

Influence of Roller Speed on Magnetic Properties and Structures of α -Fe/Nd₂Fe₁₄B Nanocomposite Magnets Prepared by Melt-spinning

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The crystallization behaviours of nanocomposite made by a function of quenching rate (roller speed) were studied. The results showed that there was one step crystallization process for the alloy quenched at roller speed of 32 m/s, which could be shown as, Am (amorphous) + α -Fe+Nd₂Fe₁₄B \rightarrow α -Fe+Nd₂Fe₁₄B. For the alloy quenched at roller speed of 40 m/s, there was two steps crystallization process taking place at different temperatures, which could be shown as, Am \rightarrow α -Fe+Nd₂Fe₂₃B₃+Nd₂Fe₁₄B+Am' \rightarrow α -Fe+Nd₂Fe₁₄B. The presence of transition phase (Nd₂Fe₂₃B₃) was harmful to get fine and uniform grain size during crystallization process. Uniform microstructures and high magnetic properties could be attained for the as-quenched alloy containing less amorphous phase and no presence of transition phase during annealing treatment. For the alloy prepared at roller speed of 32 m/s, the following properties were obtained, $B_r = 0.904$ T, $iH_c = 801$ kA/m, $(BH)_{max} = 122$ kJ/m³ and $M_r/M_s = 0.6$.

Key words : Nanocomposite, Melt-spun ribbons, Annealing, Crystallization, Exchange interaction

1. Introduction

Nanocomposite Nd-Fe-B magnets comprising soft and hard magnetic phases have attracted much attention recent years because of potential permanent magnet development [1-4]. According to traditional Stoner-Wohlfarth model, the biggest M_r/M_s of hard magnetic materials with randomly distributed uniaxial domain is 0.5 [5]. But in the nanocomposite magnets case, when the grain size of soft and hard magnetic phases was in nanoscale, exchange coupled interaction would present between soft and hard magnetic phases and the M_r/M_s value would be higher than 0.5. Researchers are very interested in this phenomenon because it directs a new way for improving magnetic properties. Many studies indicate that the magnetic properties of nanocomposite magnets depend strongly on their microstructure [6-8]. Fischer *et al.* proposed an optimum microstructure consisting of small soft magnetic grain with size of about 10 nm intermixed with hard magnetic grains with a mean grain diameter of about 20 nm [9]. The principle magnetic properties of magnets with ideal microstructure can achieve 100 MGOe. Therefore, it is very significant to study a uniform

microstructure with very fine grains of the soft phase uniformly distributed in the nanocomposite magnets in order to obtain the optimum coupling. In this work, we studied crystallization process of as-quenched alloy with different cooling rate and influence of roller speed on magnetic properties and structures of nanocomposite magnets.

2. Experiment

Alloys with compositions of Nd_{11.5}Fe_{76.7}B_{5.3}Co_{6.0}Zr_{0.5} were prepared in a vacuum induction furnace in an Ar atmosphere. Cast ingots were crushed into small pieces with dimension of about 10 mm and then the ingots were melt-spun at several roller speeds to produce ribbons with different microstructures by arc melt spinning process. The quenched ribbons were subsequently annealed by tube furnace filled with Ar gas. The magnetic properties of samples were tested by pulsed magnetometer. The crystallization temperatures of the as-spun ribbons were determined by using a differential thermal analysis (DTA). Structural studies were carried out by means of an X-ray diffractometry (XRD) with Cu K α radiation and a transmission electron microscope (TEM).

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3. Results and Discussion

3.1. Microstructures of as-quench ribbons

The XRD curves of as-quenched ribbons with different cooling roller speeds are shown in Fig. 1. The results corresponding to the roller velocity of 25, 32 and 40 m/s were α -Fe+Nd₂Fe₁₄B, Am+ α -Fe+Nd₂Fe₁₄B and Am, respectively. The cooling rate is mainly controlled by the tangential speed of the spinning roller. The faster cooling roller speed is, the faster cooling rate is produced. Thus, the roller speed influences the microstructures of ribbons. When the roller speed was very slow (25 m/s), ribbons had enough time to crystallize. There were only crystallization phases to be attained in the ribbons. When the roller speed increases, the cooling rate became so fast that partial ribbons could not finish crystallization process. Therefore, amorphous phases begin to appear in the samples like sample of 32 m/s. When the roller speed continually increased and achieved 40 m/s, all of ribbons could not finish crystallization process. The as-quenched samples were composed of full amorphous phase.

