



A novel strategy for the fabrication of high-performance nanostructured Ce-Fe-B magnetic materials *via* electron-beam exposure

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ABSTRACT Ce₂Fe₁₄B compound has a great potential to serve as a novel permanent magnet alternative thanks to the abundant and inexpensive rare-earth element (cerium), while its low magnetocrystalline anisotropy and energy product severely restrict its applications. In this work, a novel strategy combining melt-spinning and electron-beam exposure (EBE) aiming for fabricating high-performance Ce-Fe-B magnetic materials is reported to solve the above-mentioned problem. Remarkably, this strategy facilitates developing a suitable grain boundary configuration without using any additional heavy rare-earth element. Under the optimal EBE condition, the maximum energy product $((BH)_{\max})$ of pure Ce-Fe-B alloy is 6.5 MGOe, about four times higher than that obtained after conventional rapid thermal processing method for the same precursor. The enhanced intergranular magnetostatic coupling effect in the EBE sample is validated by mapping the first-order-reversal-curve (FORC) diagrams. The *in-situ* observation of magnetic domain wall motion for Ce-Fe-B alloy using Lorentz transmission electron microscopy reveals that the boundary layers are very effective in pinning the motion of domain walls, leading to the increased coercivity under EBE, and this pinning effect is further verified by micromagnetic simulations. Our results suggest that CeFeB materials using EBE could be a promising candidate after further processing, which could fill the performance “gap” between hexaferrite

and Nd-Fe-B-based magnets.

Keywords: nanoscale magnets, rapid thermal annealing, Ce-Fe-B, magnetic properties, nanocrystalline material

INTRODUCTION

The growing industrial reliance on renewable energy and increasing concern on energy efficiency throughout the complete lifecycle call for clean energy technologies, where rare-earth-based permanent magnets are playing an important role and can serve as wind turbines and the electromobility sector [1]. However, even for the practical rare-earth-based permanent magnets, it is still a great challenge to achieve a high coercivity (H_{cj}) approaching the magneto-crystalline field to meet the requirements for high-performance magnets. The H_{cj} of rare-earth permanent magnets is extremely sensitive to the microstructure in particular the grain size, grain alignment, and grain boundary (GB) configuration: (1) H_{cj} changes with the variation of grain size and keeps at a high level for nanostructured magnets [2–4]; (2) H_{cj} can be enlarged significantly by the alignment of the hard magnetic grains [5–7] and the formation of an adequate GB configuration [8–9]. Usually, nanostructured magnets are obtained through a rapid solidification process, and optimized GB configuration is achieved by conventional furnace an-

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nealing such as composition design, post-annealing, and/or GB diffusion methods [10–14], which all have a low heating rate below 10 K s^{-1} . Inevitably, this low heating rate induces the grain growth and coarsening, resulting in a detrimental effect on permanent magnet performance [15–19]. Here, we report a novel rapid thermal processing (RTP) strategy, electron-beam exposure (EBE) based on a large beam energy, which can lead to extremely high heating rates in low-dimensional systems such as ribbons and films [7,20,21].

Contrary to the conventional furnace annealing strategies, the extremely high heating rate (in the order of 10^3 K s^{-1}) enabled by EBE makes it possible to engineer GB while maintaining a refined microstructure by preventing the undesired grain growth. In our earlier work, we reported that EBE can tailor the microstructure effectively, such as grain sizes, grain shapes, and grain alignments [7,20,21]. This strategy is technically new and has been proved to be extensible to other magnetic materials. The choice of Ce-Fe-B here is based on its competitive permanent magnetic properties and the wide availability of cerium. Cerium is one of the most abundant rare-earth elements obtained as a byproduct from neodymium refining. A successful Ce-Fe-B magnet may alleviate the resource shortage problem of rare-earths [22,23].

The EBE heating rate achieved in this study is about 6000 K s^{-1} , as measured on the ribbon surface by an infrared thermometer [7]. The *in-situ* observation of magnetic domain wall motion under an external magnetic field indicates a strong pinning model of coercive mechanism under such an improved microstructure. This result, combined with mapping the first-order-reversal-curve (FORC) diagrams, shows that the origin of coercivity development is through magnetostatic coupling enhancement. Consequently, the maximum energy product obtained by EBE is higher than that obtained by RTP. It is expected that this new fabrication technology may help achieve the desired microstructure of rare-earth permanent magnetic materials and hence enable efficient and balanced utilization of cerium resources.

EXPERIMENTAL SECTION

Material fabrications

Alloy ingots with nominal compositions of $\text{Ce}_{7.2}\text{Fe}_{86.6}\text{B}_{6.2}$, $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$, and $\text{Ce}_{17.2}\text{Fe}_{76.6}\text{B}_{6.2}$ were prepared by induction melting of 99.9% pure primary materials in a high-purity argon atmosphere. More details about the EBE experimental configuration and the set-up can be

found in our earlier study [7]. The ribbon thickness was approximately $20\text{ }\mu\text{m}$, and the length and width were tailored to 1 cm . The corresponding mass was estimated to be $0.153 \times 10^{-3}\text{ g}$, assuming the density was 7.67 g cm^{-3} . The temperature ramping time and annealing time in this work were set at 0.1 and 1 s , while the accelerating voltage and focus current were set at 10 kV and 165 mA , respectively. For comparison, a conventional RTP was carried out by placing the as-spun ribbons onto a silicon wafer in the chamber equipped with a uniform lamp array that can be accurately controlled and monitored. To obtain a proper nanocrystalline structure, the amorphous ribbons were annealed with various durations in the range of 673 – 1073 K . The heating rates of RTP and EBE were as high as 30 – 70 and 3000 – 6000 K s^{-1} , respectively.

