014

Keywords:
Nanocrystalline alloys
Soft magnetic materials
Coercive field
Random anisotropy model

ABSTRACT

An approach to improve the soft magnetic properties of nanocomposite FINEMET-like alloys is presented by the study of the theoretical behavior of the coercive field with the crystallized fraction, $H_c(v_{cr})$, curves where the total anisotropy is taken into account. These curves can exhibit a minimum value that should be used for a proper design of soft magnetic alloys, consisting in matching that value with the optimal crystallized fraction of the alloy. For this, the effect of Si, Ge, Co and Al on the magnetic properties on the material phases and on its microstructure was analyzed while a good relationship between theoretical results and data in the literature was found. Small amounts of Ge and/or Al can improve the magnetic properties of FINEMET while the addition of Co deteriorates the coercive field as is predicted by the theory. Simultaneous addition of two or more solute elements to the α-Fe crystals was also evaluated and discussed. Results indicate the importance of knowing the effect of the alloying element on the crystalline magnetostriction constant and on the crystallized fraction of the microstructure.

1. Introduction

Since its development, soft magnetic nanocrystalline materials have attracted great attention and much effort has been made by researchers in order to improve their soft properties, for example, with the addition of elements that increase the saturation magnetization and permeability, and reduce the coercive field and the magnetic losses. A small grain size, of the order of tens of nanometers, is the key to their good properties and, often, the addition of these elements was conducted for this purpose neglecting other important features. In a previous work [1], we have discussed the parameters to design nanocrystalline soft magnetic materials such as FINEMET, NANOPERM, HITPERM and NANOMAT types and the effect of some alloy elements on them. In order to do this, we have modeled the theoretical behavior of the coercive force, H_c, with the crystalline fraction, v_{cr}, of the nanocomposite materials taking into account some material parameters, like the grain diameter, D, the magnetocrystalline constant, K_1, the crystalline and amorphous magnetostriction constants, λ_{am} and λ_{cr}, respectively, the internal stresses, σ_i, and v_{cr}, which act in magnetocrystalline and uniaxial anisotropies according to models in literature. In the case of FINEMET™ alloys (typical composition $Fe_{73.5}Si_{13.5}B_9Cu_1Nb_3$), they present a clear minimum in the $H_c(v_{cr})$ curve that can be exploited to soften the material. In this work, we go deeper into previous research focusing our attention on FINEMET-like nanocrystalline systems, analyzing the origin of this minimum, studying the magnetic effect of some solutes on the crystalline phase (compiling several charts from literature) and the behavior of the magnetostriction constant of the amorphous matrix with the crystallized fraction, which improving the results obtained in [1]. The theoretical results are first compared with a whole series of a $Fe_{73.5}Cu_1Nb_3Si_{22.5-x}B_x$ alloys from literature validating the model, and the effect of Ge, Al, Co and Ge + Al on the α-Fe(Si) phases are discussed. The results give new tools for a proper soft magnetic nanocrystalline alloy design.

2. Material and methods

Our studies are based on an average anisotropy model that takes into account the random anisotropy model for the magnetocrystalline anisotropy [2] and the uniaxial anisotropies such as magnetoelastic and field-induced ones. A greater understanding of the subject can be found in [2,3] and a brief summary in [1]. Here, we write the equations used in the present work and the corresponding constants without going into details. It is known that the coercive field, H_c, depends on the total anisotropy, $\langle K \rangle$, and the magnetic polarization, J_s, as:

$$H_c = p_c \cdot \langle K \rangle / J_s \tag{1}$$

with p_c equal to ~0.2 for our materials. Before defining $\langle K \rangle$, we must study the anisotropies acting in our materials. The most significant are the magnetocrystalline, K_1, and magnetoelastic, Ku_{el}, ones, as field induced anisotropies are negligible or not present in the current studied material. In the random anisotropy model, K_1 is average as:

$$\langle K_1 \rangle = \beta \cdot v_{cr} \cdot K_1 \cdot (D/L_{ex})^{3/2} \tag{2}$$

where β is a constant estimated in ~0.5 and L_{ex} the correlation length of the ferromagnetic exchange to be defined later. With respect to the magnetoelastic anisotropy, we have that:

