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Effect of Zr doping on the magnetic properties and FORC diagrams of (Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_{2}B alloys

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The microstructure and room temperature magnetic properties for (Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_{2}BZr_{x} (x = 0-5) nanocomposite alloys prepared by melt-spinning method and subsequent thermal annealing have been investigated. Zr addition has proved to result in relevant improvements in the microstructure and magnetic properties. The enhanced coercivity $H_{cj}$ of 6.02 kOe and maximum energy products ($BH_{max}$) of 15.2 MGOe have been obtained at the optimum temperature $T_a$ with the Zr content at x = 2 and x = 3, respectively. The results of X-ray diffraction (XRD) and transmission electron microscope (TEM) indicate that a small amount of Zr addition can refine the grain size and cause a uniform distribution of soft magnetic $\alpha$-Fe phase. A first-order-reversal curve (FORC) analysis is performed for the Zr-free (x = 0) sample and the Zr-doped (x = 3) sample. The FORC diagrams suggest the Zr-doped sample with a higher maximum $\rho$ of the major peak and the flat surface is responsible for the better magnetic performance. The results of Kronmüller plot show that nucleation model is the dominating mechanism for the magnetization reversal in the Zr-free and Zr-doped alloys and give the evidence for the increase in microstructural parameter $\alpha_{ex}$ resulting in a drastic increase of coercivity in the Zr-doped alloy. © 2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/1.5041996

I. INTRODUCTION

Nd_{2}Fe_{14}B permanent magnets are the most widely used type of rare earth permanent magnets due to their outstanding combination of magnetic properties.¹–³ However, the use of expensive rare-earth element Nd increases the cost of the Nd_{2}Fe_{14}B-type permanent magnets, which is one of the critical problem that hinder the development of the Nd-Fe-B magnets.⁴ Recently, the scientific researchers pay a great interest in low cost permanent magnets by reducing the total rare-earth content or substituting Nd element by La or Ce.⁵,⁶ However, the magnetic properties of low cost Ce or/and La substituted NdFeB magnets are not impressive due to low intrinsic magnetic properties of the (Ce/La)_{2}Fe_{14}B. In 2015, Pathak et al.⁷ obtained a favorable magnetic performance ($H_{cj}$ = 7.8 kOe, ($BH_{max}$ = 16 MGOe) in (Nd_{0.8}Ce_{0.2})_{2}Fe_{12}Co_{2}B nanocrystalline alloy by simultaneous substitution of Nd by Ce, and Fe by Co. Unfortunately, the remanence ($B_r$) is not impressive after the replacement of Nd by Ce element. It is well established that the enhanced $B_r$ can be obtained by the remanence enhancement between the hard and soft magnetic phase in exchange-coupled nanocomposite alloys.⁸,⁹ Therefore, as reported in Ref. 10 in our previous study for the (Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_{2}B nanocomposite magnets, we obtained the enhanced remanence ($B_r$ = 10.1 kG) for the low rare-earth (Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_{2}B sample. However, the intrinsic coercivity ($H_{cj}$ = 4.9 kOe) and maximum energy product ($BH_{max}$ = 13.7 MGOe) of the (Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_{2}B nanocomposite alloys

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Footnotes:

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are inferior to the (Nd<sub>0.8</sub>Ce<sub>0.2</sub>)<sub>2</sub>Fe<sub>12</sub>Co<sub>2</sub>B nanocrystalline alloys. Hence, enhancing the H<sub>c</sub> and (BH)<sub>max</sub> of the (Nd<sub>0.8</sub>Ce<sub>0.2</sub>)<sub>1.6</sub>Fe<sub>12</sub>Co<sub>2</sub>B nanocomposite alloy with low rare-earth is very important for achieving high performance in Nd-Ce-Fe-Co-B magnets.