Material characterizations

The annealed ribbons were characterized by X-ray powder diffraction (XRD, Cu-K α radiation, $\lambda = 1.54187\text{ \AA}$) on a single-crystal silicon substrate. Transmission electron microscopy (TEM, JEM-2100F, Talos F200X) with energy dispersive spectroscopy (EDS) was used to examine the microstructure and phase composition for the melt-spun ribbons. Talos F200X was used for *in-situ* observation of the magnetic domain wall motion with a maximum applied magnetic field of 2.2 T . The high-resolution TEM (HRTEM) samples were prepared by the ion milling technique (Gatan691/695), and the Lorentz TEM (LTEM) samples were prepared by the focused ion beam (FIB, Auriga-Carl Zeiss). Magnetic measurements were performed with a physical property measurement system (PPMS-9) and vibrating sample magnetometer (VSM, Lakeshore8600) at room temperature (RT). The maximum energy product $(BH)_{\text{max}}$ at RT was calculated using the density of 7.67 g cm^{-3} and a demagnetization correction ($N \approx 0.977$).

Micromagnetic simulations

Magnetization reversal behaviors in the nanostructured Ce-Fe-B magnetic materials were simulated by a finite element software based on the Landau-Lifshitz-Gilbert-Langevin equations. The models were constructed into a geometry consisting of cubic-shaped grains with sizes of 40 – 100 nm . For the EBE route, the width of the GB was set as 4 nm . Moreover, for RTP, according to our experimental observations, it was considered that grains touch each other without GB. However, abundant small particles with the size of 8 nm distributed randomly on the interface between grains. The magnetic anisotropy

constant (K_1), saturation magnetization ($\mu_0 M_s$), and exchange stiffness (A) of $\text{Ce}_2\text{Fe}_{14}\text{B}$ phase for all models were 1.5 MJ m^{-3} , 1.0 T , and 5 pJ m^{-1} , respectively [24].

RESULTS AND DISCUSSION

Comparison of the crystallization process, magnetic properties and microstructure between RTP and EBE

Different heating rates were achieved by RTP and EBE. During RTP, the crystallization process of the amorphous state is regulated by the power of the lamp array with a heating rate up to $30\text{--}70 \text{ K s}^{-1}$. In the case of EBE, the

heating rate and heating energy determining the crystallization process are regulated by varying the accelerating voltage, beam current, and irradiation time. The phase evolution with the heating rate is illustrated in Fig. 1. Firstly, melt spinning is used to obtain an amorphous precursor with a uniform atomic distribution of each element. EBE with a rapid heating rate is used in the next step to irradiate the amorphous precursor with a controllable injection energy. The infiltration depth and exposure area radius of the electrons are several micrometers and millimeters in size, respectively, depending on the choice of accelerating voltage and focus

