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LOW-FIELD THERMOMAGNETIC CURVES OF *MELT-SPUN* Nd–Fe–B RIBBONS

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Abstract. Low-field thermomagnetic behaviour of $Nd_{15}Fe_{77}B_8$ as-cast ingots and melt-spun ribbons quenched at different cooling rates as well as for some Nd–Fe–B magnets, produced by conventional powder metallurgy, and by hot pressed melt-spun ribbons have been studied in the temperature range: room temperature – Curie temperature.

1. Introduction

Hopkinson-type thermomagnetic behaviour of Nd–Fe–B magnetic alloys has been reported since the beginning of their investigation [1, 2] and it is still an object of current interest [3–6]. The maxima of the low-field thermomagnetic curves, plotted for zero-field cooled samples in the vicinity of their Curie temperatures (T_c) were attributed to the decrease of magnetic anisotropy down to zero when temperature increases up to T_c [2].

O. Popov and M. Mikhov [3] showed that the maxima of low-field thermomagnetic curves for random system of single-domain uniaxial particles (or clusters) can be easily explained and predicted in terms of Stoner–Wohlfarth model [7] after taking into consideration the specific temperature dependences of magnetic anisotropy constant and spontaneous magnetization. Thus the low-field thermomagnetic behaviour of the random system of single-domain $Nd_2Fe_{14}B$ particles was predicted on the basis of existing in the literature data for temperature dependences of spontaneous magnetization and first anisotropy constant.

Later this approach was used by J. B. Oliveira et al. [5] for explanation of thermomagnetic behaviour of melt-spun $Nd_{16}Fe_{76}B_8$ at different magnetic fields, and fairly good similarities between measured thermomagnetic curves and curves calculated by using first and second anisotropy constants were demonstrated.

In spite of this interest to the thermomagnetic behaviour of Nd–Fe–B magnetic alloys, to the best of our knowledge, there are no published experimental investigations on the Hopkinson-type effect for samples produced by different technologies or at different technological conditions and thus having different microstructures.

The main purpose of the present paper is: investigation of the Hopkinson-type thermomagnetic effect of $Nd_{15}Fe_{77}B_8$ melt-spun ribbons, produced at different cooling rates, and for some available Nd–Fe–B permanent magnets, produced by hot-pressed melt-spun ribbons and by conventional powder metallurgy. Some complimentary investigation of temperature dependence of hyperfine field on the iron nucleous in the range — room temperature and the Curie temperature for samples of the melt-spun ribbons, having well pronounced Hopkinson-type behaviour, is also performed.

2. Experimental

$Nd_{15}Fe_{77}B_8$ ribbons were prepared by melt spinning in Ar atmosphere at several surface speeds v_s of the wheel: 17, 19, 21, 23, 27 and 31 m/s.

X-ray analysis shows that their structure changes from amorphous for v_s = 31 m/s to crystalline, identical with those of $Nd_2Fe_{14}B$ for v_s = 17 m/s [8].

The ^{57}Fe Mössbauer spectra were recorded at room temperature as well as at several elevated temperatures up to the Curie temperatures in Ar on a conventional transmission spectrometer. The speed scale of the spectra corresponds to the center of standard α-Fe. Some of the spectra were numerically filtered by using convolution and stepped deconvolution [9]. The Mössbauer spectra at room temperature match the diagnostic spectrum of $Nd_2Fe_{14}B$ [10,11].

Samples of some available permanent magnets, produced by melt-spun and hot-pressed ribbons (General Motors), as well as produced by sintering procedures (Sumitomo Special Metals Co and Vacuumschmeltze) were also studied.

Thermomagnetic measurements were performed by VSM in fields between 1 kOe and 7 kOe over the temperature range from 300 K to 700 K in Ar. The samples were thermally demagnetized before every thermomagnetic curve plotting.

3. Experimental Results

The results from magnetic measurements can be summarized as follows:

(i) Hopkinson-type maxima have not been observed in the low-field thermomagnetic curves for as-cast master alloys and for magnets, produced by powder metallurgy procedures.

(ii) Hopkinson-type maxima have been observed for all of the melt-spun samples: as-spun ribbons and magnet, hot-pressed from ribbons. Some typical thermomagnetic curves are shown in Fig. 1*a* and Fig. 2.

