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# Nanocrystalline $\text{Nd}_2\text{Fe}_{14}\text{B}/\alpha\text{-Fe}$ permanent magnet<sup>①</sup>

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**[Abstract]**  $\text{Nd}_{8.5}\text{Fe}_{75}\text{Co}_5\text{Cu}_1\text{Zr}_3\text{Nb}_1\text{B}_{6.5}$  bonded magnet was prepared by melt-spinning ( $v_s = 18$  m/s) and subsequent heat treatment (670 °C, 4 min). Excellent magnetic properties of the bonded magnet were achieved:  $B_r = 0.68\text{T}$ ,  $iH_c = 620.3$  kA/m,  $(BH)_{\max} = 74$  kJ/m<sup>3</sup>. The addition of Cu and Zr elements shows to be advantageous in improving an intrinsic coercivity and squareness of hysteresis loop, as well as energy product. It has a remarkable remanence enhancement and the isotropic saturation remanence ratio  $M_r/M_s$  is 0.83.

**[Key words]** Nd-Fe-B; permanent magnet; nanocrystalline; grain size

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## 1 INTRODUCTION

Materials with enhanced remanent magnetic polarization were initially prepared by Coehoorn et al.<sup>[1]</sup> from melt-spun  $\text{Nd}_{3.8}\text{Fe}_{77}\text{B}_{19.2}$  ribbons by heat treatment. A similar behavior was also observed in rapidly quenched two-phase samples of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  and  $\alpha\text{-Fe}$ <sup>[2]</sup>. Recently, nanocrystalline Nd-Fe-B consisting of two suitably dispersed ferromagnetic and mutually exchange-coupled phase was widely studied<sup>[3~8]</sup>, one of which is  $\text{Nd}_2\text{Fe}_{14}\text{B}$  that has a high coercive field, while the other may be  $\alpha\text{-Fe}$  providing a high saturation remanence.

The effect of addition of Cu, Zr elements, wheel speeds and heating temperature on the magnetic properties is investigated in the present work.

## 2 EXPERIMENTAL

The alloy composition adopted in this investigation was  $\text{Nd}_{8.5}\text{Fe}_{75}\text{Co}_5\text{Cu}_1\text{Nb}_1\text{Zr}_3\text{B}_{6.5}$  (mole fraction, %). The alloy was initially melted in vacuum induction furnace and subsequently casted into bar, which was subsequently annealed at 1 000 °C for 8 h. For melt-spinning, the alloy bar was crushed into pieces of less than 8 mm in size.

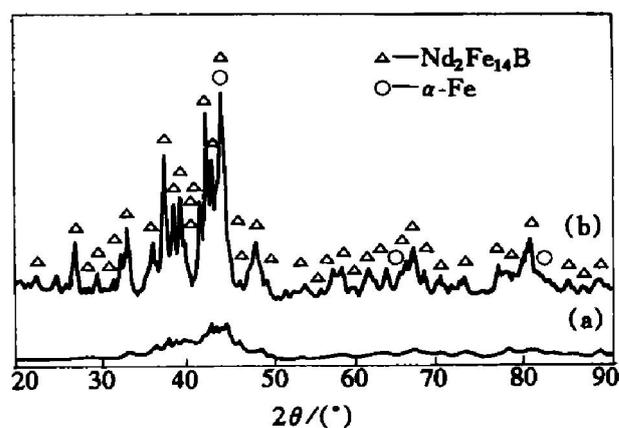
The pieces were melt-spun in ZK-10TCIII vacuum flash hardening furnace with wheel speed ( $v_s$ ) ranging from 15 m/s to 21 m/s. Then the ribbon was subsequently heat treated at 600~710 °C for 2~14 min. The phases were identified by X-ray diffractometry and the mean grain size was determined by using the Scherrer formula. The crystallization temperature for as-quenched ribbon was determined by Perkin Elmer DSC7. The microstructures of the heat treated ribbons were examined by TEM (JEM200CX). The

samples for the measurements of magnetic properties were made by mixing the powder with 4% (mass fraction) epoxy resin and compressing to the shape of  $d 10 \times 10$  mm. The density of bonded compact was about 6.1 g/cm<sup>3</sup>. The magnetic properties of bonded magnets were measured by DGY-2C magnetometer at room temperature.