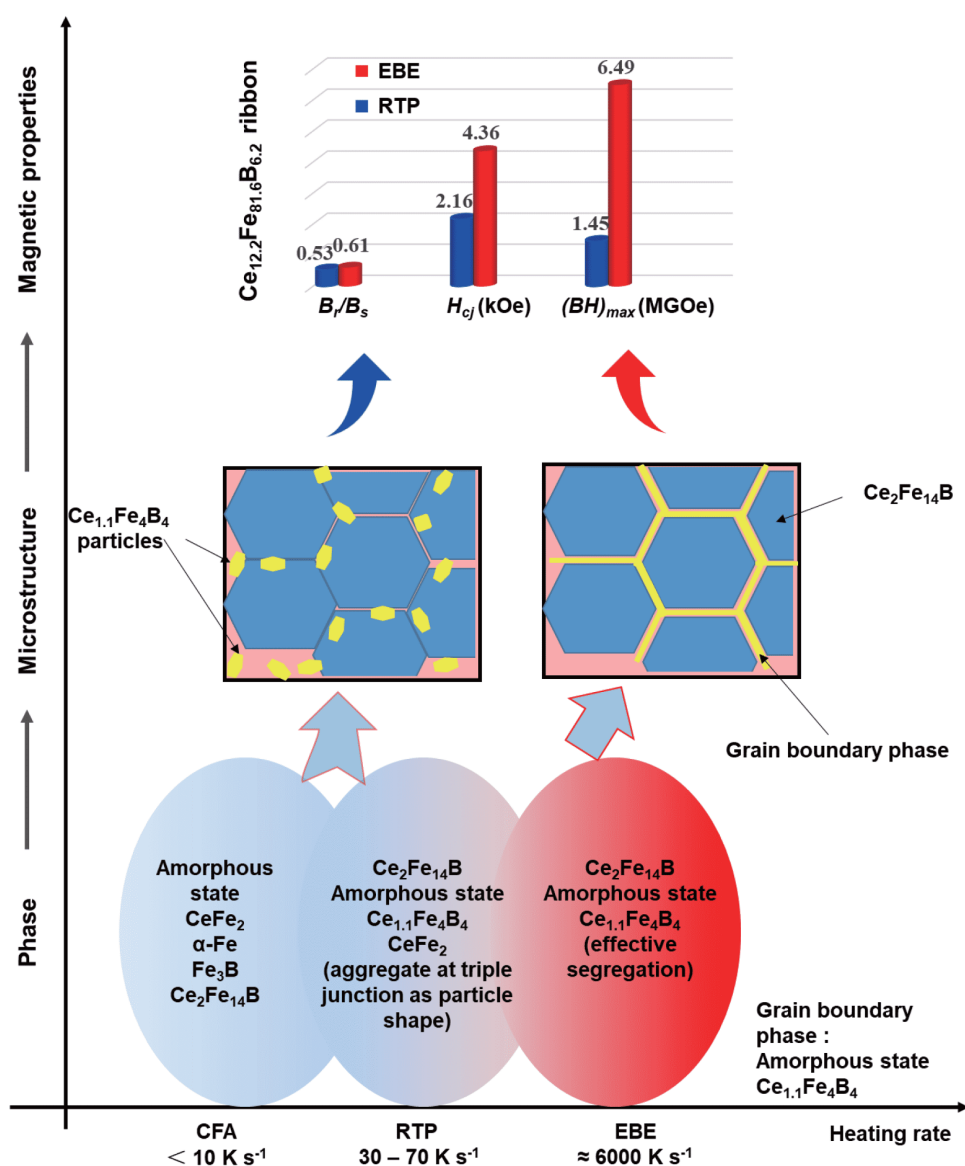


Figure 1 Schematic of the phase evolution with the heating rate.

current. These ranges of infiltration depth and exposure area are adequate to produce the heat required for inducing the crystallization from top to bottom of the ribbons (thickness of 15–30 μm and width of 5–10 mm). This can be verified by the XRD results shown in Figs S1 and S2. The XRD patterns of both the wheel side and free side show the typical 2:14:1 tetragonal structure. However, for $\text{Ce}_{17.2}\text{Fe}_{76.6}\text{B}_{6.2}$ composition, no preferential peaks were observed on the wheel side or free side, which is different from the Pr-Fe-B system reported before. In Pr-Fe-B with Pr-rich composition, instead of forming an undesired Pr-Fe phase, a quasi-texture nanostructure was induced by EBE due to the high wettability of Pr-rich phase when there is an excess of Pr [7]. When Ce is excessive in Ce-Fe-B, a small amount of undesired CeFe_2 phase is observed. This phase may facilitate the long sintering process but inhibit the alignment of the Ce-Fe-B grain in the short annealing period due to its high thermal stability and preferential growth [15,18]. For $\text{Ce}_{7.2}\text{Fe}_{86.6}\text{B}_{6.2}$ and $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$, similar results to Pr-Fe-B were obtained, an extra α -iron phase appeared due to the excess of iron, and a single 2:14:1 phase was observed due to the appropriate Ce/Fe ratio. Compared with the highest heating rate of RTP (70 K s^{-1}), the heating rate achieved by EBE in our experiments is in the range of 3000–6000 K s^{-1} . This extremely fast heating rate greatly improves the efficiency of crystallization [25,26]. The amorphous Ce-Fe-B alloy has been crystallized within 1 s under EBE. To the best of our knowledge, this is by far the most effective technique to complete the crystallization process of Ce-based permanent magnetic materials.

The strength of a permanent magnet is characterized by H_{cJ} and $(BH)_{\text{max}}$. The theoretical upper limit values of the H_{cJ} and $(BH)_{\text{max}}$ of $\text{Ce}_2\text{Fe}_{14}\text{B}$ are $H_{\text{a}} = 26 \text{ kOe}$ and $\frac{1}{4}M_{\text{s}}^2 = 34 \text{ MGOe}$ [27–29], respectively. Fig. 2 shows the second quadrant of the hysteresis loop for the three different Ce-Fe-B compositions under EBE and optimal RTP conditions (Table 1). These values have been chosen on the basis of a systematic study resulting in the evolutions shown in Fig. S8. For reference, beam currents of 0.4–0.8 mA approximately correspond to temperatures of 700–840 K on the sample surface.

As a first observation from Fig. 2, the EBE-treated sample always provides a higher H_{cJ} than the RTP-treated sample. Especially for the $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ composition, both the coercivity and remanence are enhanced significantly. The EBE-treated $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ composition reaches values as high as $H_{\text{cJ}} = 4.4 \text{ kOe}$ and $(BH)_{\text{max}} =$

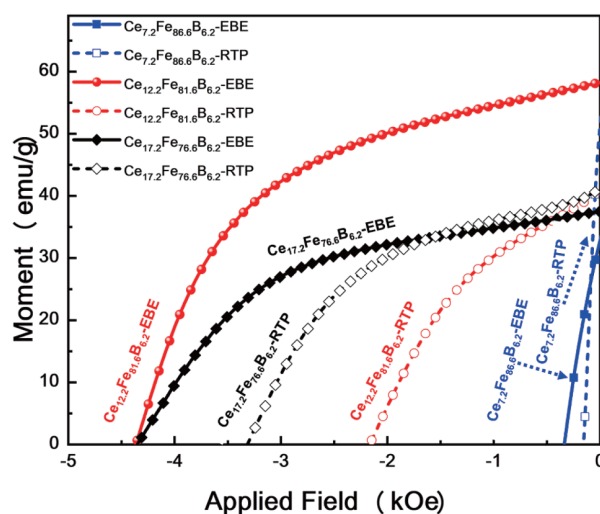


Figure 2 Demagnetization curves of the $\text{Ce}_{7.2}\text{Fe}_{86.6}\text{B}_{6.2}$, $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$, and $\text{Ce}_{17.2}\text{Fe}_{76.6}\text{B}_{6.2}$ ribbons under EBE (full symbols and solid line) and RTP (empty symbols and dashed line).