(iii) There is a well-expressed correlation between the shape of the low-field thermomagnetic curves for the samples of as-spun ribbons and the corresponding cooling rate. It can be seen from Fig. 1 that:

(a) The room temperature magnetization at a constant field decreases when decreasing the cooling rate;

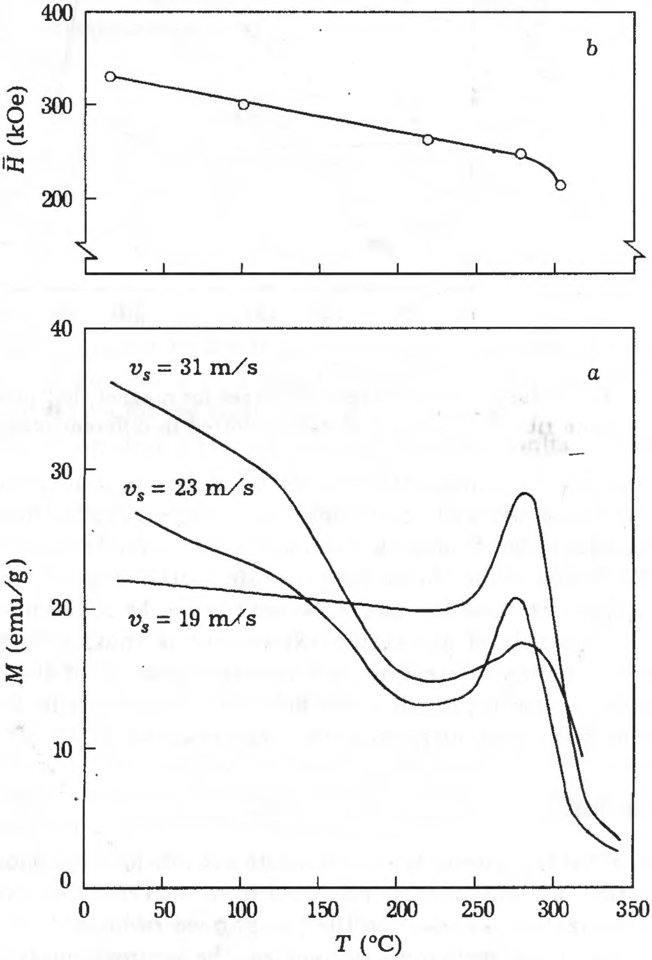

Fig. 1.

(*a*) Initial thermomagnetic curves for $Nd_{15}Fe_{77}B_8$ ribbons quenched at different surface speeds of the wheel, v_s, plotted in a field of 4 kOe;
(*b*) Temperature dependence of hyperfine field $\bar{H}$ for ribbons, quenched at $v_s = 23$ m/s

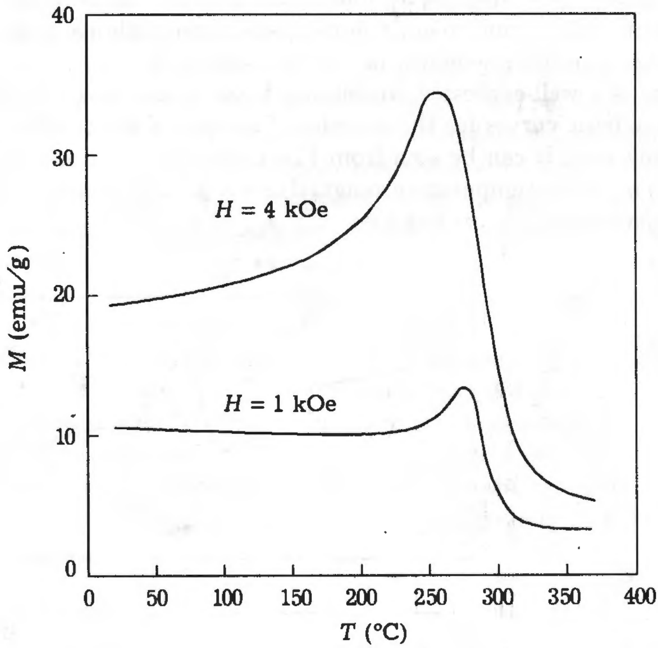

Fig. 2. Initial thermomagnetic curves for magnet, hot pressed from melt-spun ribbons (General Motors), plotted in different magnetic fields