## 3 RESULTS AND DISCUSSION

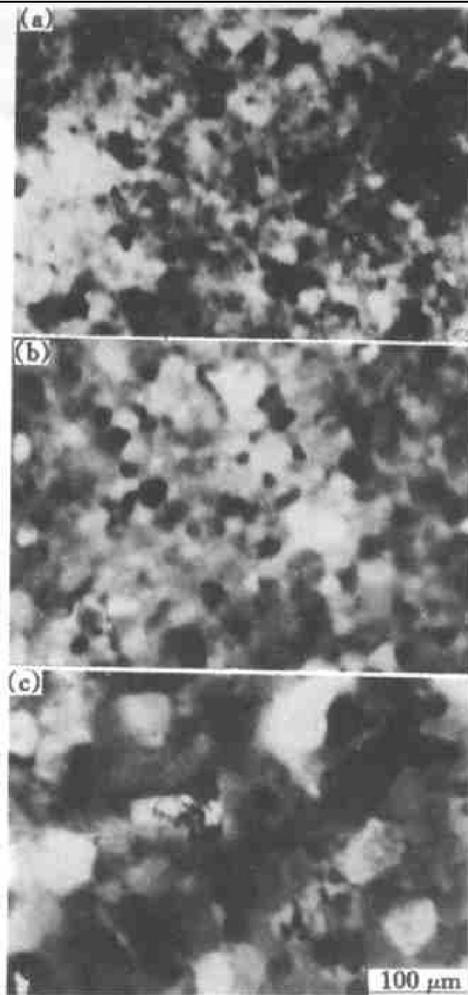
The melt-spun ribbons ( $v_s = 18$  m/s) were found by X-ray diffraction to be amorphous and nanocrystalline (as shown in Fig. 1(a)). Annealing the ribbons at 670 °C for 4 min results in the formation of the tetragonal  $\text{Nd}_2\text{Fe}_{14}\text{B}$ -type crystal structure which coexists with the  $\alpha\text{-Fe}$  phase (as shown in Fig. 1(b)). The mean grain size of the two-phase was about 50 nm.

Fig. 2 shows the TEM micrographs of ribbons



**Fig. 1** XRD patterns of  $\text{Nd}_{8.5}\text{Fe}_{75}\text{Co}_5\text{Cu}_1\text{Nb}_1\text{Zr}_3\text{B}_{6.5}$  alloy  
(a) —Melt-spun ( $v_s = 18$  m/s);  
(b) —Heat treated at 670 °C for 4 min

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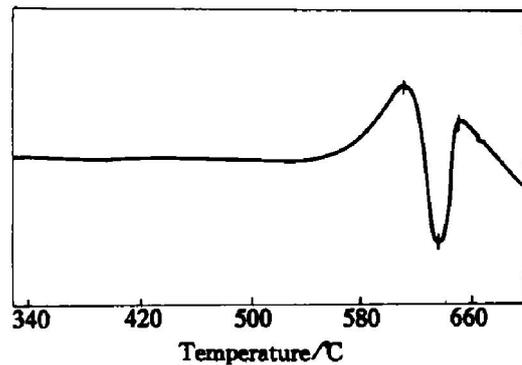


**Fig. 2** TEM micrographs of melt-spun ribbons annealed at different temperatures for 4 min  
(a) -610 °C; (b) -670 °C; (c) -710 °C

