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The Effect of Annealing in a Magnetic Field on the Microstructures and Magnetic Properties of $(\text{Nd}_{0.8}\text{RE}_{0.2})_{2.2}\text{Fe}_{12}\text{Co}_2\text{B}$ (RE = La, Ce) Alloys

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Abstract: The development of high-performance and cost-effective Nd-Fe-B permanent magnets is crucial to meet the ever-growing requirements of renewable and clean energy applications. Here, we use low-cost and highly abundant La and Ce to partially substitute Nd and investigate the effect of annealing treatment with a 1 tesla (T) magnetic field on the microstructures and magnetic properties of $(\text{Nd}_{0.8}\text{RE}_{0.2})_{2.2}\text{Fe}_{12}\text{Co}_2\text{B}$ (RE = La, Ce) ribbons. The remanence (B_r) and maximum energy product $((\text{BH})_{\text{max}})$ of studied alloys can be improved by magnetic field annealing. The respective B_r and $(\text{BH})_{\text{max}}$ of annealed $(\text{Nd}_{0.8}\text{La}_{0.2})_{2.2}\text{Fe}_{12}\text{Co}_2\text{B}$ alloy are increased to 0.86 T and 124 kJ/m³. In comparison to melt-spun $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{2.2}\text{Fe}_{12}\text{Co}_2\text{B}$ alloy, the B_r and $(\text{BH})_{\text{max}}$ of the magnetic field-annealed alloy are improved by 5% and 8%. The underlying mechanism of improved magnetic properties of La- and Ce-substituted alloys is different. The interaction magnetic domain size in $(\text{Nd}_{0.8}\text{La}_{0.2})_{2.2}\text{Fe}_{12}\text{Co}_2\text{B}$ alloy can be increased by magnetic field annealing, leading to the enhancement of exchange coupling interaction, which results to the improvement in B_r and $(\text{BH})_{\text{max}}$. In the $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{2.2}\text{Fe}_{12}\text{Co}_2\text{B}$ alloy, the concentration of (Fe + Co) of ferromagnetic intergranular phase is increased after magnetic field annealing, resulting in the increase in B_r and $(\text{BH})_{\text{max}}$.



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Keywords: rare-earth permanent alloy; magnetic field annealing; magnetic property; intergranular phase

1. Introduction

Nd-Fe-B permanent magnets have been widely used in clean energy applications such as wind turbines and hybrid electric vehicles due to their excellent hard magnetic properties [1,2]. The ever-growing demand for high-performance Nd-Fe-B magnets leads to the increasing consumption of key rare-earth (RE) elements such as Nd, Pr, and Dy. However, these key RE raw materials exhibit high prices and limited natural resources, resulting in the increase in the manufacturing cost [3]. Therefore, La and Ce elements are usually chosen to substitute for Nd because of their low prices and abundant resources [4,5]. Nevertheless, the intrinsic magnetism of $\text{La}_2\text{Fe}_{14}\text{B}$ and $\text{Ce}_2\text{Fe}_{14}\text{B}$ is inferior to that of $\text{Nd}_2\text{Fe}_{14}\text{B}$, leading to the deterioration of magnetic performance [6]. Hence, the development of high-performance and cost-effective Nd-Fe-B magnets, with partial substitution of Nd by La or Ce, has attracted more attention in the recent years [7–10].

To improve the magnetic properties of Ce- and La-substituted Nd-Fe-B magnets, two main strategies are employed to optimize their microstructures. One strategy is termed as grain boundary diffusion engineering in which Pr-Cu or Nd-Cu alloys are used to trigger

grain boundary diffusion process, leading to the increase in coercivity (H_{ci}) [11–14]. The other strategy is the modification of non-magnetic and ferromagnetic intergranular phases. The non-magnetic intergranular phase consists of more than 65 at.% RE elements, leading to the increase in coercivity by isolating $Nd_2Fe_{14}B$ grains [15,16]. In contrast, at least 65 at.% of ferromagnetic elements (Fe and/or Co) is present in the ferromagnetic intergranular phase, resulting in the improvement in remanence (B_r) by strengthening the exchange coupling interaction [17]. Tang et al. reported that the H_{ci} of $(Nd_{0.9}Ce_{0.1})_2(FeCo)_{14}B$ magnet could be enhanced because a higher concentration of rare earth was present in the intergranular phase, leading to a stronger pinning effect [18]. Kim et al. improved the magnetic properties of Ce/La-substituted Nd-Fe-B magnets by tuning the Ce and La content in the intergranular phase [19].

