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FERROMAGNETIC NANOCOMPOSITES WITH TAILORED HYSTERESIS LOOP BY MAGNETIC AND TENSILE STRESS ANNEALING

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New, annealing time reducing technologies for heat treatments with and without magnetic field of amorphous precursor of soft magnetic nanocrystalline alloys will be presented for traditional Finemet alloy ($Fe_{73.5}Si_{15.5}B_7Nb_3Cu_1$). Small – scale thermal cycles with steadily increasing average temperatures can replace successfully the presently applied long term heat treatments with small heating velocity. In the case of continuous tensile stress annealing, the pulling velocity of ribbon can be increased up to 90 m/min which makes the productivity of this method competitive with the magnetic field annealing. The ultimate goal is to flatten the hysteresis loop on demand extending the linearity up to the induced anisotropy field value. The effective permeability can be decreased down to 20 000 by applying transversal magnetic field annealing and further decrease down to 80 value can be obtained by applying tensile stress annealing up to 500 MPa. A properly conducted magnetic field annealing improves the coercive field below 1 A/m. The heterogeneities introduced by tensile stress annealing can be controlled to keep the coercive field below 10 A/m.

Keywords: amorphous magnetic materials, magnetic properties, perpendicular magnetic anisotropy, soft magnetic materials

1 INTRODUCTION

The eddy current limits for metallic cores should be shifted above 1 MHz in order to keep the operating frequencies about 100 kHz to 500 kHz for inductors and transformers. The high operating frequency permits the mass and volume reducing which is a major demand nowadays in aerospace and automobile applications. Specially, in the context of aerospace applications, a magnetic material that can maintain a lower core loss over the applicable temperature range can provide a basis for nosing out the competition with ferrites (limited temperature range) and with crystalline permalloy of 17 μm thick (limited plastic yields strength).

Maximum permeability of at least 300 000 was available for the Permalloy-type Ni-Fe cores in our lab. Applying a proper annealing cycle for Finemet type nanocrystalline alloy, round (R) hysteresis loop with remanence ratio greater than 0.7 and with maximum permeability as high as one million were obtained. The temperature dependence of the permeability is also satisfactory for the applications (±20-30%). Toroidal cores for AC-type Residual Current Circuit Breakers (RCCBs) can be produced using such R type cores with weight reduction down to 40 – 70 % compared to the Permalloy-type cores for similar application.

Nevertheless, even in zero external field annealing, the induced anisotropy, K_u, by the local magnetic flux within the domain for annealing temperatures below the Curie point can not be excluded. The ideal value of remanence-to-saturation ratio (J_r / J_s) is 0.83 for randomly oriented cubic system. A smaller value indicates the presence of induced uniaxial anisotropies. This “self” induced anisotropy can be eliminated by annealing in rotating magnetic field with sufficient high amplitude to overcome the local demagnetizing field and saturate the sample [1].

Annealing in static magnetic field (both longitudinal and transversal) goes back to the first papers on Finemet type nanocrystalline alloy [2].

It has been recognized that for low loss and wide applicable temperature demand magnetic field [3] and stress [4] annealed traditional Finemet ($Fe_{73.5}Si_{15.5}B_7Nb_3Cu_1$) represent an advanced material permitting new solutions for high frequency magnetic applications. Compact toroids without gap can be prepared with high quality factor and high current inductors [5]. This work presents some new results concerning the magnetic field and stress annealing of this soft magnetic nanocrystalline alloy with most extensive applications at the time being.

2 MAGNETIC FILED ANNEALING

When a magnetic material is heated below its Curie temperature but the temperature is still high enough for short-range atomic mobility, a local ordering takes place which stabilize the magnetization into the direction of the applied field during annealing [2]. This transversal induced anisotropy can be as high as 30 J/m^3 in the case of Finemet alloys. Prior to the magnetic field annealing the amorphous precursor sample must be transformed into a nanocrystalline structure. Due to the self heating of the sample during the amorphous –crystalline transformation the temperature of the sample can run out the interval between the two crystallization steps (about 150 K [2]) of the Finemet alloy. Entering into the second crystallization stage is detrimental to the soft magnetic properties due to the precipitation of hard Fe_2B phase. The higher the heating rate and the larger the mass of the core, the larger the temperature increase is. For example, we have found for a core of 100 gram a temperature increase of 100 degree above the programmed one at a heating rate of 2 K/min. This is why the annealing in magnetic field is performed with a very low heating rate (as law as 0.2 K/min in the 480°C-540°C temperature region) and a continuous H_2 gas cooling should be applied to avoid the self-heating of the samples.