Table 1 Experimental conditions used by EBE and RTP to achieve and optimized combination of coercivity and remanence in the CeFeB alloys under study

Composition	EBE conditions		RTP conditions	
	Beam current (mA)	Irradiation time (s)	Temperature (K)	Annealing time (s)
$\text{Ce}_{7.2}\text{Fe}_{86.6}\text{B}_{6.2}$	0.8	1	1023	300
$\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$	0.4	1	823	300
$\text{Ce}_{17.2}\text{Fe}_{76.6}\text{B}_{6.2}$	0.6	1	823	300

6.5 MGOe. These results indicate that magnetic properties of Ce-Fe-B alloy not only depend on the specific composition but are strongly influenced by the microstructure, which are in turn affected by the heating rate. By comparing the experimental magnetic properties with the theoretical upper limit values of $\text{Ce}_2\text{Fe}_{14}\text{B}$, plenty of room for improvement of the magnetic properties of Ce-based magnets remains open. The EBE provides a new promising approach to further improving the magnetic performance. The considerable increase in magnetic properties by EBE treatment can be explained with further understanding of the microstructure and magnetic domain configuration. A comparison with the other two samples is shown in the Supplementary information. Results obtained in the other two samples also demonstrate the amelioration effect of EBE for Ce-based materials. It must be admitted that although EBE could be scalable for manufacturing materials such as powders, ribbons, or films, it is not well applicable for the bulk magnets. The challenge for the bulk magnets is the cost-

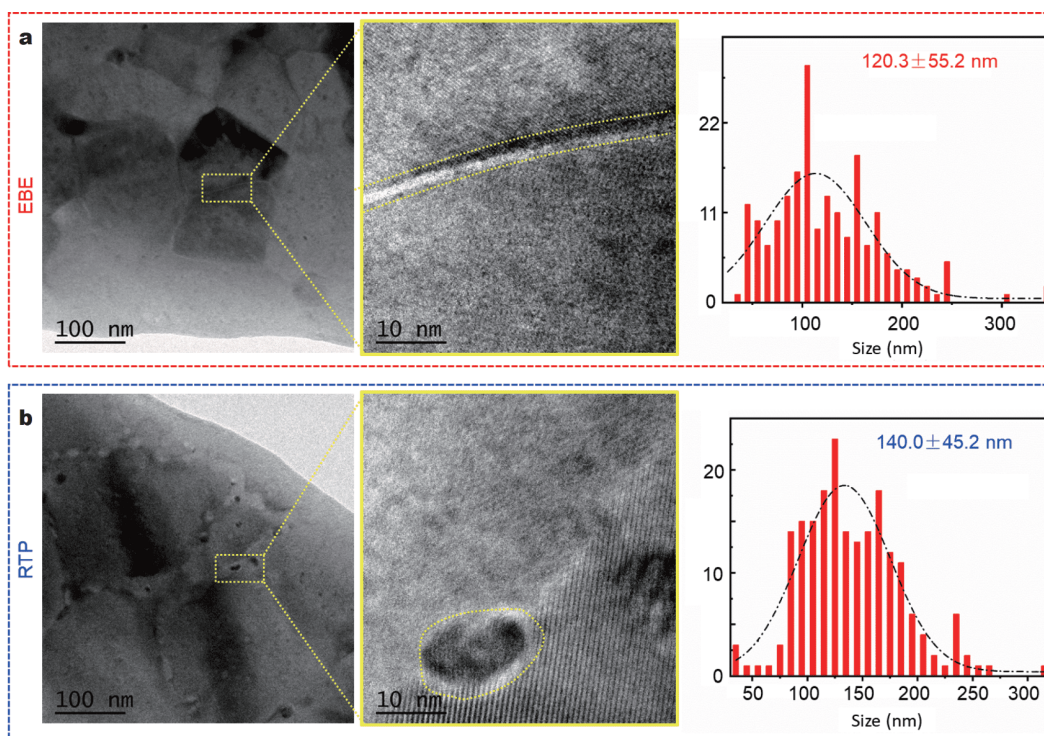


Figure 3 TEM images and the corresponding grain size distributions of $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ under (a) EBE and (b) RTP.

effective issue, which may be solved by exploring other promising methods for the time being, such as spark plasma and explosive compaction.

Extrinsic magnetic properties are intimately related to the microstructure for permanent magnets. To further understand the relation between the heating rate and the microstructure, we employed the TEM and LTEM techniques to characterize the $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ composition in detail. As shown in Figs 3 and 4, different microstructures were obtained under RTP and EBE. RTP leads to large-sized hard magnetic grains with few GBs. The grain sizes determined from TEM pictures are 140.0 ± 45.2 nm for RTP and 120.3 ± 55.2 nm for EBE, respectively. A large number of small particles are dispersed randomly along with the GB of the hard-magnetic phase Ce-Fe-B in the sample. Unlike the results obtained on Ce-Fe-B ribbons annealed by conventional furnace [15,16], these small particles (about 8 nm in size) have been identified as $\text{Ce}_{1.1}\text{Fe}_4\text{B}_4$ by high-angle annular dark-field imaging (HAADF) (Fig. 4b) and HRTEM (Fig. 5b) combined with EDS analysis (Fig. S3). $\text{Ce}_{1.1}\text{Fe}_4\text{B}_4$ particles are paramagnetic at RT, which is deleterious to the magnetic properties of the Ce-Fe-B ribbon. The coercivity is sensitive to the grain orientation. If a grain alignment along (00 l) is formed, the coercivity would be enlarged sig-