Since Nd-Fe-B melt-spun ribbons usually used as a precursor of magnets (e.g., bonded magnets and hot-pressed magnets), the microstructural optimization of melt-spun ribbons is critical for improving the magnetic performance of Nd-Fe-B magnets. La- and Ce-substituted Nd-Fe-B melt-spun ribbons have attracted considerable interest as they contain less-expensive rare-earth elements. The strategy of modifying microstructures is also effective in these ribbons. For example, Hussain et al. investigated $(Nd_{1-x}La_x)_{10}Fe_{84}B_6$ ($x = 0-0.7$) ribbons and found that $x = 0.1$ alloy showed the highest H_{ci} due to fine-grain microstructure [20]. The $(Nd_{1-x}La_x)_{13}Fe_{81}B_6$ ($x = 0-0.5$) ribbons were investigated, and the magnetic property of $x = 0.4$ alloy was improved because of its refined and uniform microstructure [21]. Zhao et al. found that the magnetic properties of $(Ce_{1-x}Nd_x)_{16}Fe_{78}B_6$ ($x = 0-1$) ribbons were sensitive to the Nd/RE ratio, and the large H_{ci} obtained in high Ce-containing alloys was due to RE segregation at grain boundaries [22]. Tao et al. increased the H_{ci} of $(Nd_{0.5}Ce_{0.5})_{30.5}Fe_{68.3}Cu_{0.2}B_{1.0}$ melt-spun ribbons by forming a continuous Ce-rich intergranular phase [23]. It is reported that high H_{ci} is obtained in $(Nd_{0.8}Ce_{0.2})_{12.2}Fe_{81.6}B_{6.2}$ ribbons by controlling grain growth and the formation of a single-domain structure [24].

Notably, Pathak et al. reported that a high H_{ci} (1409 kA/m) was achieved in melt-spun $(Nd_{0.8}Ce_{0.2})_{2.4}Fe_{12}Co_2B$ ribbons, which is larger than that of Dy-containing ribbons [25]. However, the magnetic mechanism is not clearly revealed. Our previous study showed that the high coercivity of $(Nd_{0.8}Ce_{0.2})_{2.4}Fe_{12}Co_2B$ ribbons was ascribed to strong domain wall pinning [26]. Since magnetic field annealing treatment is an effective approach to improve the magnetic properties of rare-earth permanent magnets [27–29], we also used magnetic field annealing treatment in our previous study [26] to optimize the microstructure and found that the B_r and $(BH)_{max}$ of $(Nd_{0.8}Ce_{0.2})_{2.4}Fe_{12}Co_2B$ ribbons could be increased by magnetic field annealing. This is due to changes in distribution and chemistry of the ferromagnetic intergranular phase. In this study, we decrease the RE content from $(Nd_{0.8}Ce_{0.2})_{2.4}Fe_{12}Co_2B$ to $(Nd_{0.8}Ce_{0.2})_{2.2}Fe_{12}Co_2B$ to further lower the cost of raw materials. Meanwhile, cost-effective La is also chosen to partially substitute Nd. Hence, the magnetic properties and microstructure of $(Nd_{0.8}RE_{0.2})_{2.2}Fe_{12}Co_2B$ (RE = La, Ce) ribbons are investigated in this study. Moreover, the scanning transmission electron microscope (STEM) and Lorentz microscope are used to illustrate the effect of magnetic field annealing treatment on magnetic properties and microstructures.

2. Materials and Methods

Alloy buttons with nominal composition of $(Nd_{0.8}RE_{0.2})_{2.2}Fe_{12}Co_2B$ (RE = La, Ce) ($(Nd_{0.8}RE_{0.2})_{12.79}Fe_{69.77}Co_{11.63}B_{5.81}$ in at.%) were prepared from pure metals such as Nd, La, Ce, Fe, and Co (purity > 99.95%) and FeB alloy with 18.69 wt.% boron by arc melting in an argon (Ar) atmosphere. The Ti getter was used to remove unwanted oxygen, and all ingots were re-melted four times to ensure good chemical homogeneity. A small portion of an ingot weighing 5 g was re-melted by induction melting under an Ar atmosphere

and ejected onto a rotating copper wheel at 15 m/s. An infrared thermometer was used to measure the quenching temperature. The respective chamber pressure and quenching temperature during melt spinning were 0.05 MPa and $1588 \text{ K} \pm 5 \text{ K}$. The magnetic annealing treatments were performed in the range of 573–773 K for 15 min with an applied magnetic field of 1 tesla (T) by a vacuum magnetic annealing furnace (F800-35/EM7, East Changing technologies, Beijing, China). The longitudinal axis of ribbons was parallel to the direction of the applied magnetic field. The magnetic properties of ribbons were determined at room temperature with an applied magnetic field of 5 T using a multifunctional physical property measurement system (PPMS, Quantum Design, San Diego, CA, USA). The X-ray diffraction (XRD) patterns in the scanning angle of $20\text{--}90^\circ$ were measured by a D/max-2550 diffractometer with Cu $K\alpha$ radiation (Rigaku Corporation, Akishima-Shi, Japan). A Helios 600i focused ion beam (FIB, FEI Corporate, Hillsboro, OR, USA) was used to prepare transmission electron microscopy (TEM) samples. Scanning transmission electron microscopy (STEM) images, including those of the high-angle annular dark-field (HAADF) micrograph, were collected by JEM-2100F at 200 kV (JEOL Ltd., Tokyo, Japan). The magnetic domain structure was observed by the JEM-2100F Lorentz microscope in the Fresnel mode (JEOL Ltd., Tokyo, Japan).

