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Effect of magnetic fields on melt-spun Nd$_2$Fe$_{14}$B-based ribbons

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The effect of a magnetic field on microstructure and magnetic properties of Nd$_2$Fe$_{14}$B-based melt-spun ribbons is investigated. The magnetic field was applied in perpendicular or parallel direction to the ribbon plane during quench with a field strength up to several kilo Oersteds. The XRD patterns and TEM graphs show a strong grain size reduction upon the magnetic field application. The magnetic field also enhances the (00l) texture of ribbons when the field is perpendicular to the ribbon plane. The refined microstructure with significantly reduced grain size leads to enhanced magnetic exchange interactions between the hard and soft phases in the Nd$_2$Fe$_{14}$B/Fe nanocomposite ribbons. This magnetic field-assisted melt-spinning technique is promising for producing nanocomposite magnets with enhanced energy density. © 2012 American Institute of Physics. [doi:10.1063/1.3679161]

I. INTRODUCTION

High-performance permanent magnets are the goal of numerous research efforts. The prediction of high energy product of exchange-coupled hard/soft phase nanocomposite magnets until now is not satisfied and the search of this kind of magnets is continuing intensively. Magnetic field annealing and processing have been widely used to improve magnetic performance of soft and hard magnets as well as properties of non-magnetic systems.¹–⁹ The grain size refinement, texture enhancement and magnetic property improvement were observed in the bulk NdFeB magnets prepared by injection casting in a magnetic field.¹⁰ The grain size refinement under a magnetic field during the solidification was observed in the Sn–Pb system.¹¹ It was also found that the field changes the phase equilibrium of the system, e.g., shifts the eutectic point in the Mn–Sb system to the Mn-rich concentration zone and even shifts the temperature-time-transformation (TTT) diagram of the isothermal martensitic transformation in the Fe$_{24.9}$Ni$_{13.9}$Mn alloy.¹² The field strength used in the experiments was in the Tesla range¹–⁵,⁷–¹³ except the small value of 90 Oe used in the work for the amorphous ribbons.⁶

In this work the effect of a magnetic field acting directly on the rapid quench solidification of the ferromagnetic system NdFeB/Fe was investigated. We present the experimental facts of the field-assisted grain size refinement as well as the texture and magnetic property enhancement. Some suggestions for explaining the obtained results are also presented.

II. EXPERIMENTS

A commercial melt-spinner has been modified with the home-made cooper wheel attached with commercially available permanent magnets, which allows to perform different configurations of fields either perpendicular (H$_\perp$) or parallel (H$_\parallel$) to the wheel surface. The magnetic field strength given in the text below is the maximal value in the surface.

The commercial Nd$_2$Fe$_{14}$B$_x$B$_{8−x}$ ingots have been used as the starting material. Pure iron in a fixed amount was added to the Nd$_3$Fe$_{77}$B$_8$ alloy for making composite samples. The wheel speed and the field strength were changed in the experiments while other parameters were kept constant. In the chamber 0.8 atm of argon gas was used during melt spinning.

The morphology and crystalline structure of the ribbons were characterized by transmission electron microscopy (TEM) and X-ray diffraction (XRD) using Cu K$_\alpha$ radiation. The grain size was determined with error bars by using the JADE software with Rietveld refinement option for the full width of half maximum (FWHM) analysis of the peaks taken in the 2θ range from 22° to 88°. Magnetic properties were measured in a superconducting quantum interference device (SQUID) magnetometer with a maximum applied field of 60 kOe.

III. RESULTS AND DISCUSSION

A. Grain size refinement effect

Grain size of melt-spun ribbons is usually controlled by the crystal growth process governed by high temperature gradient V_T perpendicular to the ribbon plane from the contact surface (with the wheel surface) to the free surface. During the melt-spinning process, the melt of starting alloys is ejected and solidified on the wheel surface at a temperature near room temperature. For NdFeB-based ribbons, at the first moment the iron-rich seeds are formed on the contact surface of the ribbons, and on top of these seeds the Nd$_2$Fe$_{14}$B grains are grown to the free surface of the ribbons under high V_T. Since the seeds are formed at the wheel surface temperature which was far below the Curie temperature of Fe (≈770 °C), they can be magnetized and absorb energy from an external magnetic field applied on the wheel surface, which could result in some changes in the ribbon solidification process.
In order to estimate the average grain size with XRD the produced ribbons must be ground into fine powders. The grain size determined by the FWHM of XRD powder diffraction peaks then reflects the average size of grains in the ribbon. Figure 1 presents the field effect on the grain size of ribbons melt-spun at different wheel speeds with and without the field $H_\perp = 1.5$ kOe. This field effect was more pronounced for large wheel speed values. As shown in the inset of Fig. 1, the ribbon melt-spun at 30 m/s without a field and in $H_\perp = 1.5$ kOe has much broadened peaks indicating a decreased grain size.

The grain refinement effect was proved also by TEM observations. For comparison, two ribbons melt-spun at the same condition except field application $H_\perp = 1.5$ kOe were observed and the typical TEM result was presented in Fig. 2. As we see from the TEM images, there is a strong reduction in grain size in the ribbon melt-spun in the field.

The reason for the grain size reduction may be explained by the magnetic field induced seed-size reduction effect. Normally, the critical seed size is controlled by the balance between its volume and surface energy. It is understandable that, an external magnetic field may shift the balance to the smaller volume, in other words, to the smaller critical size of the seeds.