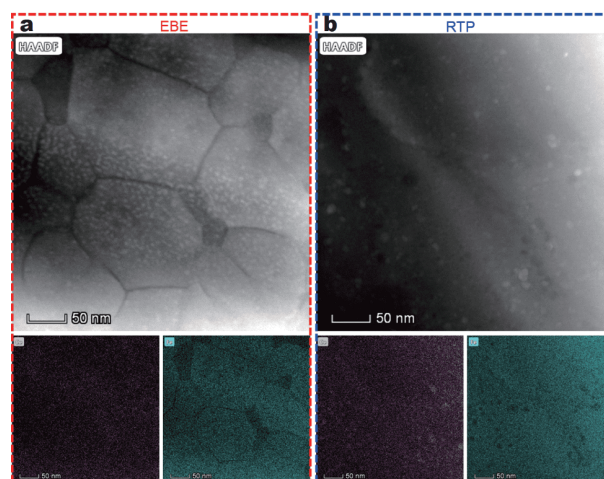


Figure 4 HAADF images of $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ under (a) EBE and (b) RTP, and the corresponding EDS mappings for Ce and Fe in $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$.

nificantly, as reported in our earlier work about Pr-Fe-B system [7]. We calculate the zone axes of the observed grains as shown in Fig. 5. The zone axes are [276], [201] and [101] for grains #1, #2 and #4, respectively. No obvious texture is observed; hence the contribution of grain orientation to coercivity in Ce-Fe-B system may not be the major reason of coercivity enhancement in the EBE-

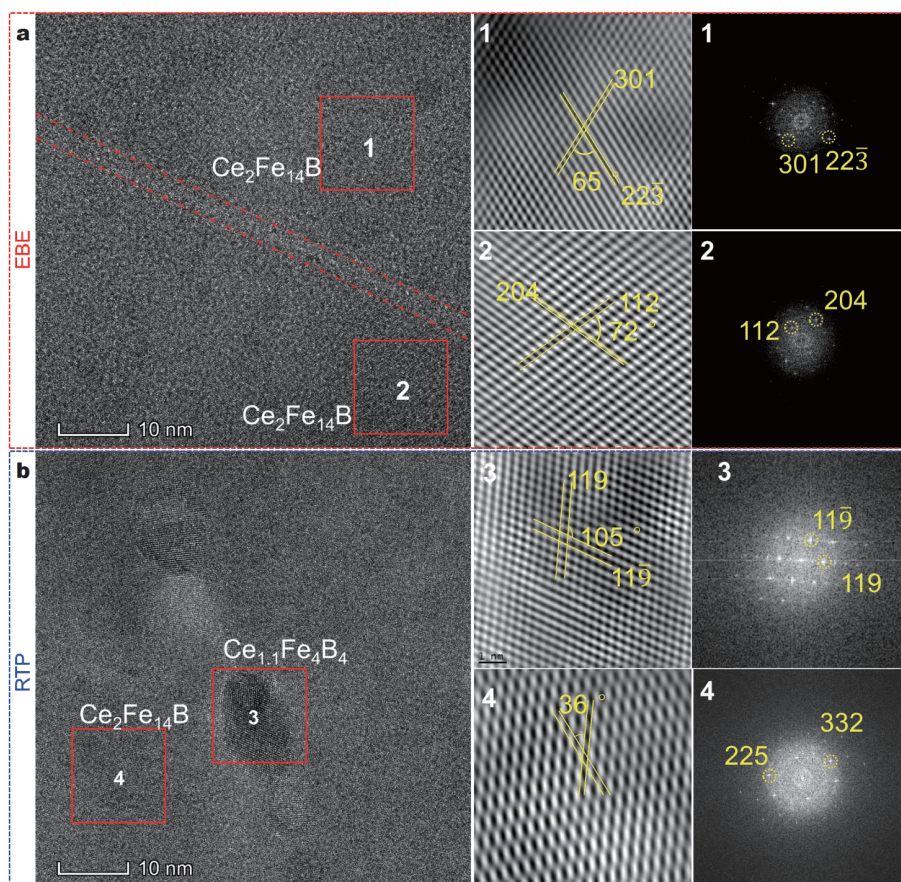


Figure 5 HRTEM images of $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ under (a) EBE and (b) RTP. #1–#4 are the corresponding high-magnification images and fast Fourier transform patterns for the selected regions (marked with red squares on the left-hand side images) in (a) and (b).

treated sample. The coercivity is also strongly correlated with the configurations of the RE-rich (RE: rare earth) GB phase, which directly determines the magnetic interaction between neighboring grains [8,30–34]. By comparison with results on the RTP-treated samples, continuous uniform GBs (Fe-lean as shown in Fig. 4a) similar to that observed in high-performance sintered or hot deformed Nd-Fe-B magnets are formed under EBE, with a thickness reaching 2.5 nm (see Figs 3 and 5). This might be one of the main reasons for the improved magnetic response obtained by EBE. The observed GB configuration most likely inhibits the propagation of reversed magnetic domains across grains by improving the intergranular magnetostatic coupling and therefore facilitating the coercivity enhancement in the Ce-Fe-B system [35]. The EBE technique, providing extremely high heating and cooling rates caused by the impingement and collision of thousands of electrons in a fairly short time, is beneficial for achieving the observed GB configuration. Commonly to both techniques, RTP and EBE, the heat energy ge-

nerated during the process promotes the crystallization from the amorphous state constituting the melt-spun precursor. The significant difference in the heating rate of both techniques is responsible for the difference observed in the crystallite size (coarser with decreasing heating rate). What is more significant, however, is the effect of the heating rate on the induced mobility of the atoms during crystallization and, accordingly, on the resulting GB configuration. When the heating rate is low, as occurring by RTP, a long annealing time is required to complete the crystallization process (300 s). This indicates under RTP condition, Ce travels farther than it does under the EBE condition. Hence the Ce atoms are more likely to form CeFe_2 nanocrystal particles with high thermal stability (see Figs 3, 4b, and Fig. S3), enriched at the GBs, thus preventing the formation of a continuous GB phase [36]. When the heating rate increases, according to Fick's law of diffusion [37,38], the diffusion coefficients of the atom will be strengthened, resulting from the ultra-fast energy injection and the quasi-stress

induced by the electron beam irradiation [7]. Thus the crystallization process is significantly accelerated. A delicate balance may be achieved between the competition of crystallization and Ce diffusion under EBE condition, causing the heavier Ce atoms to remain at the GB and form a continuous and uniform GB. This explanation fits well the enhanced coercivity observed in the EBE-processed sample.

