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Differences in the structure and magnetic properties of $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$ (X=Nb, Zr) ribbons by conventional and microwave-assisted annealing treatment

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Abstract: Amorphous ribbons with $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$(X=Nb, Zr) nominal composition were annealed by conventional and microwave-assisted annealing furnaces, respectively. The thermal decomposition process, structure and magnetic properties of products were characterized by a thermal differential scanning calorimeter (DSC), X-ray diffraction (XRD) and a vibrating sample magnetometer (VSM). The addition of Nb and Zr increased the glass-forming ability (GFA) of as-spun ribbons. The proportion of $Nd_2Fe_{14}B$ and α-Fe could be adjusted with power from 800 to 2000 W in microwave annealing process, during which the well-coupling between the soft and hard magnetic phase and higher coercivity reached up to 780.2 and 815.4 kA/m for (Nb,Zr)-doped alloys. The best magnetic properties of ribbons could be obtained by annealing at 650 ºC for 10 min under the microwave power of 2000 W.

Keywords: magnetic properties; α-Fe/$Nd_2Fe_{14}B$; crystallization kinetics; microwave-assisted annealing; rare earths

Nd-Fe-B magnets, composed of α-Fe soft magnetic phase and $Nd_2Fe_{14}B$ hard magnetic phase[1–3], were first reported by Coehoorn et al. in 1989[4], which have attracted great attention because of their excellent magnetic properties[5–7]. The interaction coupling is a short range effect in soft and hard magnetic phase of the $Nd_2Fe_{14}B$/α-Fe composite permanent magnetic materials[8,9]. A fine and uniform microstructure is a key factor improving magnetic properties. The usual way of optimizing microstructure is process optimization[10–12] or elements addition[13–15]. The addition of elements can increase the glass-forming ability (GFA) and thermal stability of ribbons, refine and homogenize the microstructure and increase magnetic properties of the $Nd_2Fe_{14}B$/α-Fe nanocomposite magnets[16–18]. The annealing conditions have great effect on microstructure and magnetic properties when the composition of alloys is determined.

Conventional heating is an indirect heating process through the heat conduction and convection between heating devices and materials, with a temperature gradient from external to internal. Microwave heating is a selective heating process in which materials couple with special bands of microwave. Microwave heating generates heat within the material first and then heats the entire volume, which is applied to ceramics, semiconductors, inorganic and polymeric materials[19–22]. Compared with the conventional heating, the microwave heating is more favorable for the improvement of the diffusion of ions, grain growth and the physical properties[23]. Wang et al.[24] synthesized $CoFe_2O_4$ ferrite with ultra fine microstructure by microwave calcination. Kotagiri et al.[25] studied the effect of microwave annealing on the magneto impedance of $Fe_{66}Ni_7Si_7B_{20}$ ribbons, resulting in uniform grain growth and the enhancement of the magneto impedance of the ribbons. According to the anisotropic magnetic permeability of $Nd_2Fe_{14}B$, Iwabuchi et al.[26] reported that microwave process was beneficial to the *c*-axis oriented and nano-sized columnar grains through the crystallization of amorphous Nd-Fe-B.

In this work, the differences in structure, phase transformation and magnetic properties of $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$ (X=Nb, Zr) ribbons annealed by conventional and microwave-assisted annealing treatment were investigated.

1 Experimental

The ingots with nominal composition of $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$ (X=Nb, Zr) were prepared by non-consumably arc-melting the elements Nd, Nb, Zr, Fe

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and FeB (B 20.05 at.%) under a high-purity Ar atmosphere. The ingots were remelted four times to ensure homogeneity and crushed into small pieces with lengths of 5–7 mm subsequently. These samples were re-melted and spun onto copper rolls with a wheel speed of 22 m/s. The uniform as-spun ribbons (about 5 cm long, 30 μm thick and 0.9 mm wide) were annealed by a tube vacuum furnace at 650 ºC for 5 min in vacuum with 5×10^{-3} Pa under magnetic field with intensity from 0 to 0.6 T. In order to study the effect of microwave-assisted annealing on crystallization, the uniform as-spun ribbons with copper sheet wrap were sealed by a quartz tube to maintain a vacuum with 5×10^{-3} Pa, and then annealed at 650 ºC for 5 min under power of 800, 1500 and 2000 W for 5, 10 and 15 min using a HAMiLab-V3 microwave furnace with a frequency of 2.45 GHz, respectively. Thermal analysis was employed to characterize the as-spun ribbons using a differential scanning calorimeter (DSC) at heating rates of 5, 20 and 40 K/min, respectively. The structure of the as-spun and annealed ribbons was identified by X-ray diffraction (XRD) (D/max-γB) with Cu-Kα radiation. The magnetic properties of the ribbons were measured by a vibrating sample magnetometer (VSM) with a maximum applied magnetic field of 2.0 T.