As a feasible method for enhancing the coercivity and maximum energy product, doping Zr element has been frequently employed to refine the microstructure and modifying the phase formation process. Liu et al. suggested that a small amount of Zr can improve the coercivity, thermal stability and exchange coupling of nanocomposite Nd-Dy-Y-Fe-B alloys. Ni et al. found that a certain amount Zr content could refine the microstructure and enhance the exchange coupling interaction in Ce-Fe-B alloys. In this paper, the effects of Zr doping on the magnetic properties of (Nd<sub>0.8</sub>Ce<sub>0.2</sub>)<sub>1.6</sub>Fe<sub>12</sub>Co<sub>2</sub>B alloys were systematically investigated. Furthermore, the magnetization reversal behavior and exchange coupling for these alloys were also studied by analyzing the first-order reversal curves (FORC), which shows the information about the distribution of interaction or switching field and the ratio of the (ir)reversible magnetization processes in the sample. Such study will be valuable to reduce the rare earth content and improve the magnetic properties, providing the possibility of industrial applications of this type magnet.

II. EXPERIMENTAL DETAILS

A series of Zr-added samples with compositions (Nd<sub>0.8</sub>Ce<sub>0.2</sub>)<sub>1.6</sub>Fe<sub>12</sub>Co<sub>2</sub>BZrx (x = 0, 1, 2, 3, 4 and 5 wt%), hereafter named Zr-0, Zr-1, Zr-2, Zr-3, Zr-4 and Zr-5 respectively were synthesized by argon arc melting followed by melt spinning. Metals of Nd, Ce, Fe, Co and Zr with purity of 99.99%, and FeB (B: 18.69 wt%) were used. The ribbon samples were obtained by using the melt spinning in an argon atmosphere with a wheel speed of 25 m/s to produce ribbons. Furthermore, the as-spun ribbons were subsequently isothermally treated for 5 min at 813-1023 K under a vacuum of lower than 5 × 10<sup>-4</sup> Pa to determine the temperature T<sub>a</sub> at which the energy product (BH)<sub>max</sub> was maximized. The phase constitutions of these ribbons were characterized by X-ray diffraction (XRD) using Cu Kα radiation. The thermal behaviors were analyzed by differential scanning calorimeter (DSC, METTLER TOLEDO-1LF1600) at a constant heating rate of 10 K/min under argon atmosphere. The magnetic properties of all samples were accomplished by a Vibrating Sample Magnetometer (VSM) in the temperature range of 300-500 K. The samples for the magnetic measurements were 1-2 mm-wide, 6 mm-long and 30-μm-thick. Magnetization measurements were made with the magnetic field direction in the plane of the ribbons in order to minimize the demagnetization effect. The microstructure was investigated by FEI Technai F20 transmission electron microscope (TEM).

First-order-reversal-curve (FORC) technique has been employed to study the magnetic interactions for the Zr-free and Zr-doped nanocomposite alloys. The measurement of a FORC begins by saturating the sample with a large positive applied field. Then, reducing the field to a reversal field H<sub>s</sub>, and the FORC is the magnetization curve that the applied field starts at H<sub>a</sub> and proceeds back to saturation. Repeating this procedure for a set of different values of H<sub>a</sub>, a family of FORCs is obtained. The FORC distribution ρ(H<sub>a</sub>, H<sub>b</sub>) is defined as a mixed second derivative of the applied field H<sub>b</sub> and the reversal field H<sub>b</sub>:

\[ \rho(H_a, H_b) = -\frac{1}{2} \frac{\partial^2 M(H_a, H_b)}{\partial H_a \partial H_b} \]  \hspace{1cm} (1)

Meanwhile, the FORC diagram can be transformed into in 2-dimensional (2-D) and 3-dimensional (3-D) contour plots, mathematically converting the coordination from \{H<sub>a</sub>, H<sub>b</sub>\} to \{H<sub>a</sub>=(H<sub>a</sub>+H<sub>b</sub>)/2, H<sub>c</sub>=(H<sub>b</sub>-H<sub>a</sub>)/2\}, where the H<sub>c</sub> is the local coercivity and H<sub>a</sub> is the bias field.