(b) The low-field magnetization for all of the as-spun ribbons changes non-monotonously with the temperature, passing firstly through the minimum at about 200 °C and then through the maximum below T_c;

(c) With decreasing the cooling rate the initial slope of the thermomagnetic curves decreases the rate, however the height of the maximum increases;

(iv) The main result of Mössbauer experiment is that, in contrast to the low-field thermomagnetic curves, the average value $\bar{H}$ of the hyperfine field on Fe-nucleous has no peculiarities below T_c. As shown in Fig. 1*b* $\bar{H}$ decreases monotonously with increasing temperature up to T_c.

4. Discussion

The origin of the Hopkinson-type peculiarities in low-field thermomagnetic curves of melt-spun ribbons, and magnets produced from such ribbons, have to be connected to the magnetization processes in the fine grained ribbons.

The grains of the melt-spun ribbons can be approximately considered as disordered uniaxial single-domain particles [12] and their thermomagnetic behaviour can be described in terms of Stoner–Wohlfarth model. As shown in [3] the low-field thermomagnetic behaviour of such a system can be of Hopkinson-type, due to the specific temperature dependences of spontaneous magnetization and magnetic anisotropy constant.

The easy magnetization directions of the grains are randomly distributed and the system could be considered similar to a system with random magnetic anisotropy. In the thermally demagnetized sample the vectors of the grains magnetic moments are uniformly directed along their easy axes. The total magnetization measured in a low-magnetic field is very small and it is produced predominantly by reversible moments rotations. With increasing temperature, the magnetic anisotropy decreases and the irreversible rotations of the grains magnetic moments start to take place, causing an increasing of the total magnetization in a given field. With further increasing of the temperature, in the vicinity of T_c, the particles magnetic moments start to decrease more rapidly following the steep slope of the Brillouin function, and the magnetization measured starts to decrease after passing through the Hopkinson-type maximum.

As shown in [13] the thermal activation gives a considerable contribution to the low-field thermomagnetic curves of small particles systems. When the particles become smaller the Hopkinson-type maxima smear out. In the real samples the grains have no uniform sizes. Particularly in the melt-spun ribbons the grains on both opposite surfaces are formed at different cooling conditions and their average sizes could be different. The thermomagnetic curves for a disordered system of single-domain uniaxial particles, having two different average sizes (large enough, but not so large to be multidomain and small enough, but not too small to be superparamagnetic) are characterized by Hopkinson-type maximum preceded by a well expressed minimum. This could be a possible reason for the observed minima in the thermomagnetic curves for the melt-spun ribbons studied.

Having in mind the above considerations the experimental results reported can be explained as follows.

Melt-spun $Nd_{15}Fe_{77}B_8$ ribbons consist of small single-domain grains and their low-field thermomagnetic behaviour with clear Hopkinson-type effect is typical for such systems. The particles sizes are grouped around two distinct average sizes and, because of the thermally activated processes of moments rotations the Hopkinson-type maxima are preceded by well expressed minima. With increasing cooling rate the grains become smaller and thermally activated magnetization processes (including superparamagnetism) start to play much more important role apart from their single-domain Stoner–Wohlfarth-type behaviour. This leads to a weakening of the Hopkinson-type maxima near T_c, increasing the slope of the thermomagnetic curves before the minima, and increasing the values of the room temperature magnetization measured in constant low-magnetic field.

In the as-cast ingots and in the magnets, produced by conventional powder metallurgy the grains are too large to be considered as single domain particles [14], and the Hopkinson-type behaviour, produced by moments rotations cannot be observed at the experimental conditions used in the present work.

5. Conclusion

Hopkinson-type peculiarities were observed for all the melt-spun samples: as-spun ribbons and magnet, hot pressed from ribbons. The magnitude of the Hopkinson-type maximum decreases with increasing the cooling rate. This peculiarities were not observed for as-cast ingots and for magnets, produced by powder metallurgy procedures. The existence of the Hopkinson-type effect in the melt-spun samples was explained by their fine single-domain-like grains structure in terms of Stoner–Wohlfarth model.

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