Improved soft magnetic properties in nanocrystalline FeCuNbSiB Nanophy® cores by intense magnetic field annealing

Rajasekhar Madugundoa,b,c, Olivier Geoffroya,b,c, Thierry Waeckerlebd, Bianca Frincua,b, Stéphanie Kodjikian a,b, Sophie Rivoirard a,b,*

a Univ. Grenoble Alpes, Inst NEEL, F-38000 Grenoble, France
b CNRS, Inst NEEL, F-38000 Grenoble, France
c Grenoble Electrical Engineering Laboratory (G2Elab), Bâtiment GreEn-ER, 21 avenue des martyrs, 38031 Grenoble, France
d Aperam Research Center, 58160 Imphy, France

ABSTRACT

The effect of high external magnetic field (up to 7 T) on soft magnetic properties in nanocrystalline Fe74.1Si15.7Nb3.1B6.1Cu1 Nanophy® cores has been investigated. The as-quenched amorphous ribbons were nanocrystallized by annealing between 540 and 620 °C in transverse magnetic field. By varying annealing field from 0 to 7 T, induced anisotropy ranging from as low as 4 J/m³ to as high as 41 J/m³ is obtained. It is responsible for an increase in the cut-off frequency up to 300 kHz when the material is submitted to dynamic magnetic excitations. A minimum coercivity of 0.74 A/m is observed in the core annealed in 1 T associated to low losses. The relative permeability decreases on increasing the annealing field intensity with a minimum value of 13,654 at 7 T. Such permeability level opens the way to new applications of the Nanophy® alloys.

1. Introduction

From very promising materials in the 80’s [1], nanocrystalline FeCuNbSiB based soft-magnetic materials have now emerged to a wide range of highly efficient commercial products in many applications needing electrical devices operating at low or medium frequencies [2–6]. These nanocrystalline materials, obtained by partial nanocrystallization of amorphous matrix – as cast from the liquid in thin ribbons – exhibit excellent soft magnetic properties such as low coercivity (Hc), high saturation induction (~1.25 T) very low and adjustable magnetostriiction (< 2×10⁻⁵), and significantly better thermal stability of soft magnetic properties as compared to their amorphous counterparts [1,4,7,8]. These attractive properties, attributed in the major part to the nanosize of the randomly oriented bcc Fe(Si) crystals (10–15 nm) in amorphous matrix [7,9] are further emphasized – as in any ultra-soft magnetic materials – by uniaxial anisotropy controlled both by alloy composition and magnetic field annealing [10–15]. Such induced anisotropy enables to control the shape of the hysteresis loop and thus meets the demands of various applications. For example, the high remanence ratio obtained by longitudinal magnetic field annealing makes the soft magnetic material suitable for saturable reactors and high frequency transformers, whereas a flat B–H curve obtained by transverse magnetic field annealing can be used in choke coils applications [10]. In the nanocrystalline FeCuNbSiB alloy family, Herzer reports that the magnitude of the anisotropy is proportional to the metalloid content, with a maximum of 100 J/m³ at the composition Fe73.5Cu1Nb3Si13.5B9 [11]. The mechanism of induced anisotropy is assumed to be Fe–Si pair ordering in the nanocrystalline phase [16–18]. The present study investigates the effect on the soft magnetic properties of an intense magnetic field (up to 7 T) applied during the crystallization of Nanophy® (Fe74.1Si15.7Nb3.1B6.1Cu1) cores.

2. Experimental details

Amorphous ribbons of composition Fe74.1Si15.7Nb3.1B6.1Cu1, were prepared by rapid solidification (10 mm in width and ~20 µm in thickness). The ribbons were wound into toroidal cores and annealed under pure H2 at different temperatures in the range of 540–620 °C for 1 h and with magnetic field intensities between 0 and 7 T. The DC magnetic field was applied along the core axis. After annealing, quasi-static magnetic measurements were carried out on the cores using a hysteresis loop recorder at f=2.3 Hz. Beside, dynamic magnetic properties were measured in the frequency range up to 2 MHz at the magnetic induction of 0.1 T. The cut-off frequency was determined as the frequency, where the imaginary part of permeability is maximum. Effect of magnetic field annealing on microstructure was investigated by using a transmission electron microscope (TEM). The images were analyzed to extract the grain size distribution by using Visilog® soft-
3. Results and discussion