3. Results and Discussions

3.1. Effect of Magnetic Field Annealing on Magnetic Properties

The studied $(\text{Nd}_{0.8}\text{RE}_{0.2})_{2.2}\text{Fe}_{12}\text{Co}_2\text{B}$ (RE = La, Ce) alloys are hereafter termed as the Nd-La alloy and the Nd-Ce alloy. The hysteresis loops of melt-spun ribbons are shown in Figure 1a. The H_{ci} of the Nd-La alloy is 914 kA/m, which is lower than 1090 kA/m of the Nd-Ce alloy. However, the B_r and $(BH)_{\max}$ of the Nd-La alloy are 0.84 T and 117 kJ/m^3 , which are higher than 0.78 T and 98 kJ/m^3 of the Nd-Ce alloy, see Table 1. It indicates that the partial substitution of Nd by La or Ce produces a difference in the magnetic property. The reason for this difference is discussed in Section 3.3.

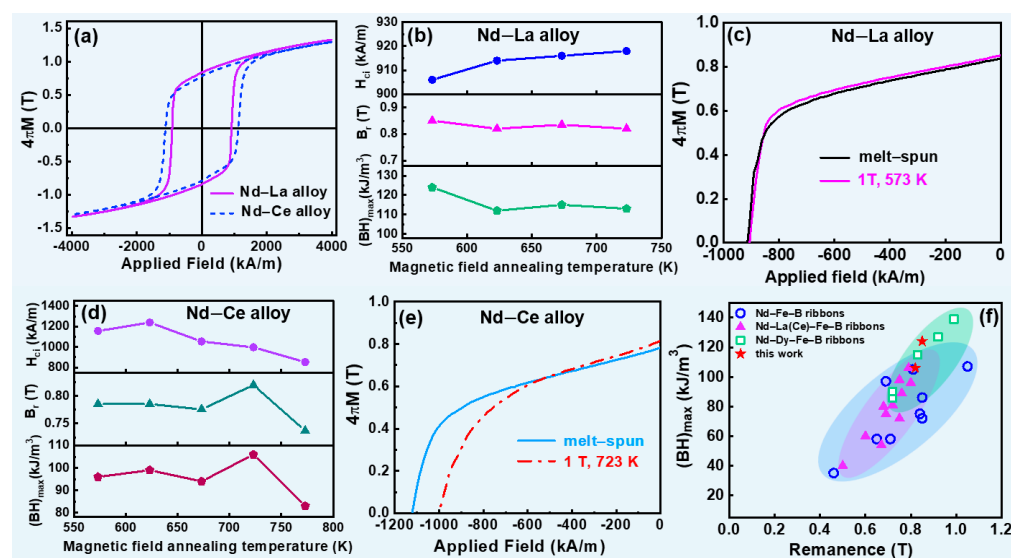


Figure 1. (a) The hysteresis loops of the Nd-La and Nd-Ce alloys; (b) the magnetic properties of the Nd-La alloy as a function of magnetic annealing temperatures; (c) the demagnetization curves of the melt-spun and magnetic field-annealed Nd-La alloy at 573 K; (d) the magnetic properties of the Nd-Ce alloy as a function of magnetic annealing temperatures; (e) the demagnetization curves of the melt-spun and magnetic field-annealed Nd-Ce alloy at 723 K; and (f) the comparison of B_r and $(BH)_{\max}$, where red pentagrams represent alloys assessed in this study.

Table 1. The intrinsic coercivity (H_{ci}), remanence (B_r), and maximum energy product ($(BH)_{max}$) of the melt-spun and magnetic field-annealed Nd-La and Nd-Ce alloys at an optimum magnetic field annealing temperature.