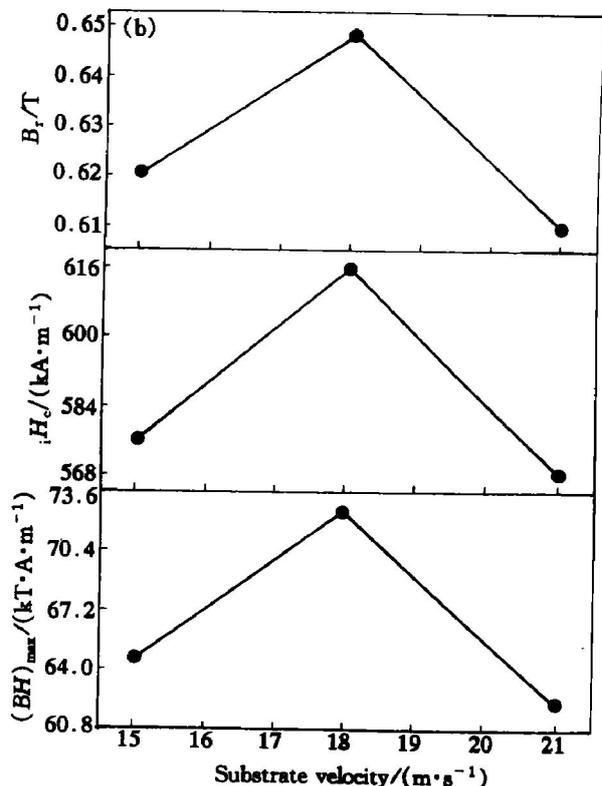
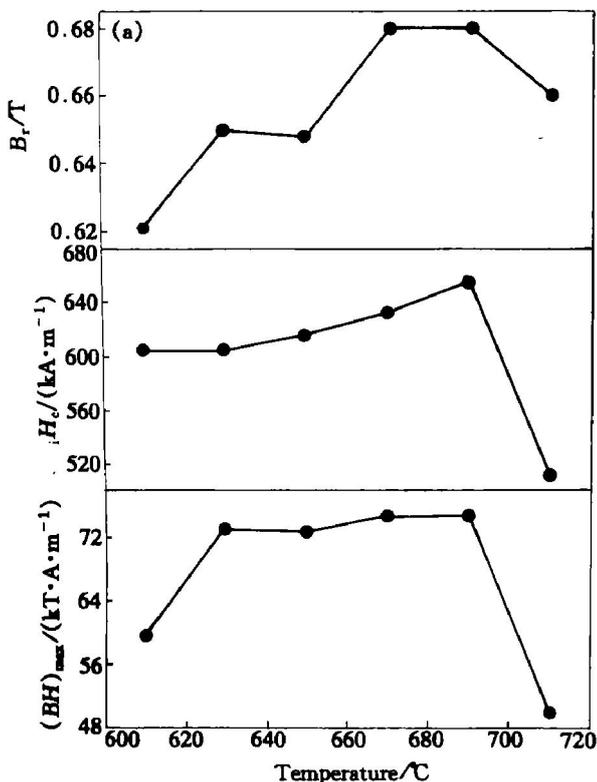
( $v_s=18$  m/s) after being treated at different heating temperatures. After annealing the ribbons at 610 °C for 4 min, the mean grain size was about 20~30 nm (as shown in Fig. 2(a)), and amorphous phase was found coexisting with  $\alpha$ -Fe phase. The mean grain size of ribbons heat treated at 670 °C for 4 min was about 50 nm (as shown in Fig. 2(b)), while the grain size of ribbons annealed at 710 °C for 4 min was larger than 70 nm (as shown in Fig. 2(c)). The grain size determined by XRD was in good agreement with those measured directly by TEM.

The DSC curve of the melt-spun ribbons ( $v_s=18$  m/s) was shown in Fig. 3 at heating rate of 20 °C/min. It can be seen that the crystallization peak is at 629 °C.

Fig. 4(a) shows the relationship between magnetic properties of bonded magnet and the annealing



**Fig. 3** DSC curve for melt-spun ribbons ( $v_s=18$  m/s) with heating rate of 20 °C/min



**Fig. 4** Magnetic properties of bonded magnets vary with annealing conditions  
(a) - $v_s=18$  m/s,  $t=4$  min; (b) - $\theta=670$  °C,  $t=4$  min

temperature for the melt-spun ribbons ( $v_s = 18$  m/s). The magnetic properties of bonded magnet increase slightly with increasing annealing temperature from 610 °C at the beginning then decrease sharply at 710 °C. Excellent magnetic properties of bonded magnet,  $iH_c = 620.3$  kA/m,  $(BH)_{\max} = 74$  kJ/m<sup>3</sup>, were obtained at 670 °C.