Interactions among the magnetic particles and *in-situ* observation of magnetic domain wall motion under EBE and RTP

To further clarify the interactions among the grains with different microstructures (especially different GB configurations) and their correlations with the magnetic properties, FORC was employed to study the coercive mechanism. Talos F200X with a maximum applied field of 2.2 T was used for *in-situ* observation of the dynamic magnetic domain motion during the magnetization and demagnetization processes of the EBE- and RTP-treated ribbons.

An FORC measurement begins with saturating the samples by a sufficiently large positive applied magnetic field, after which the sample is brought to a reversal field, H_a , and then the magnetization is measured as the applied field H increases from H_a back up to saturation. The magnetization at applied field H_b on the FORC with reversal point H_a is denoted by $M(H_a, H_b)$, where $H_b \geq H_a$. The measurement process is repeated for different reversal fields to trace out the FORCs. An FORC distribution is defined as the mixed second derivative:

$$\rho(H_a, H_b) \equiv -\frac{\partial^2 M(H_a, H_b)}{\partial H_a \partial H_b}. \quad (1)$$

The FORC diagram shown here is plotted by the FORCinel program designed by Harrison *et al.* [39], and the smoothing factor is chosen as 3 consistently to make a clear comparison between FORC diagrams while keeping noise to an acceptable level.

When an FORC distribution is plotted by FORCinel, the coordinates are changed from $\{H_a, H_b\}$ to $\{H_u \equiv (H_b - H_a)/2, H_c \equiv (H_b + H_a)/2\}$ to visualize the interactions. After the coordinate transformation, an FORC diagram is a contour of an FORC distribution with H_c and H_u on the horizontal and vertical axes, respectively [40]. According to Pike *et al.* [41], H_c is equivalent to the particle coercivity and H_u is equivalent to the local interaction field. As shown in the FORC diagrams (Fig. 6), the vertical spread along the H_u axis is wider in the EBE-treated sample compared with that of the RTP-treated one. This indicates that the magnetostatic coupling is stronger in the EBE-treated ribbon [40,41]. As it is well-known, the GB phase plays an important role in the interactions between the grains [8,9,11,42]. In the EBE-treated ribbon, the GB is well defined, uniform, and continuous as illustrated in Figs 3–5. Hence, the stronger magnetostatic coupling should result from this engineered GB configuration. The role of GB in the magnetostatic coupling was further verified from the dynamic structural changes of the magnetic domain walls observed using *in-situ* LTEM. Fig. S4 shows that most of the magnetic domains in the EBE-treated ribbon are single domains and the domain walls coincide with the GBs. During the initial magneti-

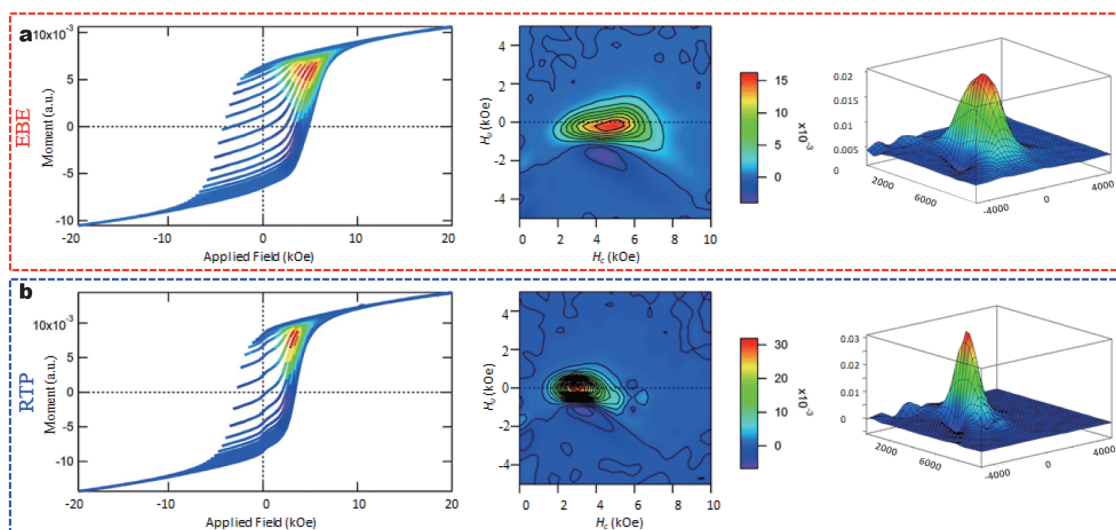


Figure 6 2D and 3D FORC diagrams of the $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ ribbon under (a) RTP, and (b) EBE. The left columns are the colored FORC curves with the FORC distributions plotted inside the hysteresis loops.