E-mail address: jmoya.fi.uba@gmail.com

http://dx.doi.org/10.1016/j.jallcom.2014.10.124
0925-8388/

$$Ku_{el} = 3/2 \mid \lambda_s \mid \cdot \sigma_i \quad (3)$$

where σ_i is the internal (mechanical) stresses and λ_s is the effective magnetostriction constant of the nanocomposite material that can be expressed as:

$$\lambda_s = v_{cr} \cdot \lambda_{cr} + (1 - v_{cr}) \cdot \lambda_{am} \quad (4)$$

being λ_{am} and λ_{cr} the magnetostriction constants of the amorphous and nanocrystalline phases, respectively, and v_{cr} the crystallized fraction.

The total anisotropy, $\langle K \rangle$, can be calculated as the average:

$$\langle K \rangle = \sqrt{\beta^2 \langle K_1 \rangle^2 + Ku_{el}^2} \quad (5)$$

The correlation length is then defined as $L_{ex} = \varphi\ (\beta \cdot A_{cr}/\langle K \rangle)^{1/2}$ with the constant $\varphi = 1.5$ and the exchange stiffness, A_{cr}, set in $0.6 \cdot 10^{-11}$ J/m, as it was reported in experimental results [2]. Eq. (5) becomes self-consistent and it can be solved by iteration. Introducing Eq. (5) in Eq. (1) we can obtain the behavior of H_c with v_{cr}, $H_c(v_{cr})$.

3. Calculation

3.1. Nanocrystalline phase considerations

FINEMET-like nanocomposite alloys are formed by α-Fe(Si, Si + M) solid solution nanocrystals, with M equal to one or more elements as Al, Ge, Co, among others, surrounded by a Fe-rich amorphous matrix. The total solute, Si + M, content of the nanograins range approximately from 15 to 22 at.% The effects of the mentioned solute elements on the magnetic properties on Fe crystals are shown in Fig. 1. The elements Si, Al and Ge reduce J_s of the Fe crystals in approximately the same way up to ~20% of solute content, while Co increases it and it has a maximum at ~40% of Co content (Fig. 1a). Also, all of them provoke a decrease in K_1 constant of the Fe crystals (Fig. 1b). This fact is important because, between certain limits, the soft magnetic properties of the nanocomposite material become more and more independent on grain size as $\langle K_1 \rangle$ is reduced (see Eq. (2)). Finally, Fig. 1c shows the way these solutes change the magnetostriction constant, λ_{cr}, of the Fe crystals, which affects the magnetoelastic anisotropy Ku_{el} (Eq. (6)). When $\langle K_1 \rangle$ is reduced (either by reducing D or K_1 of nanocrystals) Ku_{el} governs the total anisotropy and it can be reduced via the magnetostriction balance of both phases; as the amorphous matrix is a Fe-rich phase, it has a positive λ_{am} value and, in order to be balanced, we must have a negative λ_{cr}. In our examples of Fig. 1c, this condition is satisfied by a free-solute Fe crystal (not a FINEMET alloy) or by Fe crystal containing more than ~15% of Si or Ge. Alloying Fe with Al or Co will not satisfy this condition.

The effect of the internal stresses, σ_i, has also to be computed in the value of Ku_{el} since a total balance of the effective magnetostriction constant, λ_s, is not always possible, as we will see later on. Ribbons obtained in the as-quenched state have a high stress level reported in 30–15 MPa [12,13] that are reduced with subsequent heat treatment until the first stages of nanocrystallization (low v_{cr}), where there is an increase in σ_i due to the grain nucleation and a relatively low annealing temperature. At optimum annealing temperature with a relatively high v_{cr}, stresses can be released up to 2.5–0.2 MPa. A further increase in v_{cr} may result in a new increase in σ_i either due to a growth in the lattice mismatch in grain boundaries and/or to higher differences in densities between the crystalline and amorphous phases.