3.2. Crystallization process of as-quenched ribbons with different wheel speeds

The DTA curves of ribbons with different roller speeds are shown in Fig. 2. The curve (a) and (b) show the DTA curves of the amorphous (40 m/s) and partial amorphous (32 m/s) samples, respectively. In curve (a) case, there were two exothermic peaks. It indicated that the crystallization of the amorphous alloy took place through two steps at different temperatures. The first stage occurred at about 590°C and the second one occurred at about 630°C. For the curve (b), there was only one peak at 570°C. It

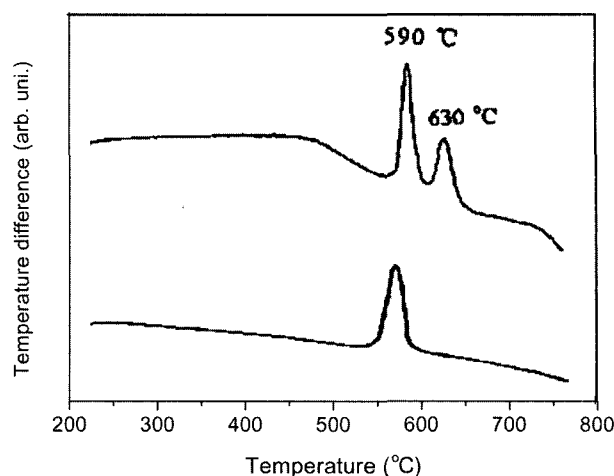


Fig. 2. DTA curves of the ribbons at roller speed of (a) 40 m/s and (b) 32 m/s.

indicated that only one-step took place in the course of the crystallization. In order to examine the crystallization reaction corresponding to each exothermic peak, the as-quench alloys were heated at the peak temperature for 3.6 ks and then the resulting samples were identified by XRD. For ribbons of 32 m/s, there were α -Fe and Nd₂Fe₁₄B phase to be detected in annealed samples. It indicated that crystallization occurred at 570°C and the reaction was: Am+ α -Fe+Nd₂Fe₁₄B \rightarrow α -Fe+Nd₂Fe₁₄B. For the ribbons quenched at roller speed of 40 m/s, there were α -Fe, Nd₂Fe₂₃B₃, Nd₂Fe₁₄B and Am' phases to be detected in the samples annealed at 590°C, and there were α -Fe and Nd₂Fe₁₄B to be detected in the samples annealed at 630°C. It indicated that the reaction corresponding to the two exothermic peaks are: Am \rightarrow α -Fe+Nd₂Fe₂₃B₃+Nd₂Fe₁₄B+Am' \rightarrow α -Fe+Nd₂Fe₁₄B.

3.3. Magnetic properties and structures of annealed samples

The as-quenched samples with different cooling rate were annealed in optimal heating treatment processes. The magnetic properties of the annealed samples are shown in Table 1. It was found that higher magnetic properties were attained in the samples made by cooling roller speed of 32 m/s. The best magnetic properties: $B_r = 0.904$ T, $H_c = 801$ kA/m, $(BH)_{max} = 122$ kJ/m³ and $M_r/M_s = 0.6$, were achieved for the sample. The structures of annealed samples were detected by means of TEM. The results are shown in Fig. 3. It was observed that there were coarse grains in the samples made by cooling roller speed of 40 m/s and there were more uniform grains in the samples made by cooling roller speed of 32 m/s.

The main reason of coarse grains in the samples of 40 m/s was found in the presence of transition phase during

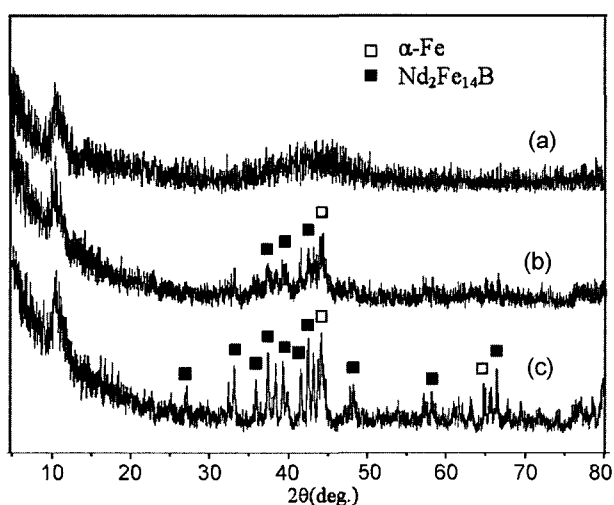
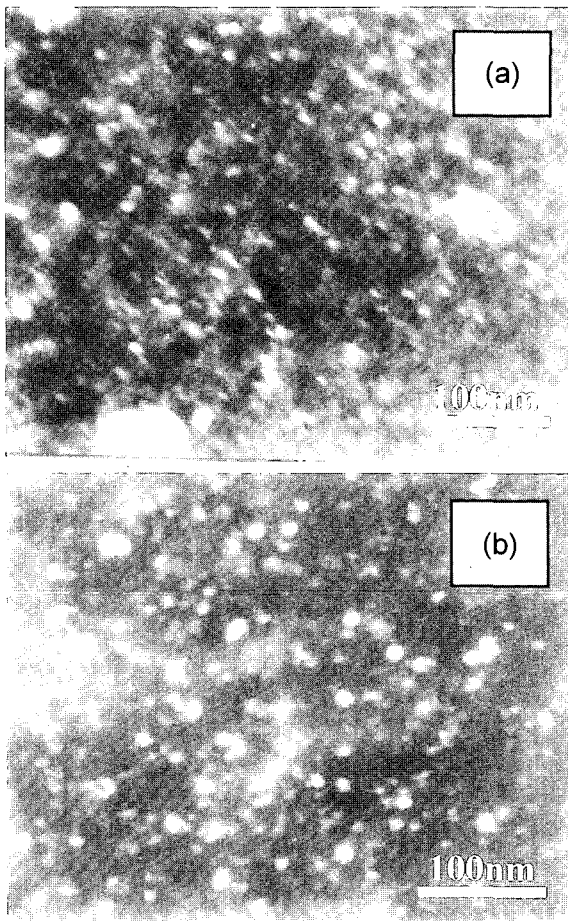