Alloy	Condition	H_{ci} (kA/m)	B_r (T)	$(BH)_{max}$ (kJ/m ³)
Nd-La	melt-spun	914	0.84	117
	1 T, 573 K/15 min	906	0.86	124
Nd-Ce	melt-spun	1090	0.78	98
	1 T, 723 K/15 min	997	0.82	106

To further improve the magnetic properties of the Nd-La and Nd-Ce alloys, annealing treatment in a 1 T applied magnetic field is performed in the temperature range of 573–773 K. The hysteresis loops of the magnetic field-annealed Nd-La alloy at different temperatures are shown in Figure S1a of Supplementary Materials. Figure 1b shows the H_{ci} , B_r , and $(BH)_{max}$ of the Nd-La alloy as a function of magnetic field annealing temperatures. The optimum temperature is 573 K because the largest $(BH)_{max}$ is achieved. The demagnetization curves in Figure 1c show that the B_r and $(BH)_{max}$ of the Nd-La alloy are increased to 0.86 T and 124 kJ/m³ after magnetic field annealing, and the H_{ci} is slightly decreased by 0.8% to 906 kA/m. It indicates that the B_r and $(BH)_{max}$ of the Nd-La alloy can be improved by magnetic field annealing without sacrificing H_{ci} . The hysteresis loops of the Nd-Ce alloy after magnetic field annealing at various temperatures are presented in Figure S1b of Supplementary Materials. The optimum magnetic field annealing temperature of the Nd-Ce alloy is 723 K; see Figure 1d. Figure 1e shows that the B_r and $(BH)_{max}$ of the magnetic field-annealed Nd-Ce alloy is increased by 5% to 0.82 T and by 8% to 106 kJ/m³ when compared with the B_r (0.78 T) and $(BH)_{max}$ (98 kJ/m³) of the melt-spun alloy. For comparison, Figure 1f exhibits the B_r and $(BH)_{max}$ of Nd-Fe-B ribbons [30–35], La- or Ce-substituted (named as Nd-La(Ce)-Fe-B) ribbons [22,25,36–40], Dy-containing (termed as Nd-Dy-Fe-B) ribbons [41–44], and magnetic field-annealed ribbons in this study (marked by red pentagrams). It is found that the studied alloys in current study exhibit good magnetic properties.

3.2. Effect of Magnetic Field Annealing on Microstructure

Figure 2 shows the XRD patterns of the melt-spun and magnetic field-annealed Nd-La and Nd-Ce alloys. The single Nd₂Fe₁₄B (2:14:1) phase is observed in the studied alloys, indicating that the phase constitution is not changed after magnetic field annealing. Nd₂Fe₁₄B shows a tetragonal structure (space group $P4_2/mnm$, $a = 8.80$ Å, $c = 12.20$ Å) [6]. The lattice parameter (a , c) is obtained by Le Bail refinement. The respective value of a and c of the melt-spun Nd-La alloy is 8.79 Å and 12.21 Å and that of the melt-spun Nd-Ce alloy is 8.80 Å and 12.21 Å. It indicates that the partial substitution of Nd by La or Ce has a marginal effect on the lattice parameters of Nd₂Fe₁₄B phase. After magnetic field annealing, the lattice parameters of the Nd-La alloy ($a = 8.78$ Å, $c = 12.19$ Å) and Nd-Ce alloy ($a = 8.81$ Å, $c = 12.20$ Å) both show a slight change when compared with those of the melt-spun Nd-La and Nd-Ce alloys. Using the Scherrer formula [45], the respective average grain sizes of the melt-spun and magnetic field-annealed Nd-La alloy are approximately 24 nm and 32 nm. In the melt-spun and magnetic field-annealed Nd-Ce alloy, they are estimated to be 44 nm and 34 nm. It suggests that the grain size in the Nd-La alloy is increased, while it is reduced in the Nd-Ce alloy after magnetic field annealing.

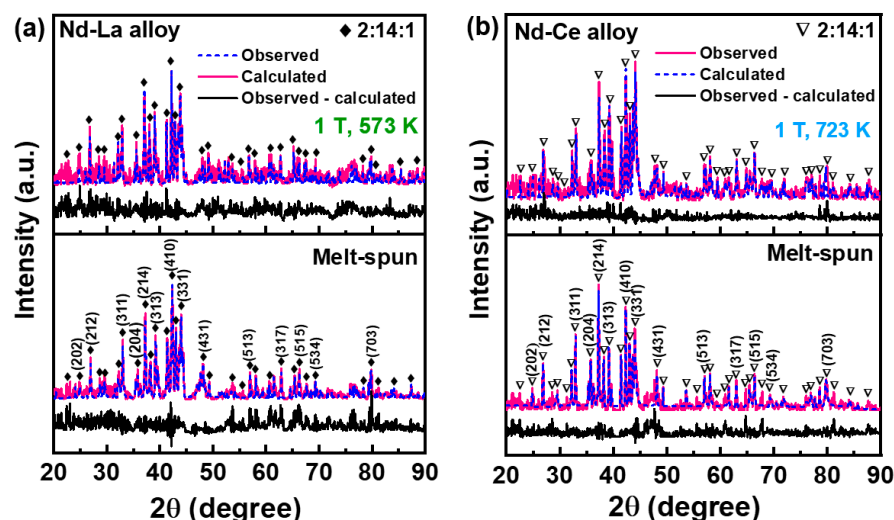


Figure 2. XRD patterns of melt-spun and magnetic field-annealed Nd-Li alloy (a) and Nd-Ce alloy (b).