2 Results and discussion

Fig. 1 shows the X-ray diffraction patterns of $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$ (X=Nb, Zr) as-spun ribbons. The patterns of the as-spun ribbons present a broad non-crystalline peak with few weak diffraction peaks, indicating a small amount of crystalline phase.

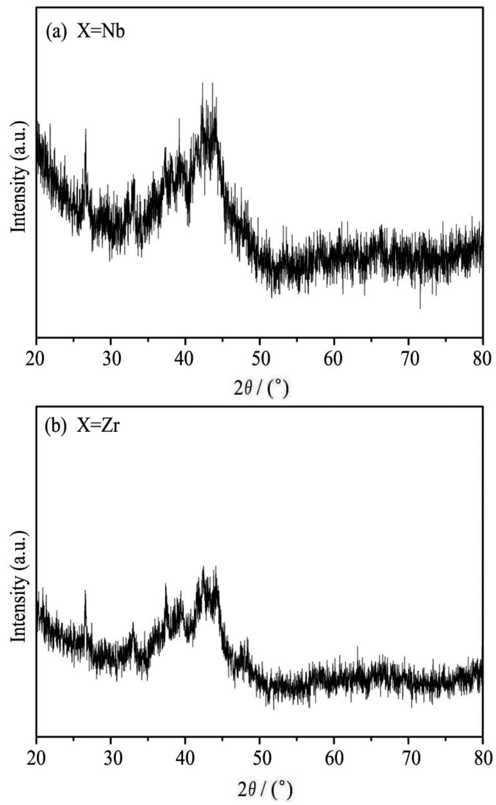

Fig. 1 X-ray diffraction patterns of the as-spun $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$ (X=Nb, Zr) ribbons
(a) X=Nb; (b) X=Zr

Fig. 2 shows DSC curves of the $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$ (X=Nb, Zr) as-spun ribbons heated from room temperature to 1000 ºC at heating rates of 5, 20 and 40 ºC/min, respectively. One exothermic peak can be found, which reveals that the precipitation of soft and hard magnetic phases is formed at the same temperature. The synchronous growth of α-Fe phase and $Nd_2Fe_{14}B$ phases is conducive to avoiding the coarsening of the first precipitated phase and obtaining the fine and homogeneous microstructure. With the increase of heating rate, the crystallization characteristic temperature of as-spun ribbons increases, presenting obvious crystallization kinetics characteristics. Crystallization effective activation energy is a key parameter of the thermal stability for the amorphous alloy, which reflects the statistical average of the activation energy in crystallization process. The higher the effective activation energy value is, the smaller the crystallization tendency of the amorphous alloy is. The effective activation energy of amorphous alloy was obtained by Kissinger equation[27] as follows:

$$\ln\frac{T^2}{B}=\frac{E}{RT}+C \quad (1)$$

where B is the heating rate, T is the crystallization characteristic temperature, E is the crystallization activation

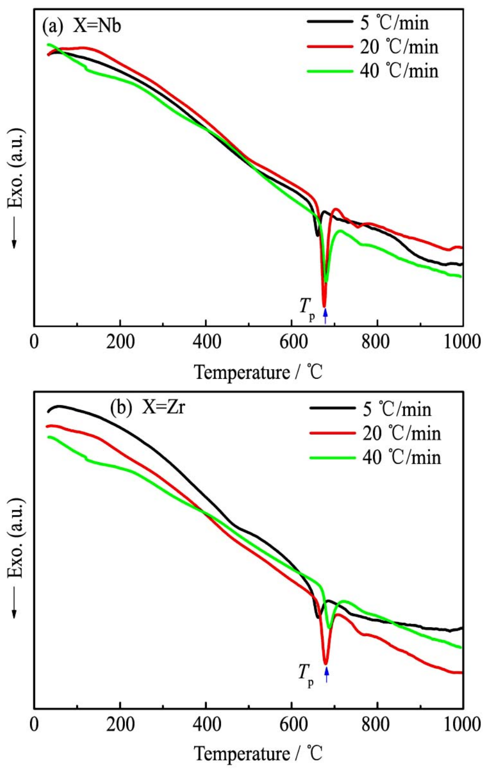

Fig. 2 DSC curves of the as-spun $(Nd_{0.75}Pr_{0.25})_{9.5}Fe_{76}X_4B_{10.5}$ (X=Nb, Zr) ribbons
(a) X=Nb; (b) X=Zr