III. RESULTS AND DISCUSSION

Fig. 1 shows the XRD patterns of the as-spun (Nd<sub>0.8</sub>Ce<sub>0.2</sub>)<sub>1.6</sub>Fe<sub>12</sub>Co<sub>2</sub>BZrx alloys. It can be seen that the as-spun Zr-free (Zr-0) sample possesses a large amount of amorphous phase as well as a small partial crystalline 2:14:1 phase. The volume fraction of amorphous phase increases significantly as a consequence of the increased Zr content, suggesting that Zr adding can improve the glass forming ability of the (Nd<sub>0.8</sub>Ce<sub>0.2</sub>)<sub>1.6</sub>Fe<sub>12</sub>Co<sub>2</sub>BZrx alloys.
The DSC curves for as-spun ribbons with different Zr content are shown in Fig. 2, each sample has an obvious exothermic peak caused by crystallization process. The onset crystallization temperatures ($T_x$) of $(Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_2BZr_x$ increase from 816 K to 898 K as Zr content increases from 0 to 5. The increasing of $T_x$ indicates that the thermal stability of the $(Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_2BZr_x$ samples improves with a further increase of Zr content. All the values of $T_x$ are listed in Table I.

In accordance with the above DSC results, the as-spun alloys were annealed at 813-1023 K for 5 min and then quenched to obtain the optimum temperature $T_a$ at which the maximum energy product is maximized. The demagnetization curves for the $(Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_2BZr_x$ heat treated at the optimum temperature $T_a$ are presented in Fig. 3, it can be seen all the demagnetization curves of the annealed samples show a single-phase-like behavior. All the values of magnetic properties and the optimum temperature $T_a$ are listed in Table I. Fig. 4 presents the variations in intrinsic coercivity $H_{cj}$, maximum energy product ($BH_{max}$), remanence $B_r$ and squareness $H_k/H_{cj}$ as a function of Zr concentration for the $(Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_2BZr_x$ alloys at the optimum temperature $T_a$. It shows that the intrinsic coercivity $H_{cj}$ increases first and reaches the maximum value ($H_{cj}$: 6.02 kOe) for the Zr-2 alloy, then decreases gradually with a further increase of x. However, the maximum energy product ($BH_{max}$), remanence $B_r$, and squareness $H_k/H_{cj}$ of the Zr-1 alloy decreases, compared with the Zr-free (Zr-0) alloy. With further increasing Zr content, the magnetic parameters ($BH_{max}$, $B_r$ and $H_k/H_{cj}$) of Zr-doped alloys present a tendency that increases first and then decreases gradually. The optimal magnetic properties are obtained for the Zr-3 alloy: $B_r = 9.5$ kG, $H_{cj} = 5.99$ kOe and ($BH_{max}$ = 15.2 MGOe. That means the addition of proper Zr content tends to result in a strong increase in $H_{cj}$ and ($BH_{max}$ but a slight decrease in $B_r$ only for the isotropic melt-spun Nd-Ce-Fe-Co-B alloys.

FIG. 1. XRD patterns of the as-spun $(Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_2BZr_x$ alloys.

FIG. 2. DSC traces for the as-spun $(Nd_{0.8}Ce_{0.2})_{1.6}Fe_{12}Co_2BZr_x$ alloys (heated at 10 K/min).
TABLE I. The onset crystallization temperatures $T_x$, optimum temperature $T_a$, remanence $B_r$, intrinsic coercivity $H_c$, squareness $H_{k}/H_c$ and maximum energy product $(BH)_{\text{max}}$ for $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{1-x}\text{Fe}_{12}\text{Co}_{2}\text{B}_{x}$ alloys.

| Alloys | $T_x$ (K) | $T_a$ (K) | $B_r$ (kG) | $H_c$ (kOe) | $H_{k}/H_c$ | $(BH)_{\text{max}}$ (MGoe) |
|--------|-----------|-----------|------------|------------|-------------|------------------|
| Zr-0   | 816       | 903       | 10.1       | 4.90       | 0.36        | 13.7             |
| Zr-1   | 829       | 923       | 9.1        | 5.28       | 0.32        | 11.8             |
| Zr-2   | 839       | 943       | 9.4        | 6.02       | 0.35        | 12.9             |
| Zr-3   | 867       | 963       | 9.5        | 5.99       | 0.38        | 15.2             |
| Zr-4   | 880       | 973       | 9.2        | 5.85       | 0.37        | 14.7             |
| Zr-5   | 898       | 993       | 9.1        | 5.64       | 0.34        | 14.2             |