The bright-field TEM image of the melt-spun Nd-Li alloy in Figure 3a shows the presence of equiaxed grains with an average grain size of approximately 26 nm. The average grain size of the annealed Nd-Li alloy at an optimum temperature (termed as 1 T, 573 K) is increased to 34 nm; see Figure 3b. The plate-shaped grains are observed in the melt-spun Nd-Ce alloy with an average grain size of 47 nm; see Figure 3c. After magnetic field annealing at 723 K, the average grain size is reduced to 36 nm; see Figure 3d. It is in good agreement with values obtained from the XRD result.

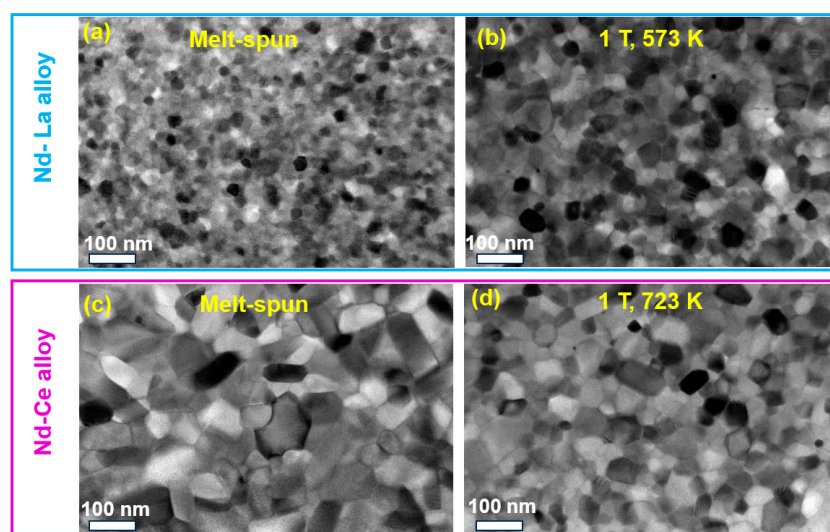


Figure 3. The TEM images of the melt-spun and magnetic field-annealed alloys: (a,b) the Nd-Li alloy and (c,d) the Nd-Ce alloy.

Figure 4 shows the STEM-HAADF images and line scans of the Nd-Li and Nd-Ce alloys. The HAADF image shows atomic number contrast (Z-contrast), indicating that the element with a higher atomic number exhibits a brighter contrast [46]. No bright contrast at the grain boundaries is shown in Figure 4a, and the line scan of L1 in Figure 4b exhibits a uniform distribution of elements. It indicates that no intergranular phase is observed in the melt-spun Nd-Li alloy. Similarly, no intergranular phase is exhibited in the magnetic field-annealed Nd-Li alloy (1 T, 573 K); see the HAADF image in Figure 4c and the line scan of L2 in Figure 4d. Notably, the intergranular phase between adjacent grains is observed in the melt-spun Nd-Ce alloy and marked with an arrow in Figure 4e. The

line scan of the melt-spun alloy including three intergranular phases (termed as IP1, IP2, and IP3) is shown in Figure 4f. The enrichment of Nd is observed at intergranular phases. The elemental composition of intergranular phase is shown in Table 2. The composition of ferromagnetic elements (Fe + Co) of three intergranular phases in the melt-spun Nd-Ce alloy is 81.04 ± 0.95 at.%, 78.70 ± 1.04 at.%, and 81.12 ± 1.01 at.%, which is more than 65 at.% that is reported in reference [17]. It indicates that IP1, IP2, and IP3 are all ferromagnetic intergranular phases. In the magnetic field-annealed Nd-Ce alloy (1 T, 723 K), the intergranular phase is also observed; see the red arrow in Figure 4g. The line scan in Figure 4h and chemistry in Table 2 show that the intergranular phase (IP1#) is ferromagnetic. Moreover, the concentration of (Fe + Co) in the ferromagnetic intergranular phase of the Nd-Ce alloy is increased to 85.81 ± 1.06 at.% after magnetic field annealing.