Fig. 1. XRD patterns of the as-quenched samples with cooling roller speed of (a) 40 m/s, (b) 32 m/s and (c) 25 m/s.

Table 1. The magnetic properties of the annealed samples.

| Sample | B_r (T) | | iH_c (kA/m) | | $(BH)_{max}$ (kJ/m ³) | |
|--------|-------------|----------|---------------|----------|-----------------------------------|----------|
| | As-quenched | Annealed | As-quenched | Annealed | As-quenched | Annealed |
| 25 m/s | 0.79 | 0.79 | 785 | 797 | 88.5 | 90 |
| 32 m/s | 0.74 | 0.90 | 690 | 801 | 51.2 | 122 |
| 40 m/s | 0.53 | 0.84 | 210 | 781 | 1.9 | 102 |

**Fig. 3.** TEM micrographs of the annealed samples produced by cooling rate of (a) 40 m/s and (b) 32 m/s.

the crystallization process. The nuclei of α -Fe, $Nd_2Fe_{14}B$ and the transition phase ($Nd_2Fe_{23}B_3$) first formed and then the nuclei clustered and grew. Some of the nuclei of α -Fe and $Nd_2Fe_{14}B$, absorbing on surface of transition phase, grew faster because transition phases decomposed to α -Fe and $Nd_2Fe_{14}B$ with increasing temperature and accelerated the growth speed of the grain. Therefore, the grains around the transition were easy to grow bigger. So, there were some coarse grains to appear in the samples. Due to presence of coarse grains, the exchange coupling was weak and resulted in the lower magnetic properties. For 32 m/s sample, there was no transition phase to present and the former α -Fe and $Nd_2Fe_{14}B$ could be as nuclei for

a rapid nucleation during annealing treatment. Therefore, a structure of uniform and fine grain with nanosize exhibited in annealed sample. The ideal structure resulted in optimal magnetic properties to be attained. To the samples with lower cooling rate (cooling roller speed of 25 m/s), the as-quenched samples were crystallized state and the as-quenched ribbons were thicker. Therefore, quenching condition between roller side and free side was different and the microstructures were not uniform on transect direction. After the heating treatment, there was broad grain size distribution in the samples. It was impossible to control all of grains in nanoscale. The overgrown grains resulted in decreasing exchange coupling and the lower magnetic properties.

These results show that the as-quenched ribbons with controlled structures, containing less amorphous phase and no transition phase, can attain optimal magnetic properties after annealing treatment.

4. Conclusion

There were different crystallization processes to take place between samples quenched at roller speed of 32 m/s (containing partial amorphous) and samples quenched at roller speed of 40 m/s (containing entire amorphous). There was one-step crystallization process for the samples of 32 m/s, which could be shown as, $Am + \alpha\text{-Fe} + Nd_2Fe_{14}B \rightarrow \alpha\text{-Fe} + Nd_2Fe_{14}B$. For the samples of 40 m/s, there was two-step crystallization process to take place at different temperature, which could be shown as, $Am \rightarrow \alpha\text{-Fe} + Nd_2Fe_{23}B_3 + Nd_2Fe_{14}B + Am' \rightarrow \alpha\text{-Fe} + Nd_2Fe_{14}B$.

After annealing treatment, uniform microstructure and high magnetic properties of nanocomposite magnets could be attained for the as-quenched alloy containing less amorphous phase and no transition phase in annealing treatment. For the alloy prepared at roller speed of 32 m/s, the following properties were obtained, $B_r = 0.904$ T, $iH_c = 801$ kA/m, $(BH)_{max} = 122$ kJ/m³ and $M_r/M_s = 0.6$.

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