Effect of Annealing Process on Microstructure and Magnetic Properties of FeSiBPCNbCu Nanocrystalline Soft Magnetic Powder Cores

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Abstract: In this work, the effect of the annealing process (heating rate (HR) and holding time (HT)) on microstructure and magnetic properties of FeSiBPCNbCu nanocrystalline soft magnetic powder cores (NPCs) are systematically studied. NPCs annealed under the HR of 100 K/min and HT of 30 min exhibit a finely uniform nanocrystalline structure with a concentrated grain size distribution and a small average grain size of 18.71 nm and show a modified magnetic domain structure where the domain walls move easily with the changes in the external magnetic field. The optimized microstructure leads to outstanding soft magnetic properties, including the low coercivity of 10.5 A/m (50 mT, 500 kHz), low core loss of 751 mW/cm³ (50 mT, 500 kHz), and highly effective permeability of 64.7 (100 kHz). We believe that the annealing process regulation can contribute to the industrial production of high-performance NPCs, which can meet the performance requirements of high-end electronic components such as molding chokes used in power supplies, mobile phones, and other terminal equipment.

Keywords: nanocrystalline powder cores; annealing process; magnetic domain structure; core loss

1. Introduction

As a new type of soft magnetic material, nanocrystalline soft magnetic alloys have been widely used in the field of power electronics due to their excellent magnetic properties, since they were first reported in 1988 by Yoshizawa, Y. et al. [1–4], and nanocrystalline soft magnetic powder cores (NPCs) produced by pressing coated nanocrystalline powder have been carried out [5–8]. Compared with traditional iron, iron silicon, sendust, and iron-nickel soft magnetic powder cores (SMPCs), NPCs show better soft magnetic properties, including lower coercivity ($H_c$) and lower core loss ($P_{cv}$) at a high frequency due to their unique structure where nano-sized $\alpha$-Fe(Si) grains are densely distributed in the residual amorphous phase, which can better meet the development trend of high frequency, miniaturization, and high energy efficiency of electronic components [9,10].

The microstructure of the NPCs is an essential factor that affects their soft magnetic properties [11]. The grain size ($D$) of nano-sized $\alpha$-Fe(Si) grains is generally smaller than the exchange length ($L_e$), and its relationship with $H_c$ ($H_c \propto D^6$) has been fully described and illustrated according to the random anisotropy model (RAM) [4,12]. To obtain NPCs with a...
uniform and fine nanocrystalline structure, the heating rate (HR) and holding time (HT) during the annealing process can be adjusted to control the nucleation and growth process of nano-sized α-Fe(Si) grains. Li, Z. et al. [13] investigated the effect of ultra-rapid annealing (URA) on the microstructure and soft magnetic properties of (Fe1-x-yCo,Ni)56B14 alloys, and nanocrystalline (Fe0.8Co0.05Ni0.15)86B14 with outstanding performances with low $H_c$ of 2 A/m was prepared by URA successfully. Jiang, L. et al. [14] adjusted the annealing process of Fe81Si5.5B14P11Cu0.5C2 alloy and found that a low HR of 10 K/min is beneficial for the alloy to achieve a fine structure where the mean grain size is 17 nm, and the $H_c$ is about 4.8 A/m. In addition, with the extension of HT, the mean size of α-Fe(Si) grains gradually becomes larger. Meng, Y. et al. [15] found that the appropriate rapid annealing with higher temperatures and shorter HT is beneficial to the higher crystallinity and the formation of the fine α-Fe(Si) grains in Fe53Si4B10P2Cu1 ribbons and leads to good magnetic properties, including high saturation magnetization ($M_s$) of 1.82 T and low $H_c$ of 6.2 A/m.

In our previous study, high-performance FeSiBPCnCu NPCs were prepared by a traditional annealing process, while the microstructure is not ideal. The low crystallization volume fraction and large grain size have a negative impact on the magnetic properties, and there is still much room for optimization. In this work, we further investigated the effects of HR and HT on the microstructure and soft magnetic properties of FeSiBPCnCu NPCs and summarized the evolution of the crystalline structure during the annealing process. It is found that a uniform nanocrystalline structure with fine α-Fe(Si) grains of 18.71 nm can be obtained when the HR and HT were 100 K/min and 30 min, respectively. Due to the optimized structure, the NPCs show excellent soft magnetic properties, including low $H_c$ of 10.5 A/m (50 mT, 500 kHz), high effective permeability ($\mu_e$) of 64.7 (100 kHz), and low $P_{ce}$ of 751 mW/cm$^2$ (50 mT, 500 kHz). We believe that this work can contribute to the industrial production of high-performance NPCs, which can meet the performance requirements of high-end electronic components.

2. Materials and Methods

Alloy ingots with the composition of Fe74.75Si8B10P3C2Nb1.5Cu0.75 were prepared by induction melting a mixture of industrial materials including Fe (99.6 wt. %), Si (99.9999 wt. %), Nb (99.9 wt. %), Cu (99.99 wt. %), pre-alloyed FeB (99.4 wt. %), and FeP (99.4 wt. %). Amorphous precursor powder was then fabricated by gas-atomizing the remelted alloy ingots with industrial raw materials at a dynamic pressure of 8 MPa. The commercial W-6C epoxy resin (2 wt. %) was chosen as the binder and evenly mixed with the gas-atomized powder. Powder cores with an outer diameter of 20.3 mm and an inner diameter of 12.7 mm were produced by pressing the mixed powder under the pressure of 1800 Mpa. The final annealing processes were carried out by controlling HR and HT using a rapid heating tube furnace (BTF-1200C-RTP-S90B) to achieve NPCs; the annealing temperature is 783 K, while the HR and HT range from 5–500 K/min and 0–90 min, respectively.