Based on the above results, for the further information on the influence of Zr adding on the exchange coupling between the hard and soft phases, the change of $\delta M(H) = [M_d(H) - M_r + 2M_r(H)]/M_r$ was estimated and plotted in Fig. 5. Here, $M_d(H)$ is the reduced remanent magnetization, $M_o(H)$ is the reduced magnetization and $M_r$ is the saturation remanence. According to the Wohlfarth’s analysis, a negative value of $\delta M(H)$ represents the static magnetic interactions, in contrast, the positive values of $\delta M(H)$ are due to the intergranular exchange coupling effect; the maximum value of $\delta M(H)$ reflects the strength of the inter-granular exchange coupling. Fig. 5 shows the $\delta M$ plots with internal magnetic field for Zr-0, Zr-1, Zr-3 and Zr-5 alloys. A positive $\delta M$ is observed in all samples, confirming the existence of exchange-coupling interaction between the soft and hard phases. Evidently, the magnitude of the positive $\delta M$ peak for the Zr-3 alloy is the largest, confirming the strongest magnetic interaction between the grains and resulting in the highest maximum energy product. Furthermore, the Zr-3 alloy also possesses the lowest negative value of $\delta M(H)$, suggesting the lowest static magnetic exchange interaction. Meanwhile, according to the reports on the static magnetic interaction decreased with decreasing content of soft magnetic phase in Refs. 11 and 19, we consider that the content of $\alpha$-Fe phase is reduced with increasing Zr doping.

The XRD patterns of $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{1-x}\text{Fe}_{12}\text{Co}_{2}\text{B}_{x}$ alloys heat treated at the optimum temperatures $T_a$ are displayed in Fig. 6. All diffraction peaks come from hard magnetic phase 2:14:1 (H) and soft magnetic $\alpha$-Fe (S). The intensities of (110) plane for $\alpha$-Fe phase and (214) plane for 2:14:1 phase diffractions are denoted as $I_{(110)}$ and $I_{(214)}$, respectively. The peak intensity ratio $I_{(110)}/I_{(214)}$ is used to estimate the relative volume fraction of the $\alpha$-Fe phase and 2:14:1 phase by employing a semi-quantitative technique. As shown in Table II, the peak intensity ratio of $I_{(110)}/I_{(214)}$ decrease from 1.59 to 1.29 with the content of Zr from 0 to 3 in the $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{1-x}\text{Fe}_{12}\text{Co}_{2}\text{B}_{x}$ alloys. With a further increase of Zr content ($x = 5$), the sample possesses a higher value of peak intensity ratio ($I_{(110)}/I_{(214)} = 1.52$). It demonstrates that the relative volume fraction of $\alpha$-Fe phase is decreasing after the addition of proper Zr element. Based on the data of XRD, the lattice constants were calculated and listed in Table II. It can be seen that the lattice constants $a$ and $c$ of the 2:14:1 phase unit cell...
FIG. 4. Variations in intrinsic coercivity $H_{cj}$, maximum energy product $(BH)_{max}$, remanence $B_r$ and squareness $H_k/H_{cj}$ as a function of Zr concentration for the $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{1.6}\text{Fe}_{12}\text{Co}_2\text{BZr}_x$ alloys at the optimum temperature $T_a$.

decrease with Zr addition. This is an evident that the doping Zr atoms entering into the 2:14:1 phase. Since the atomic radius of Zr (0.160 nm) is smaller than that of Nd (0.264 nm) and Ce (0.182 nm), but larger than that of Fe (0.124 nm) and B (0.098 nm), the reduced lattice constants indicated that Zr substitutes rare earth in the lattice. A similar result with Zr adding is also observed in the Nd-Fe-B,\textsuperscript{20} (Nd, Dy, Y)-Fe-B\textsuperscript{11} and Ce-Fe-B\textsuperscript{12} alloys. In addition, the axial ratio of $c/a$ (1.386) remains unchanged, indicating that Zr adding in $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{1.6}\text{Fe}_{12}\text{Co}_2\text{BZr}_x$ nanocomposite alloys cause a uniform contraction of the lattice in $a$ and $c$ directions.