This increases enormously the cost of the heat treatment which last more than 10 hours performed under flowing hydrogen gas. This is why; in this paper we present a meth-

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od to reduce the heat treatment time necessary for magnetically induce anisotropy. The method consists of cyclic heating and cooling within a small temperature interval of several 10 degrees, while the medium temperature is progressively increased in the interval of 480 °C - 540 °C, after each cycle. The self cooling at this relatively high temperature is enough rapid in order to achieve a rate of 10-15 minutes per cycle. A sum of 5-6 cycles is enough to obtain a nanocrystalline structure which subsequently can be cooled down to the temperature of magnetic field annealing performed usually at around 360 °C-480 °C for 2 hours in transversal field of 160 kA/m. In this way, the annealing time can be reduced to 3-4 hours even for a quantum of several kg of samples.

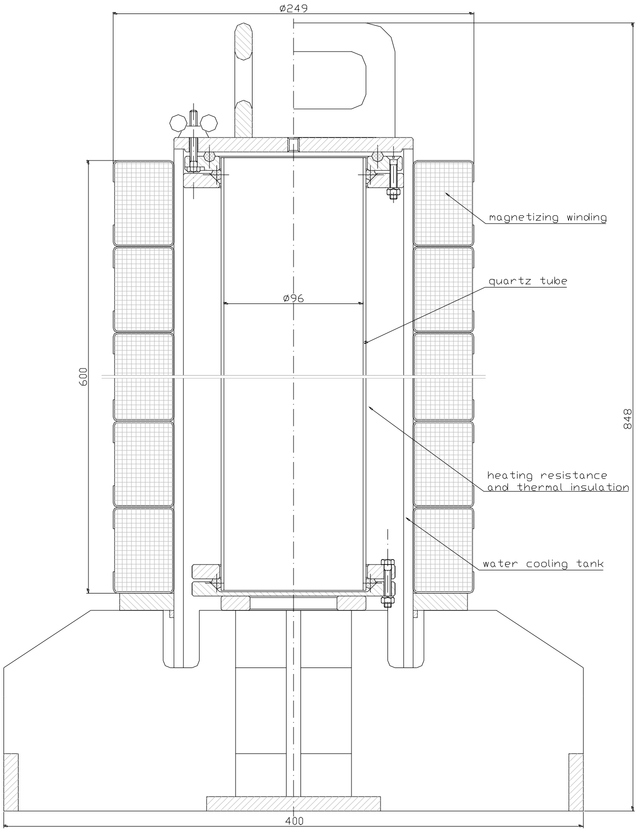

Fig. 1. Schematic drawing for the magnetic annealing furnace

A homemade annealing devise is shown schematically in Fig.1 and 2. The volume for the annealing is a cylinder of 96 mm diameter and 400 mm high.

Applying first a longitudinal field created by a DC current of 100 A through a rod placed axial to the toroidal sample (which form a Z type loop) and subsequently a transversal field of 160 kA/m created by the solenoid (which flatten the loop) one can obtain a linear (F type) loop with a coercivity smaller than that of the round (R) type loop (see Fig.3).

For Finemet type nanocrystalline alloy applying various annealing's between 380 °C – 480 °C for different times in transversal magnetic field of 160 kA/m the smallest obtainable permeability (the largest flattening) is about 20 000 corresponding to a transversal induced anisotropy of 30 J/m^3.