zation process, two typical magnetic domain wall motion models were observed. With increasing magnetic field, the domain wall moves quickly through the grain interior and stays intermittently at the GBs, which directly evidences the pinning effect of the GB, as shown in the Supplementary Video. On the contrary, for the RTP-treated sample, the increase of the magnetic field leads to shrinkage or expansion of the magnetic domain evolving continually from the corner of the domain or the smaller particles. This kind of wall motion is consistent with the nucleation model. This different behavior explains the coercivity enhancement observed in the EBE-treated ribbon. The peak corresponding to the switching field centered at around 4 kOe along the H_c axis for the EBE-treated ribbon is about 2 kOe higher than that determined for the RTP-treated ribbon (Fig. 6), which again is consistent with the coercivity trend. During the demagnetization process *in-situ* observed by LTEM, new domain walls appear more slowly (i.e., need to increase further the external field) in the EBE-treated sample compared with that of the RTP-treated one (see the I, II, and III circles marked in Fig. 7). The image contrast variation for LTEM picture is the reflection of the domain wall motion. In the EBE-treated ribbon, as the magnetic field decreases to 503 mT starting from the saturation magnetization state, a massive image contrast variation occurs. By comparison, this situation occurs at a higher magnetic field of 820.8 mT in the RTP-treated ribbon

(more specific magnetic domain wall motion is shown in Figs S5 and S6). This result demonstrates that the activation of the magnetic domain wall motion requires more energy in the EBE-treated sample, in good agreement with the higher coercivity measured for this sample.

This study employs the FORC diagrams combined with the *in-situ* LTEM observation to study the relationship among the microstructure, magnetic grain interactions, and permanent magnet properties in the Ce-Fe-B ternary alloy system. These results reveal that the EBE technique can produce a continuous uniform GB phase in the Ce-Fe-B ternary system, which can isolate magnetic grains effectively, resulting in proper strengthening of the magnetostatic coupling. The obtained results suggest that the GB phase might act as an efficient pinning site of magnetic domains during magnetization reversal, thus increasing the coercivity of the nanostructured Ce-Fe-B ribbon.

Micromagnetic simulations

To provide a more comprehensive insight into the correlation experimentally found between microstructure and magnetic properties, micromagnetic simulations were carried out. The microstructure of the sample prepared under EBE was modeled, as shown in Fig. 8a, which is composed of 125 cubic-shaped grains (40–100 nm) separated by a 4-nm GB (Fe-lean phase) layer. On the other hand, the microstructure of the sample prepared by

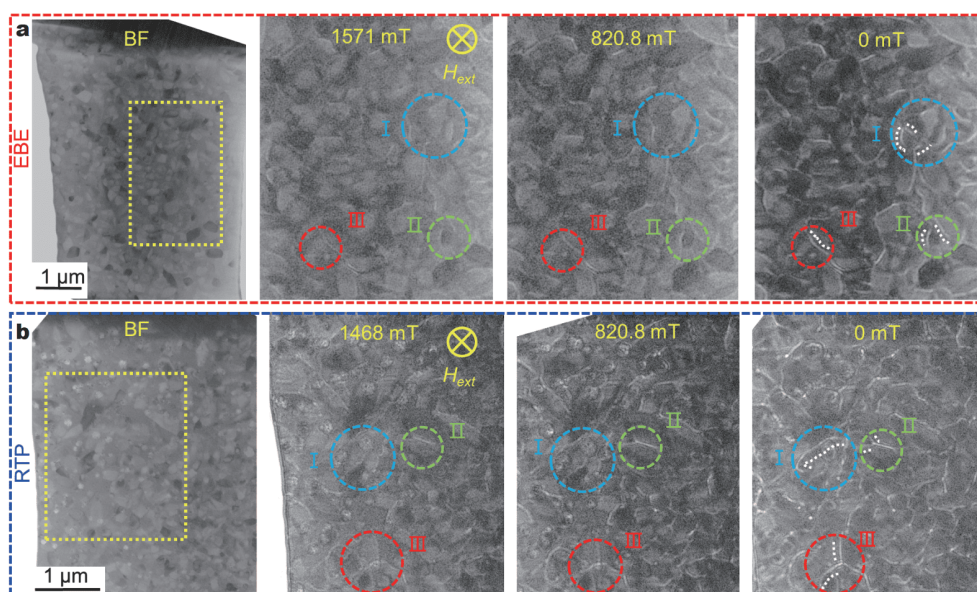


Figure 7 *In-situ* observation of demagnetization process of $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ ribbon under (a) EBE, and (b) RTP. The white dashed lines in the figures of 0 mT denote the typical magnetic domain wall (more in-detail magnetic domain wall motions are shown in Figs S5 and S6, and the dynamic domain wall motion is shown in Supplementary Video).

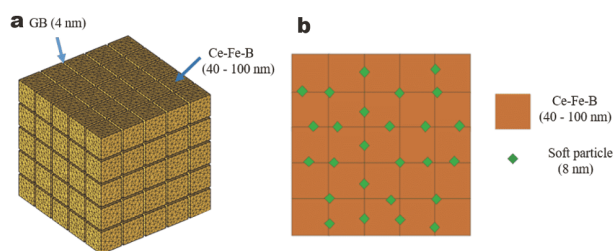


Figure 8 (a) The model for EBE-treated ribbons with cubic-shaped Ce-Fe-B grains (40–100 nm) separated by GB layer (4 nm). (b) The XY section of the bulk model for RTP-treated ribbons with Ce-Fe-B grains (40–100 nm) and soft particles (8 nm).

Table 2 Magnetic parameters of Ce-Fe-B, GB, and soft particles

Materials	K_1 (J m^{-3})	A (J m^{-1})	J_s^a (T)
Ce-Fe-B	1.5	5.0	1.17
GB	0	6.0	1.0
Soft particles	0	1.0	1.0

a) J_s is saturation magnetic polarization.

RTP has been simulated by grains (40–100 nm) in close contact with each other with the presence of soft particles with a size of ~ 8 nm. These soft particles were distributed randomly on the interfaces between Ce-Fe-B grains, as shown in Fig. 8b. The volume ratio that these particles occupy in the system is maintained at 1%. The effective magnetocrystalline anisotropy constant of the soft particles has been assumed to be a negligible value since their magnetization can randomly flip direction under the influence of temperature. The specific magnetic parameters used in micromagnetic modeling are listed in Table 2.