In addition, the v_{cr} plays a critical role in soft magnetic properties through the balance of magnetostriction constants of the nanocomposite material. The v_{cr} can be controlled in several ways, for example, it increases by introducing solutes to the α-Fe crystal, by replacing one solute element for another (like Si by Ge or Al) or by reducing B content.

3.2. Amorphous phase considerations

With respect to the saturation magnetization and the magnetostriction constant of the remnant amorphous matrix, for our calculus we can consider, on the one hand, that the saturation magnetization increases directly with its iron content (for

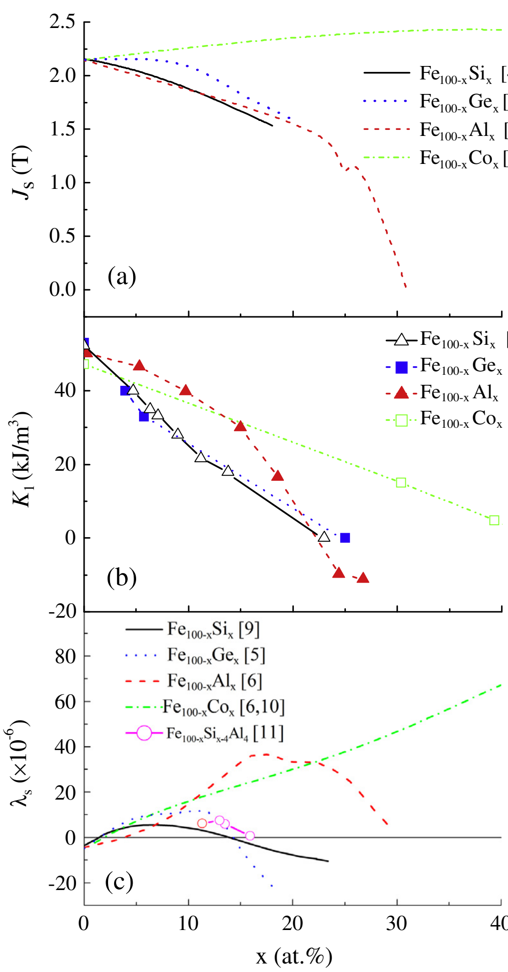

Fig. 1. Effect of Si, Ge, Al and Co solute elements on some magnetic properties of the α-Fe phase. Dependence on solute content of: (a) J_s, (b) K_1 and (c) magnetostriction constant. Data obtained from references indicated in graph legends. (See above-mentioned references for further information.)

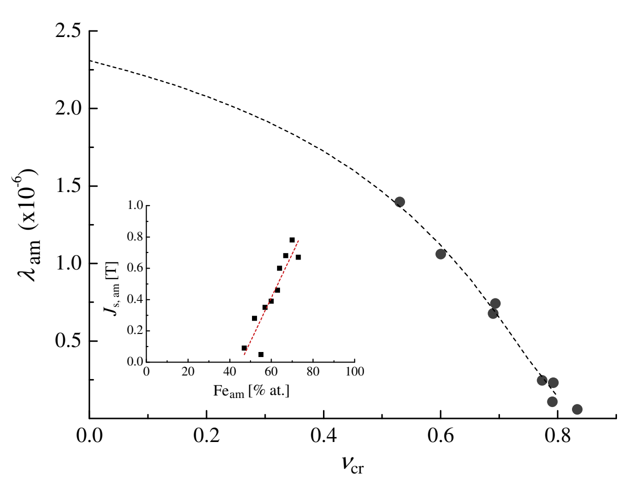

Fig. 2. Evolution of the magnetostriction constant, λ_{am}, of the amorphous remnant phase with the crystallized fraction, v_{cr}, obtained from Eq. (7). Inset, behavior of the saturation magnetization, J_s, of the amorphous phase of similar composition of the remnant matrix with its Fe content [14].