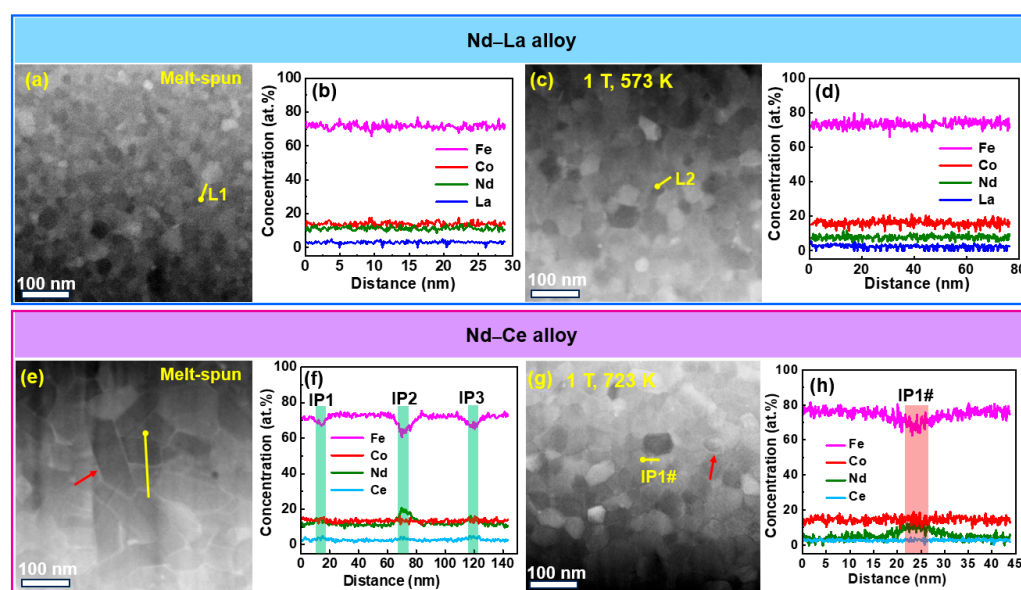


Figure 4. (a,b) The HAADF image and line scan of the melt-spun Nd-La alloy; (c,d) the HAADF image and line scan of the magnetic field-annealed Nd-La alloy at 523 K; (e,f) the HAADF image and line scan of the melt-spun Nd-Ce alloy; and (g,h) the HAADF image and line scan of the magnetic field-annealed Nd-Ce alloy at 723 K.

Table 2. The average elemental composition of intergranular phase in the melt-spun and magnetic field-annealed Nd-Ce alloy.

Condition	Intergranular Phase	Nd (at.%)	Ce (at.%)	Fe (at.%)	Co (at.%)	Fe + Co (at.%)
Melt spun	IP1	14.82 ± 0.79	4.05 ± 0.52	66.64 ± 1.17	14.40 ± 0.74	81.04 ± 0.95
	IP2	17.34 ± 0.69	3.96 ± 0.40	64.08 ± 1.21	14.62 ± 0.88	78.70 ± 1.04
	IP3	14.68 ± 0.85	4.20 ± 0.45	66.98 ± 1.26	14.14 ± 0.76	81.12 ± 1.01
Magnetic field annealing	IP1#	12.10 ± 0.51	2.09 ± 0.47	71.62 ± 1.45	14.19 ± 0.68	85.81 ± 1.06

3.3. The Magnetic Mechanism of Improved Magnetic Property by Magnetic Field Annealing

To better understand the effect of magnetic field annealing treatment, it is essential to investigate the magnetic properties of melt-spun ribbons. We use the rapid solidification of melt spinning to prepare melt-spun Nd-La and Nd-Ce ribbons. Our previous study found that wheel speed, quenching temperature, and chamber pressure during melt spinning played a critical role on the reproducibility of magnetic properties of $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{2.4}\text{Fe}_{12}\text{Co}_2\text{B}$ ribbons [47]. We could achieve ribbons with reproducible magnetic

properties by tuning the quenching temperature in the range of 1573–1623 K, chamber pressure in the range of 0.04–0.07 MPa, and wheel speed in the range of 15–22 m/s. Arreguín-Hernández et al. illustrated the importance of synthesis methods for achieving reproducible properties [48]. Hence, we optimized the quenching temperature (1588 K), wheel speed (15 m/s), and chamber pressure (0.05 MPa) in this study to achieve melt-spun Nd-La and Nd-Ce ribbons with reproducible magnetic properties.