In a first step, the optimization of annealing conditions was realized. The toroidal cores were annealed at various temperatures in the range 540–620 °C for 60 min in 7 T under H2. The dependence of induced anisotropy (Ku), relative permeability (μr), and coercivity (Hc) on the annealing temperature are shown in Fig. 1. The magnitude of Ku increases from 31 to 44 J/m³ with increasing annealing temperature from 540 to 620 °C. Consequently μr is decreased from 18,000 down to 13,000. Diffusional pair ordering in the Fe(Si) nanocrystals is invoked to be the cause of the induced anisotropy. Just above the crystallization temperature (found at 540 °C in this alloy composition) [19], the coercivity decreases down to a minimum value of 0.92 A/m obtained for a 560 °C annealing. This decrease is attributed to the development of the induced anisotropy. When the annealing temperature is further increased (above 560 °C) a coercivity increase is observed. At high temperatures, thermal stability of the microstructure is affected and magnetic hardening occurs through crystallization of secondary phases in the amorphous matrix. A good compromise of Ku, μr and Hc values being obtained in the core annealed at 580 °C, this temperature was chosen to study the influence of magnetic field intensity on the soft magnetic properties.

For that purpose, the cores were annealed in 0, 1, 3, 5 and 7 T at 580 °C for 60 min under H2. The results are shown in Fig. 2. The variations of Ku, μr, Hc values with annealing field deduced from the hysteresis loops of Fig. 2 are represented in Fig. 3. With increasing annealing field intensity, Ku is increased from 4 to 41 J/m³ and μr is decreased from 135,122 to 13,654. In both cases, this corresponds to a variation of one order of magnitude. One can observe in Fig. 3 that Ku does not reach a saturation value and further increase could be expected using higher field intensity, with however limited impact on the relative permeability. The μr value of 13,654 obtained at 7 T annealing in our work is quite different from the prior art (> 30,000) [19] and though such μr levels are reached in industry with other alloy compositions, they are associated with large magnetostric- tion and thus to detrimental effects, such as noises, strain sensitivity. This new range of μr opens the way to new products for common mode choke or inductance applications for the Nanophy wire.

The dependence of losses per cycle on frequency for the cores is shown in Fig. 4(b). At low frequency the losses are higher in the field at 580 °C annealing temperature (found at 540 °C in this alloy composition) [19], the local magnetization is not so high. It is very weak at the beginning of crystallization from the amorphous paramagnetic phase and increases when crystallization of the ferromagnetic Fe(Si) nanocrystals proceeds. On the contrary, it is shown in Fig. 1 that high external field is more effective in increasing Ku when the annealing temperature is high (at 620 °C, i.e. very close to the Curie point of the Fe(Si) nanocrystals is expected to be responsible for the induced anisotropy. It is generally admitted that directional atomic order is brought about by local spontaneous magnetization rather than external applied field, although the applied field governs the direction of the local magnetization. Our results show that the local spontaneous magnetization does not play the major role in inducing anisotropy and that the contribution of the external field is prevailing. In fact, in the temperature range of the study (above the Curie point of the amorphous phase (360 °C) but below that of the Fe(Si) nanocrystalline phase (650 °C)) [19], the local magnetization is not so high. It is very weak at the beginning of crystallization from the amorphous paramagnetic phase and increases when crystallization of the ferromagnetic Fe(Si) nanocrystals proceeds. On the contrary, it is shown in Fig. 1 that high external field is more effective in increasing Ku when the annealing temperature is high (at 620 °C, i.e. very close to the Curie point of the nanocrystalline phase and thanks to high diffusion rate, the spontaneous magnetization drops but Ku is increased). These results may indicate a role of the external field in the mechanism of directional ordering much more important than predicted by the theory [20]. Further investigations are in progress to determine other possible structural causes of anisotropy, such as contribution of crystallographic texture, as reported by Fujii et al. [21] in the case of Fe73.5Si13.5B9Nb3Cu1 alloys or effect of the external field on the crystallization process.
cores compared to the cores annealed in zero field. The losses increase with frequency and they merge for the frequencies > 20 kHz, where the performance of all samples become similar.

Fig. 5(a) and (b) shows TEM bright field images of the samples treated in 0 and 7 T fields, respectively. The grain size distributions show a Gaussian type distribution with a maximum at around 13–15 nm for the 0 T sample and 7–8 nm for the 7 T sample. Reduction in the average grain size observed in the sample annealed in magnetic field is in agreement with the results reported in nanocrystalline ribbons by Tsurekawa et al. [22] and already observed in steels [23]. The grain size reduction in the sample annealed in 7 T is consistent with the improvement of functional properties in this sample.

4. Conclusions

High magnetic field applied during the nanocrystallization has significantly influenced the magnetic properties in Fe74.1Si15.7Nb3.1B6.1Cu1 Nanophy® cores. Induced anisotropy and permeability ranging between 4 and 41 J/m³ and 135,122–13,654, respectively were realized by annealing at different annealing fields from 0 to 7 T. A minimum $H_c$ of 0.74 A/m was observed in the core annealed in 1 T. The new level of permeability (13,654) obtained in this particular composition is quite different from the prior art and opens the way to new products for common mode choke or inductance applications.
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References