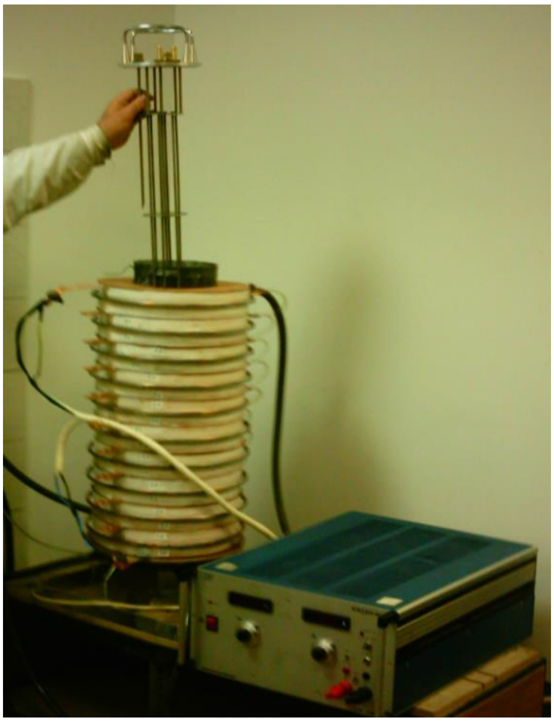

Fig. 2. The photo of the magnetic field annealing device

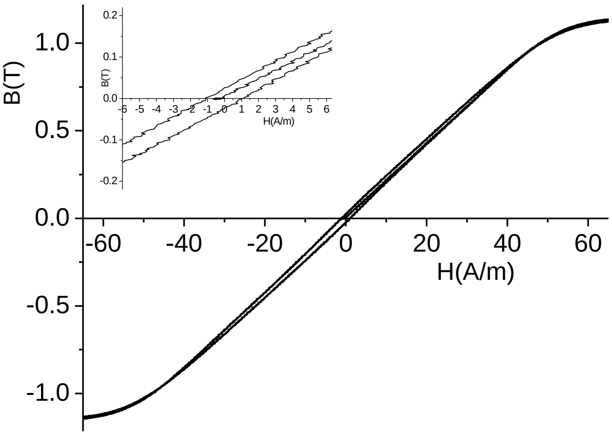

Fig. 3. Linear magnetization loop obtained after magnetic field annealing: μ_{eff} = 20 000, Hc = 1A/m

3 STRESS ANNEALING

The induced anisotropy can be increased further on up to 5000 J/m^3 applying a tensile stress during the amorphous - nanocrystalline transformation annealing. The correlation between the applied stress and induced anisotropy is roughly $K_{ind} \sim 10*\sigma$, where the tensile stress is given in MPa and K_{ind} is in J/m^3. This correlation is similar for static or continuous tensile stress annealing and is explained within the framework of the back-stress model [6]:

$$K_{ind} = -\frac{3}{2}\lambda^{s}_{cr}x_{cr}\sigma \qquad (1)$$

Where λ^{s}_{cr} is the saturation magnetostriction for $Fe_{80}Si_{20}$ crystalline precipitate, x_{cr} is the volume fraction of the crystalline phase and σ is the applied tensile stress during annealing.

For practical applications the continuous (or flash) annealing can be considered only. The precursor amorphous ribbon is placed in a continuous stress annealing equipment which is similar to that applied by others [5]. The equipment presented schematically in Fig. 4 includes an open tubular furnace cut along the generator of the cylinder in order to place the running ribbon into the middle of the furnace. No protective gas is applied during annealing. The ribbon is pulled reel-to reel at constant velocities under a constant tensile stress. The sample coils are rolled up subsequently from the bobbin containing the nanocrystalline ribbon. The pulling force between 10-500 MPa is measured with a dynamometer F and is kept constant through a feedback to the motor with controlled torque. Both servo-motors have incremental encoder which make possible to read the position values of the motors. From these positions the positive or negative strain of the ribbon can be determined. The negative strain appears at small stresses where the contraction due to the amorphous –crystalline transformation is larger than the elongation due to the creep.

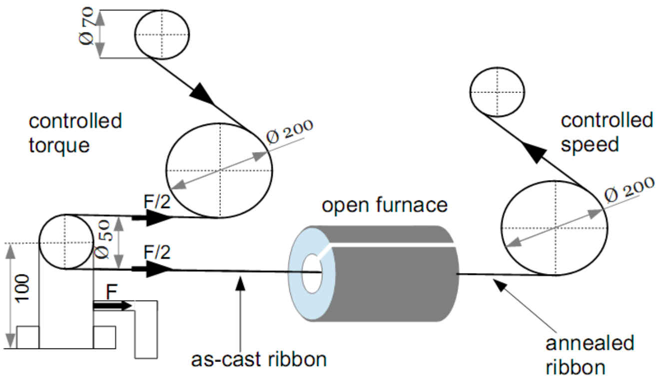

Fig. 4. Schematic drawing of the continuous stress annealing equipment

For experiments amorphous ribbon with Finemet composition ($Fe_{73.5}Si_{15.5}B_7Nb_3Cu_1$) of 6 mm wide were produced by planar flow casting in our laboratory. The pulling velocity depends on the length of the tubular furnace and is set in order to obtain 5-10 seconds for the running time through the furnace. The annealing is in air and the temperature is set between 873 and 973 K. The pulling velocity of ribbon under tensile stress annealing can be increased up to 90 m/min which makes the productivity of this method competitive with the magnetic field annealing.