FIG. 5. The variation of $\delta M$ with internal magnetic field for $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{1.6}\text{Fe}_{12}\text{Co}_2\text{BZr}_x$ alloys heat treated at the optimum temperatures $T_a$.

FIG. 6. XRD patterns of $(\text{Nd}_{0.8}\text{Ce}_{0.2})_{1.6}\text{Fe}_{12}\text{Co}_2\text{BZr}_x$ alloys heat treated at the optimum temperatures $T_a$. 
TABLE II. The peak intensity ratios of $I_{(110)S}/I_{(214)H}$ and the lattice parameters ($a$, $c$ and $c/a$) of 2:14:1 phase for (Nd$_{0.8}$Ce$_{0.2}$)$_{1.6}$Fe$_{12}$Co$_{2}$BZr$_{x}$ alloys heat treated at the optimum temperatures $T_a$.

| Alloys | $I_{(110)S}/I_{(214)H}$ | $a$(Å) | $c$(Å) | $c/a$ |
|--------|-------------------------|--------|--------|------|
| Zr-0   | 1.59                    | 8.804  | 12.202 | 1.386|
| Zr-1   | 1.32                    | 8.799  | 12.197 | 1.386|
| Zr-3   | 1.29                    | 8.793  | 12.187 | 1.386|
| Zr-5   | 1.52                    | 8.778  | 12.166 | 1.386|

To further establish the detailed nature of the structure, TEM micrographs of the Zr-0 and Zr-3 alloys heat treated at the optimum temperatures $T_a$ are performed in Fig. 7. Figs. 7(a) and (b) show the microstructure of the Zr-free sample consists of regular 2:14:1 grains and fine $\alpha$-Fe grains, which has been reported in our previous paper (see Ref. 10). In addition, Fig. 7(c) shows the bright-field image of the Zr-3 alloy heat treated at the optimum temperatures $T_a$. Apparently, the Zr-3 sample does also consist of 2:14:1 phase and $\alpha$-Fe phase with a more uniform microstructure, while the amount of $\alpha$-Fe phase decreases, which is consistent with previous results of XRD measurements and $\delta M(H)$ plots. Up to 40 grains were used to determine the average grain size which, for Zr-0 sample is

![FIG. 7. TEM micrographs of the Zr-0 and Zr-3 alloys heat treated at the optimum temperatures $T_a$. (a) bright-field image and its SAED for the bright-field image and its SAED for the Zr-0 alloy; (b) HRTEM image and its FFT diffraction pattern of the $\alpha$-Fe phase in (a); (c) bright-field image for the Zr-3 alloy, the HRTEM image and its FFT diffraction pattern corresponding to 2:14:1 phase were shown in the inset; (d) HRTEM image and its FFT diffraction pattern of the $\alpha$-Fe phase in (c). “Reproduced with permission from Tan et al., J. Magn. Magn. Mater. 437, 17 (2017). Copyright 2017 Elsevier B.V.”](image-url)
25 nm for the α-Fe phase and 43 nm for the 2:14:1 phase. These values are larger than those found for the Zr-3 sample, i.e., 12 nm for the α-Fe phase and 35 nm for the 2:14:1 phase, respectively. It means that the addition of Zr can suppress the grain growth of the α-Fe grain and the 2:14:1 grain. Furthermore, compared with the soft magnetic α-Fe phase for the two samples (Figs. 7(a) and (c)), the soft magnetic α-Fe grains are enriched at the grain boundaries and also embedded dispersively in 2:14:1 grains for the Zr-0 sample, while the soft magnetic α-Fe grains for the Zr-3 sample are uniform distribution between the hard magnetic 2:14:1 grains. It indicates that the existence of enhancement magnetic interactions between the soft magnetic and hard magnetic grains. A fine microstructure can contribute to the enhancement of exchange interaction between hard and soft magnetic grains. The increase of $H_{c}\gamma$ and $(BH)_{\text{max}}$ for the Zr-3 sample in comparison with Zr-0 sample may result from a smaller grain size and uniform distribution of soft magnetic α-Fe phase.