Fig. S7 shows simulation results on magnetization re-

versal behaviors when the sizes of Ce-Fe-B grain and soft particles are 100 and 8 nm, respectively, and the dependence of the coercivity on the grain size in the simulated Ce-Fe-B system. The result of micromagnetic simulation shows that the coercivity of a Ce-Fe-B magnet with an engineered GB configuration (EBE-type) is about twice higher than that with numerous soft particles and without uniform GB phase (RTP-type). This result supports our previous arguments explaining the differences in the coercivity values obtained using different heating techniques.

The configurations of magnetization reversal simulated for both microstructures (EBE and RTP) are shown in Fig. 9; the size of Ce-Fe-B grain is 100 nm, the thickness of GB phase is 4 nm, and the size of soft particles is 8 nm. In the model of Ce-Fe-B bulk with the engineered GB configuration (EBE-type), nucleation begins at the GB on the surface of the bulk due to the relatively low magnetocrystalline anisotropy, as illustrated in Fig. 9a; $\mu_0 H_n$ (nucleation field) is -0.7 T and $\mu_0 H_c$ (coercive field) is -0.8 T. In the model of bulk Ce-Fe-B with the inclusion of soft particles (RTP-type), nucleation begins at the soft particles with the lower applied field (Fig. 9b), due to their negligible magnetocrystalline anisotropy and low exchange stiffness; $\mu_0 H_n$ is -0.41 T and $\mu_0 H_c$ is -0.49 T.

CONCLUSIONS

This study proposes a new route to developing a high-performance ternary Ce-Fe-B permanent magnet material through a novel strategy that combines melt-spinning for producing amorphous ribbons followed by application of EBE for crystallization of the 2:14:1 structure. The combination of both techniques and, particularly important,

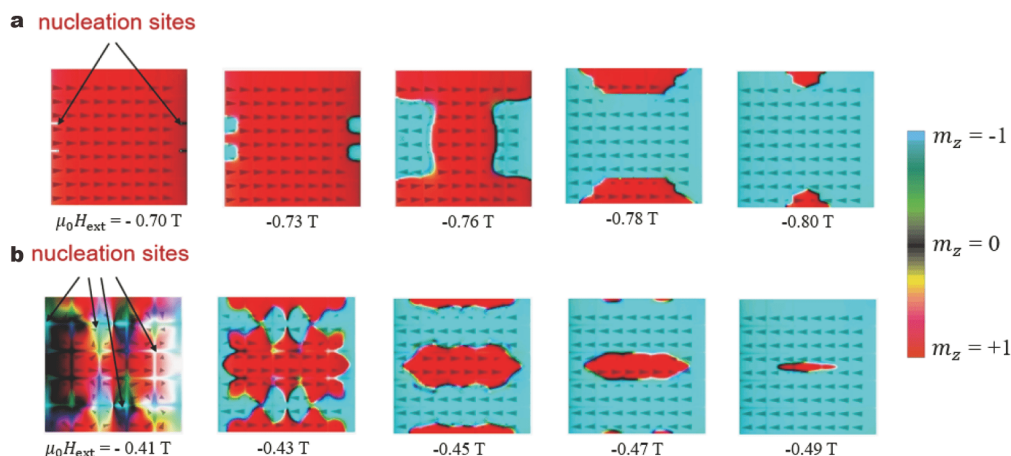


Figure 9 The variation of magnetization and nucleation sites in the models of Ce-Fe-B bulk with (a) an engineered GB configuration (EBE-type), and (b) soft particles (RTP-type) for the Ce-Fe-B grain size of 100 nm, the GB thickness of 4 nm, and the soft particle size of 8 nm.

the use of a high heating rate (about 6000 K s^{-1}) in EBE, allow for a controlled grain growth, a careful design of the GB, and an enhanced intergranular magnetostatic coupling, tunable by controlling the irradiating conditions. Following this route, an optimized composition $\text{Ce}_{12.2}\text{Fe}_{81.6}\text{B}_{6.2}$ shows a coercivity as high as 4.4 kOe and a maximum energy product of 6.5 MGOe. Microstructural and magnetic characterizations, in combination with micromagnetic simulations have proven that the GB is effective in pinning the motion of domain walls, leading to the increased coercivity, which is well above that obtained by other methods using lower heating rates. The conclusions extracted from this study open a new path towards the development of a promising candidate to fill the performance “gap” between hexaferrite and Nd-Fe-B-based magnets.

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Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Experimental details and supporting data are available in the online version of the paper.



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一种通过电子束曝光制备高性能纳米结构Ce-Fe-B磁性材料的新方法

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摘要 Ce₂Fe₁₄B作为一种基于广泛而廉价的稀土(铈)的新型永磁体, 低矫顽力和低最大磁能积严重限制了其在永磁领域中的应用。在这项工作中, 我们报道了一种将熔融纺丝技术和电子束曝光(EBE)技术结合的新方法, 旨在制造高性能Ce-Fe-B磁体。值得注意的是, 该方法可以在不使用任何其他重稀土元素的情况下调控出合适的晶界构型。在最佳EBE条件下, 纯Ce-Fe-B合金的最大磁能积为6.5 MGOe, 比常规快速热处理方法制备的合金的最大磁能积高四倍左右。这说明, 使用EBE制备的CeFeB材料作为前驱体, 经过进一步加工之后, 如热变形或烧结, 有望填补六角铁氧体与Nd-Fe-B磁体之间的市场空白。