Thus, in the case of ferromagnetic seeds, an external field reduces the critical size of the seeds formed on the contact surface of ribbons and consequently shifts the grain size distribution to the side of lower grain size. However, the estimation of this effect for NdFeB system shows that for fields of strength of several kilo-Oersteds, the reduction of the critical size of seeds is only a few percent. The significant change in grain size observed experimentally and presented in Fig. 1, especially for the large wheel speeds, may be also attributed to the shift of the continuous cooling transformation (CCT) diagram that refines the microstructure or even causes amorphous formation in ribbons at the given cooling rate.

### B. Field induced texture enhancement

As mentioned above, the solidification of the melt begins with the seed formation on the contact surface. If the seeds are ferromagnetic then they are magnetized along the direction of the applied field. The crystallization on the magnetized seeds can result in the texture enhancement in comparison with the case of the conventional melt-spinning process where the crystallization is governed only by the temperature gradient $\nabla T$. The field-induced texture enhancement was clearly observed on XRD patterns of the free surface of ribbons melt-spun in the perpendicular field (Fig. 3). The texture degree $\tau$ is estimated by the ratio $\gamma = (I_{(006)}/I_{(410)})/0.216$, where $I_{(006)}$ and $I_{(410)}$ are the intensities of the

![FIG. 2. The TEM images of the single-phase NdFeB ribbon melt-spun from the starting alloy Nd$_{15}$Fe$_{77}$B$_8$ at the wheel speed $v_w = 25$ m/s (a) without a field and (b) in the field $H_\perp = 1.5$ kOe. From the Fig. 1 the averaged grain size is 22 nm for the ribbon melt-spun in zero field and 19 nm for the ribbon melt-spun in $H_\perp = 1.5$ kOe, respectively. From the TEM images one observes the separate small grain embedded in the matrix with the dislocations inside the ribbon melt-spun without a field and the regular and smaller grains inside the field-assisted melt-spun ribbon.](https://example.com/fig2.png)

![FIG. 3. (Color online) The XRD patterns taken on the free surfaces of single phase ribbons melt-spun at 15 m/s without a field $H = 0$, in $H_\perp = 3$ kOe and $H_\parallel = 3$ kOe. The texture degree $\gamma$ of the ribbon melt-spun in $H = 0$ is 9.9, enhanced to 23.7 in the perpendicular field and declined to 1 in the parallel field.](https://example.com/fig3.png)
peak (006) and (410), respectively and 0.216 is the value of this ratio for the case of non-textured sample. This field-induced texture enhancement has been observed through XRD patterns for all ribbons of single phase and composite when they were melt-spun at low speeds \( v_w \leq 25 \) m/s. This effect was hidden in ribbons melt-spun at larger \( v_w \) when grain size becomes very small. A typical example is shown on Fig. 3 for the single phase ribbon melt-spun from the starting material \( \text{Nd}_{15}\text{Fe}_{77}\text{B}_8 \) at 15 m/s, the texture degree \( \gamma \) gains from 9.9 (in \( H = 0 \)) to 23.7 (in \( H_L = 3 \text{ kOe} \)) to 23.7 (in \( H_L = 3 \text{ kOe} \)).

To examine the field-induced texture enhancement, a similar experiment was also done for the parallel field configuration \( H // \). The XRD pattern taken for the free surface of the ribbon melt-spun in \( H // \) and is presented also in the Fig. 3 showing vanished (00l) reflection and enhanced (410) and (411) peaks, with decreased the texture degree \( \gamma \) to 1. The structural and morphological changes have also led to magnetic properties changes. The magnetization loop measurement of the sample melt spun in a parallel field has a \( M_r/M_s \) ratio of 0.4 and the coercivity \( H_C \) only 0.55 kOe.

C. Effect of fields on the soft phase addition

We also investigated the field effect on the \( \text{Nd}_2\text{Fe}_{14}\text{B}/\text{Fe} \) composite ribbons. The starting alloy \( \text{Nd}_{15}\text{Fe}_{77}\text{B}_8 \) was added with 40%wt. of Fe and was melt-spun at \( v_w = 15.8 \) m/s in \( H_L = 0 \) and 3 kOe. Their magnetization loops shown in Fig. 4 present the positive role of the field in enhancing the inter-phase exchange coupling for the case of high content of the soft phase. For comparison, the ribbon melt-spun in \( H_L = 3 \text{ kOe} \) with 12.6 MG.Oe of the energy product \((BH)_{\text{max}}\) was obtained, while for the ribbon with the same composition melt-spun without a field the \((BH)_{\text{max}}\) was only 1.5 MG.Oe. This suggests again the formation of finer grain size in the composite samples melt-spun in the field as discussed above.

IV. CONCLUSIONS

The magnetic field-assisted melt-spinning technique has been implemented for preparing hard magnetic \( \text{Nd}_2\text{Fe}_{14}\text{B} \)-based ribbons. The obtained experimental results showed that the field application of several kilo Oersteds during the ribbon formation reduces the grain size in the ribbons and enhances the ribbon’s texture in the case the field is perpendicular to the wheel surface. It has been also observed that the field application enhances the exchange coupling in the nanocomposite ribbons. It is promising to use this technique for production of high performance nanocomposite magnets.

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