In order to fully understand the process of magnetization reversal in detail, we selected the alloys with Zr-0 and Zr-3 samples using the FORC technique for more detailed analysis. To calculate the $\rho (H_a, H_b)$, we choose the FORC diagrams in this paper were evaluated with SF (smoothing factor) $= 3, 22$. Fig. 8 shows the FORCs, 2-D and 3-D diagrams of the Zr-0 and Zr-3 alloys heat treated at the optimum temperatures $T_{\alpha}$. The experimental FORC curves for the Zr-0 and Zr-3 alloys are presented in Figs. 8(a1) and (b1), under increasing applied magnetic fields, each of the FORCs traverses gradually increasing before conforming onto the major loop. As one can see the 2-D diagrams in Figs. 8(a2) and (b2) that there is only one strong spot along the $H_c$-axis, one located at $H_c \approx 5.1$ kOe for the Zr-0 sample and one located at $H_c \approx 6.3$ kOe for the Zr-3 sample, respectively. Meanwhile, on comparing with the Zr-0 sample, the 2-D diagrams of Zr-3 sample show an obvious narrower and lower slope of weak line pattern with a pair of negative and positive $\rho$, implying Zr addition results in an obvious reduction of the irreversible domains due to a presence of strong pinning sites. In both Figs. 8(a3) and (b3), it is found that only one major peak in the FORC diagram, indicating that a single-phase magnetization reversal behavior is observed for the Zr-0 and Zr-3 nanocomposite alloys. Moreover, a higher maximum $\rho$ of the major peak and the flat surface have been obtained for the Zr-3 sample, demonstrating the fact of weaker magnetostatic interaction but stronger interphase exchange coupling, which is consistent with the results of $\delta M(H)$. It is interesting to note that a distinct kink is observed at a half position of the major peak for the 3-D diagrams of the Zr-3 sample in Fig. 8(b3), despite it obtains a higher maximum $\rho$, suggesting that the weak exchange interaction between the two magnetic phases at the specific magnetic field. To make quantitative analysis of the exchange-coupling interaction behavior and coercive field distributions, the integrating horizontal line scans by the projection along the $H_c$-axis at $H_u = 1100$ Oe (Zr-0) and $H_u = 900$ Oe (Zr-3) were shown in Fig. 9. Meanwhile, a Gaussian fit of these FORC projections and the full width at half maximum (FWHM) are taken into account, it can be seen that the peak location of the Gaussian fit is increased and the FWHM is also increased from 1785 Oe to 2452 Oe for the Zr-0 and Zr-3 samples, respectively. The increase of peak location indicates the increase of the reverse field, while the increase of FWHM for the Zr-3 sample shows the enhanced strong exchange-coupling interaction. It is interesting to note that despite a kink is observed at a half position of the major peak for the Zr-3 sample, it still exhibits a higher FWHM due to an increase exchange-coupling, suggesting that the FWHM can also give the mean field interaction effect as a whole.