The following experimental results have been obtained:

- the flatness (effective permeability) depends on the applied tensile stress only (see eq.(1)), whereas the elongation (creep) depends both on the applied stress and pulling velocity. The elongation is increasing with the pulling velocity [7]. This finding is questioning the role of creep in the induced transversal anisotropy.
- The oxidation in air produced no discoloration of the ribbon and did not deteriorate the soft magnetic properties. The presence of oxide layer is evident from the fact that the annealed ribbon dissolved more difficult in acids (like aqua regia) than the as cast ribbon. What is more, the oxide layer plays the role of electric insulator for the wound up ribbon.
- With this technique we could tailor the hysteresis loop on demand with effective permeability ranging between 100 – 10 000. The coercivity remains low below 12 A/m, even for the lowest obtainable permeability (the largest tensile stress applied). In Fig.5 the quasi DC (*f* = 0.005 Hz) linear magnetization curve is shown up to the saturation with a magnetization field resolution better than 10^{-3} of the saturation field.

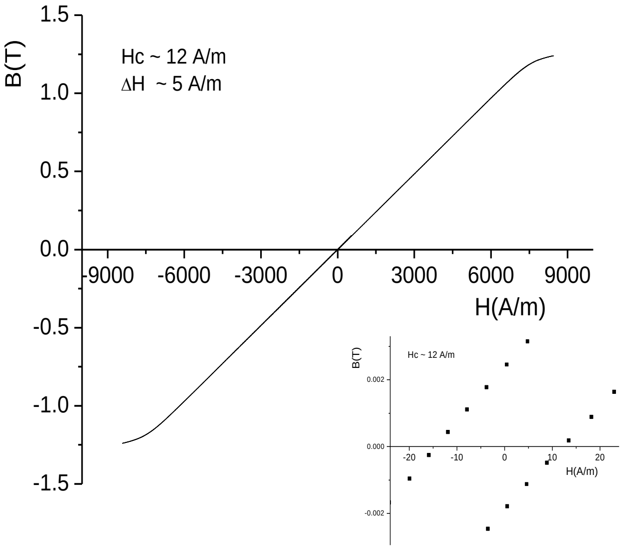

Fig. 5. The linear hysteresis loop (μ_{eff} ~ 128) obtained by continuous stress annealing. The insert shows the resolution of 5 A/m of the DC quasi –static measurement (*f* = 0.005 Hz).

The fact that the coercivity remains small even for such a large stress shows that the amorphous-nanocrystalline transformation is homogeneous both longitudinal and transversal to the ribbon and no deformation happens as it can be seen on the Fig. 6, where lighting of the ribbon passing through the middle part of the furnace can be observed due to the self-heating produced by crystallization. The average temperature of the furnace was 650°C and the pulling velocity of the ribbon was 20 m/min at an applied stress of 200 MPa.

The lighting appears fortunately due to the self heating of the exothermic crystallization which turns the claret into the reddish radiation above 650°C.

The small coercivity can be understood taking into account the smoothing effect of the induced anisotropy. The induced anisotropy enhances the average anisotropy and reduces its fluctuation [6]. The coercive field is determined by the anisotropy fluctuation and not by the averaged anisotropy. The induced anisotropy smoothes out the anisotropy fluctuation on a larger scale then the renormalized exchange length L_{ex} ~ $(A/\langle K\rangle)^{1/2}$. For a long range fluctuation length

Fig. 6. Lighting due to the exothermic nano-crystallization

$\Lambda > L_{ex}$, the coercive field will be determined by $H_c \sim K_u^{1/2}/\lambda$ [6].

The eddy current limit as obtained from the permeability spectra (maximum of the imaginary part of the permeability) is around 10 MHz for the sample with linear hysteresis loop presented in Fig. 5.

4 CONCLUSIONS

The low coercivity and the tailorable law permeability of stress annealed Finemet make it more competitive compared to the MnZn ferrites for high frequency applications up to 10 MHz. Magnetic field and stress annealing complement each other producing cores with effective permeability's between 100 000 - 20 000 and 10 000 - 100, respectively

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