Fig. 4 (b) shows that magnetic properties of bonded magnets vary with the wheel speed  $v_s$ . The  $iH_c$  and  $(BH)_{\max}$  of the bonded magnet were sensitive to the wheel speed  $v_s$ .

It is obvious that magnetic properties of bonded magnet were affected by changing the melt-spinning process parameters and heat treatment temperature.

Isotropic saturation remanence ratio  $M_r/M_s > 0.5$  may be considered a criterion for the presence of the exchange-coupled mechanism<sup>[3]</sup>. There is a remarkable remanence enhancement, and the isotropic saturation remanence ratio  $M_r/M_s$  of the bonded magnets which was melt-spun at  $v_s = 18$  m/s and subsequently annealed at 670 °C for 4 min is 0.83.

The addition of Cu and Zr elements shows to be advantageous in improving the intrinsic coercivity and squareness of hysteresis loop, and as well as energy product. It can be seen from Fig. 4 that intrinsic coercivity increases sharply from 429.4 kA/m to 620.3 kA/m and energy product from 64.6 kJ/m<sup>3</sup> to 74 kJ/m<sup>3</sup><sup>[6]</sup>.

Fig. 5 shows irreversible open circuit flux change of the Nd<sub>8.5</sub>Fe<sub>75</sub>Co<sub>5</sub>Cu<sub>1</sub>Zr<sub>3</sub>Nb<sub>1</sub>B<sub>6.5</sub> bonded magnets. The measurement was done at the temperature

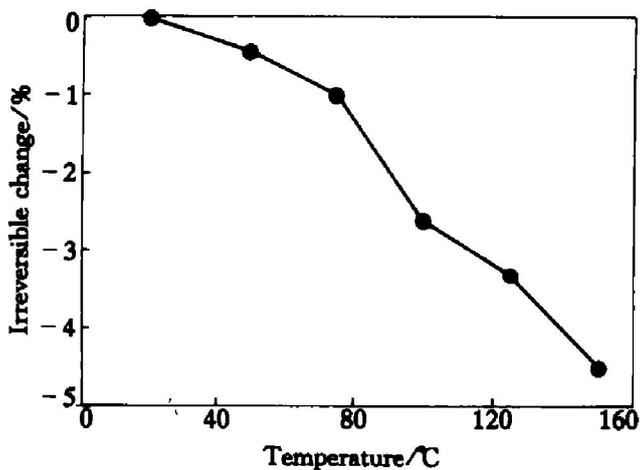


Fig. 5 Irreversible open circuit flux change for Nd<sub>8.5</sub>Fe<sub>75</sub>Co<sub>5</sub>Cu<sub>1</sub>Zr<sub>3</sub>Nb<sub>1</sub>B<sub>6.5</sub> bonded magnet melt-spun ( $v_s = 18$  m/s) then heat treated at 670 °C for 4 min

between room temperature and 150 °C. The holding time was 1 h for each test. The irreversible loss of bonded magnets at 150 °C is less than 5%.

## 4 CONCLUSIONS

1) The nanocrystalline Nd<sub>8.5</sub>Fe<sub>75</sub>Co<sub>5</sub>Cu<sub>1</sub>Zr<sub>3</sub>Nb<sub>1</sub>B<sub>6.5</sub> bonded magnet was prepared by melt-spinning ( $v_s = 18$  m/s) and subsequent heat treatment (670 °C, 4 min). Excellent magnetic properties of the magnet are achieved as  $B_r = 0.68$ T,  $iH_c = 620.3$  kA/m,  $(BH)_{\max} = 74$  kJ/m<sup>3</sup>.

2) The addition of Cu and Zr shows to be advantageous in improving the intrinsic coercivity and squareness of hysteresis loop, and as well as energy product.

3) There is a remarkable remanence enhancement, and the isotropic saturation remanence ratio  $M_r/M_s$  is 0.83.

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