For a better understanding of coercivity mechanism and the reason for the higher value of the magnetic properties in Zr-doped samples, the magnetic microstructural parameters were calculated quantitatively according to Kronmüller’s formulation of the Brown model:

\[
\frac{H_{c\gamma}(T)}{M_s(T)} = \alpha_k \alpha_{ex} \frac{H_{\text{eff}}^\text{min}(T)}{M_s(T)} - N_{\text{eff}}
\]  

(2)

Where $\alpha_k$, $\alpha_{ex}$ and $N_{\text{eff}}$ are so-called microstructural parameters and are related to the non-ideal microstructure of a real magnet. The parameter $\alpha_k$ describes the reduced surface anisotropy of nonperfect grains, $\alpha_{ex}$ describes the effect of exchange coupling between the neighboring grains on the coercive field. $N_{\text{eff}}$ is an average effective local demagnetization factor, describing the internal
stray fields acting on the grains. According to Stoner and Wohlfarth model, the minimum nucleation field $H_{N}^{\text{min}}$ represents the value for the nucleation field of the most unfavorably aligned grains belonging to misalignment angle of 45° to the applied field. Fig. 10 shows the plots of $H_{cJ}(T)/M_s(T)$ vs $H_{N}^{\text{min}}(T)/M_s(T)$ in the temperature range of 300-500 K for the Zr-0 and Zr-3 alloys samples. It shows a good linear relationship for both Zr-0 and Zr-3 nanocomposite alloys, suggesting that nucleation of reverse domains is the dominant mechanism in these nanocomposites. The fitting parameters can be obtained by a standard linear least-squares method according to Equation (2), the value of $\alpha_k\alpha_{ex}$ is 0.145 and 0.197, $N_{\text{eff}}$ is 0.002 and 0.047 for the Zr-0 sample and Zr-3 sample, respectively.
FIG. 9. The projection of the FORC distribution of the Zr-0 and Zr-3 alloys heat treated at the optimum temperatures $T_a$ onto the $H_u$-axis and Gaussian fit highlighting the mean switching field and its distribution.

FIG. 10. The plots of $H_{cj}(T)/M_s(T)$ versus $H_{min}^N(T)/M_s(T)$ in the temperature range of 300-500 K for the Zr-0 and Zr-3 alloys heat treated at the optimum temperatures $T_a$.

As known, the value of coefficient $\alpha_k$ can assume to be about 0.8,28,29 then the coefficient $\alpha_{ex}$ of the Zr-free sample and Zr-doped sample are 0.181 and 0.246, respectively. It shows that the increase in $\alpha_{ex} = 0.246-0.181 = 0.065$ in our samples result in a drastic increase of coercivity $H_{cj}$ about 1.09 kOe = 5.99-4.90 kOe in the Zr-3 sample.

In summary, a certain amount Zr doping in the (Nd$_{0.8}$Ce$_{0.2}$)$_{1.6}$Fe$_{12}$Co$_2$B alloys can be helpful for reducing the content and improving the uniform distribution of $\alpha$-Fe phase, which led to the strong exchange coupling interaction between magnetically hard and soft phases. Thus, the magnetic properties $H_{cj}$, $(BH)_{max}$ and $H_k/H_{cj}$ are all improved for the (Nd$_{0.8}$Ce$_{0.2}$)$_{1.6}$Fe$_{12}$Co$_2$BZr$_x$ alloy with the Zr addition of $x = 3$.

IV. CONCLUSIONS

In this work, the effect of Zr doping on the magnetic properties and FORC diagrams of (Nd$_{0.8}$Ce$_{0.2}$)$_{1.6}$Fe$_{12}$Co$_2$B alloys were investigated. A certain amount Zr doping could reduce the content of $\alpha$-Fe phase and enhance the exchange coupling interaction, which led to an improvement in $H_{cj}$, $(BH)_{max}$ and $H_k/H_{cj}$. The enhanced magnetic properties can be obtained at the optimum temperature $T_a$ for the Zr-3 alloy: $B_t = 9.5$ kG, $H_{cj} = 5.99$ kOe and $(BH)_{max} = 15.2$ MGOe. The FORC analysis provides more information on the magnetostatic as well as the exchange interactions. Meanwhile, nucleation was found to be the dominant magnetization mechanism in (Nd$_{0.8}$Ce$_{0.2}$)$_{1.6}$Fe$_{12}$Co$_2$BZr$_x$ alloys and the increase of $\alpha_{ex}$ resulting in a drastic increase of coercivity $H_{cj}$ in the Zr-3 sample.
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