The B_r and $(BH)_{\max}$ of the melt-spun Nd-La alloy are 0.84 T and 117 kJ/m³, which are higher than 0.78 T and 98 kJ/m³ of the melt-spun Nd-Ce alloy. This difference can be attributed to microstructural changes. (1) The partial substitution of Nd by La refines the grain size. The TEM result shows that the average grain size of melt-spun Nd-La alloy is approximately 26 nm, which is smaller than 47 nm of the melt-spun Nd-Ce alloy. Chang et al. found that La could refine the grain size of $(Nd_{1-x}La_x)_{9.5}Fe_{85.5}B_5$ ribbons, which resulted in the increase in B_r by enhancing the exchange coupling between magnetic grains [49]. (2) An intergranular phase is observed in the melt-spun and magnetic field-annealed Nd-Ce alloy but not in the Nd-La alloy. It ascribes to the partitioning position of Nd and Ce in the $Nd_2Fe_{14}B$ unit cell. Liu et al. studied the partitioning of La in the $Nd_2Fe_{14}B$ phase by first-principles density functional calculation and found that 4g and 4f sites in the $Nd_2Fe_{14}B$ unit cell had different substitution energies [50]. When 4g site is occupied by La, the substitution energy is positive, leading to the expulsion of La from the $Nd_2Fe_{14}B$ phase and its segregation at the grain boundaries. When the 4f site is occupied, the substitution energy is negative, indicating that La enters the $Nd_2Fe_{14}B$ phase. Since no intergranular phase is shown in the Nd-La alloy and it is present in the Nd-Ce alloy in this study, it is reasonable to assume that La enters the 4f site and Ce occupies the 4g site in the $Nd_2Fe_{14}B$ phase.

It is worth noting that the underlying magnetic mechanism of improved magnetic properties by magnetic field annealing in the Nd-La and Nd-Ce alloys is different. In the Nd-La alloy, no intergranular phase is observed. The B_r of the Nd-La alloy is increased after magnetic field annealing due to the enhancement of the exchange coupling interaction between adjacent grains. It is confirmed by the magnetic domain structure, as shown in the Fresnel images taken by the Lorentz microscope in Figure 5. The magnetic domain size of the melt-spun alloy (marked by a red dashed line) in Figure 5a is 88 nm, which is larger than the average grain size of 26 nm. It indicates that an interaction domain is formed, which features a magnetic domain containing several grains [51]. The interaction domain is also observed in the magnetic field-annealed alloy, which is marked by a pink dotted line in Figure 5b. Moreover, the domain size of the annealed alloy is 266 nm, which is larger than 88 nm of the melt-spun alloy. It suggests that a stronger exchange coupling interaction is present in the magnetic field-annealed alloy, leading to the improvement in magnetic properties. Hence, the B_r and $(BH)_{\max}$ of the magnetic field-annealed Nd-La alloy are increased to 0.86 T and 124 kJ/m³ when compared to the B_r (0.84 T) and $(BH)_{\max}$ (117 kJ/m³) of the melt-spun alloy.

The effect of magnetic field annealing on the Nd-Ce alloy can be understood through two aspects. (1) The average grain size is reduced by magnetic field annealing. It is interesting to find that the grain size is reduced to 36 nm after magnetic field annealing when compared with 47 nm of the melt-spun alloy. The same phenomenon is also observed in our previous study on rare-earth permanent magnetic alloys [52,53]. It is mainly due to two reasons. One reason is the effect of the length-to-width (b/a) ratio of grains. In the melt-spun Nd-Ce alloy, the plate-shaped grains are present, indicating that the ratio b/a is larger than that of equiaxed grains. The larger the b/a ratio is, easier is the occurrence of grain fragmentation, leading to the refinement of grains. The other reason is the effect of internal stress. Our previous study used convincing evidence from the precession electron

diffraction of TEM to illustrate that the stress concentration was easily present at grain boundaries, and magnetic field annealing treatment could increase the strain, resulting in the reduction in grain size [53]. In this study, the intergranular phase is observed in the melt-spun Nd-Ce alloy, while it is absent in the melt-spun Nd-La alloy, which suggests that more stresses are present in the Nd-Ce alloy. Hence, the grains size of the Nd-Ce alloy is decreased after magnetic field annealing. (2) The chemistry of intergranular phase can be changed by magnetic field annealing. The ferromagnetic intergranular phase is observed in both melt-spun and magnetic field-annealed Nd-Ce alloys. Moreover, the concentration of (Fe + Co) in the ferromagnetic intergranular phase can be increased by magnetic field annealing. It can strengthen the exchange coupling interaction, leading to the improvement in B_r and $(BH)_{max}$ [54]. Hence, the B_r and $(BH)_{max}$ of the magnetic field-annealed Nd-Ce alloy is increased by 5% and 8% when compared with that of the melt-spun alloy.

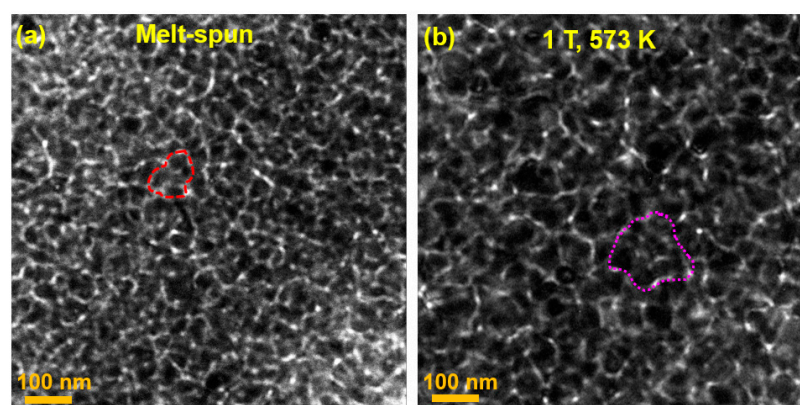


Figure 5. Lorentz microscopy images: (a) melt-spun Nd-La alloy and (b) magnetic field-annealed Nd-La alloy.

The H_{ci} of the Nd-La alloy (906–914 kA/m) in this study is higher than that reported for $(Nd_{1-x}La_x)_{10}Fe_{84}B_6$ ($x = 0.1-0.4$) ribbons ($H_{ci} < 500$ kA/m) [20], which is due to the presence of $La_2Fe_{14}B$ that shows an inferior magnetic property than $Nd_2Fe_{14}B$ in reference [20]. In comparison to the reported B_r (< 0.78 T) and $(BH)_{max}$ (< 100 kJ/m³) of $(Nd_{1-x}La_x)_{13}Fe_{81}B_6$ ($x = 0.1-0.5$) ribbons [21], the studied Nd-La alloy shows higher B_r (0.84–0.86 T) and $(BH)_{max}$ (117–124 kJ/m³). This ascribes to the microstructural refinement of the Nd-La alloy. The $(BH)_{max}$ (106 kJ/m³) of the studied Nd-Ce alloy after magnetic field annealing is higher than 100 kJ/m³ of the reported $(Nd_{0.8}Ce_{0.2})_{2.4}Fe_{12}Co_2B$ ribbons [25], which is due to the presence of the ferromagnetic intergranular phase in the Nd-Ce alloy. The B_r and $(BH)_{max}$ of the Nd-Ce alloy are larger than those of $(Nd_{0.5}Ce_{0.5})_{30.5}Fe_{68.5}B_{1.0}$ [37] and $(Nd_{0.8}Ce_{0.2})_{13}Fe_{82}B_5$ [38] due to the refined microstructure of the Nd-Ce alloy.

Notably, the increment in the B_r and $(BH)_{max}$ of the magnetic field-annealed Nd-La alloy is smaller than that of the Nd-Ce alloy. The B_r and $(BH)_{max}$ of the Nd-La alloy are increased from 0.84 T and 117 kJ/m³ of the melt-spun alloy to 0.86 T, i.e., by 2%, and 124 kJ/m³, i.e., by 6%, of the magnetic field-annealed alloy. The B_r and $(BH)_{max}$ of the melt-spun Nd-Ce alloy are 0.78 T and 98 kJ/m³, and they are increased by 5% to 0.82 T and by 8% to 106 kJ/m³ after magnetic field annealing. These results suggest that annealing in a magnetic field has a stronger effect on the magnetic properties of the Nd-Ce alloy. This may be due to the presence of the ferromagnetic intergranular phase in the Nd-Ce alloy. Our previous study found that the magnetic properties could be improved by tuning the distribution and composition of the intergranular phase [29,54].

4. Conclusions

In this study, we investigate the magnetic properties and microstructures of melt-spun Nd-La and Nd-Ce ribbons. We also study the effects of annealing with a 1 T magnetic field on both their microstructures and magnetic properties. The partial substitution of Nd by La or Ce has a marginal effect on the lattice parameters of the Nd₂Fe₁₄B phase. But La substitution refines the grain size, while Ce substitution results in intergranular phases. These substitutions result in different magnetic properties between partial La- and Ce-substituted (Nd_{0.8}RE_{0.2})_{2.2}Fe₁₂Co₂B (RE = La, Ce) alloys and also affect how the annealing in a magnetic field tailors the magnetic properties of these alloys. Annealing in a 1 T magnetic field improves the magnetic properties of the Nd-La and Nd-Ce alloys. Data analyses suggest that the improvement in the Nd-La alloy comes from the increase in the interaction magnetic domain size, leading to the enhancement of exchange coupling between magnetic grains. However, in the Nd-Ce alloy, annealing in a magnetic field increases the concentration of (Fe + Co) in the ferromagnetic intergranular phase. Our findings provide an approach to develop cost-effective and high-performance rare-earth permanent magnets by magnetic field annealing treatment.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/met15010005/s1>, Figure S1: Hysteresis loops of Nd-La alloy (a) and Nd-Ce alloy (b) after annealing at different temperatures in 1 T magnetic field.

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