The thermal stability of the gas-atomized powder was measured by a differential scanning calorimetry (DSC, 404C, Netzsch, Selb, Germany with a heating rate of 40 K/min. The hysteresis loop of the gas-atomized powder was measured by a vibrating sample magnetometer (VSM, Lakeshore 7410, Columbus, OH, USA) with an applied field up to 800 kA/m. The microscopic morphology of the gas-atomized powder and NPCs were observed by a scanning electron microscope (SEM, ZEISS EVO 18, ZEISS, Oberkochen, Germany) with Cu-Kα radiation and a transmission electron microscopy (TEM, JEM2100, JOEL, Tokyo, Japan). Specimens of NPCs for TEM were processed by a focused ion beam/scanning electron microscope (FIB/SEM, Carl Zeiss Auriga, ZEISS, Oberkochen, Germany). The dynamic magnetic domains of the NPCs were observed by a Lorentz transmission electron microscope (JEM-2100F, JOEL, Tokyo, Japan). The $\mu_e$ of NPCs were measured by an impedance analyzer (Agilent 4294A, Palo Alto, CA, USA) from 1 kHz to
10 MHz. The $P_{ev}$ and $H_c$ of NPCs were measured by an alternating current (AC) B-H loop analyzer (SY-8218, IWATSU, Tokyo, Japan).

3. Results and Discussion

3.1. Characterization of Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ Powder and NPCs Coating Behavior

The SEM image of gas-atomized Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ powder with no heat treatment is shown in Figure 1a. The powder fabricated by gas atomization shows excellent surface morphology, including smooth surface, high sphericity, and uniform shape, which is conducive to uniform coating and the minimization of $P_{ev}$ [16–18].

![Figure 1](image-url)

**Figure 1.** (a) SEM image, (b) XRD pattern, (c) DSC curve, and (d) hysteresis loop of gas-atomized Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ powder.

The XRD pattern of gas-atomized Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ powder is shown in Figure 1b, only a broad peak at around $2\theta = 45^\circ$ can be observed in the pattern, indicating that the gas-atomized powder has a fully amorphous structure. Figure 1c shows the DSC curve of the powder. There are two endothermic peaks with the starting temperatures accruing at 784 K and 857.4 K, which are defined as the first crystallization temperature ($T_{x1}$) of the $\alpha$-Fe(Si) phase and the second crystallization temperature ($T_{x2}$) of the non-soft magnetic Fe-B phases, respectively [19]. The $\Delta T (\Delta T = T_{x2} - T_{x1})$ of the amorphous powder is considered a parameter of the thermal stability of the remaining amorphous state above $T_{x1}$, and the value is 73.4 K. The hysteresis loop of the powder is shown in Figure 1d. The powder shows a high saturation magnetization ($M_s$) of 150.9 emu/g, 13.6% higher than that of the commercial 1K107 powder (~133 emu/g) [3], which is beneficial for the miniaturization of electronic devices. In addition, the powder exhibits a low $H_c$ of 0.1265 kA/m, which is propitious to the loss reduction of devices during operation and can improve the utilization rate of energy.

Figure 2a shows the cross-section morphology of a selected powder inside the Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs when the HR and HT were 100 K/min and 30 min, respectively. The dividing line between the powder and epoxy resin can be observed clearly, confirming that the powder can still be uniformly coated by the epoxy resin after the annealing process, which can significantly increase the resistivity of NPCs and reduce the eddy current loss.
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(Pc). Figure 2b shows the enlarged image of the selected area from Figure 2a, and a path in the enlarged image was selected to perform a line scan of the element distribution. The corresponding main elements distribution lines are shown in Figure 2c. It can be seen that the elements are evenly distributed inside the powder, when the scanning position reaches the boundary between the powder and epoxy resin, the signal intensities of Fe and Si drop sharply, while the signal intensities of C and O rise accordingly, which are the constituent elements of epoxy resin, proving that epoxy resin has an effective isolation effect on the magnetic powders.

Figure 2. (a,b) Cross-section morphology and (c) corresponding main elements distribution lines of a selected powder inside the Fe74.75Si8B10P3C2Nb1.5Cu0.75 NPCs.

3.2. Magnetic Properties and Microstructures of Fe74.75Si8B10P3C2Nb1.5Cu0.75 NPCs Annealed at Different HR

Annealing processes with different HR (5, 40, 100, 300, and 500 K/min) and the same HT of 60 min were applied for the fabrication of Fe74.75Si8B10P3C2Nb1.5Cu0.75 NPCs. Figure 3a shows the μe of the NPCs annealed at different HR. The μe all show good high-frequency stability, the values are basically unchanged even under the high frequency of 10 MHz, which is beneficial for the stable operation of the devices at a high frequency. The NPCs annealed at the HR of 5 K/min show a low μe of 59.2 (100 kHz); as the HR gradually rises to 100 K/min, the μe gradually increases to the maximum value, which is 63.2. When the HR continues to rise, the μe shows a downward trend. As the HR further rises to 500 K/min, the μe gradually decreases from the maximum value of 63.2 to 60.9.

The dependence of Pc on frequency is shown in Figure 3b, and the applied field is 50 mT. The Pc rises with the increase in frequency for all NPCs, and the NPCs annealed at the HR of 100 K/min show the lowest Pc at all frequencies. The comparison of Pc tested at the frequency of 500 kHz is shown in Figure 3c. When the HR increases from 5 to 100 K/min, Pc gradually decreases from 906 mW/cm³ to the minimum value, which is 759 mW/cm³. As HR continues to rise to 500 K/min, the Pc deteriorates to 933 mW/cm³. Figure 3d shows the corresponding Hc change curve, which is similar to Pc. The Hc of the NPCs decreases from 12.9 to 10.8 A/m as the HR rises from 5 to 100 K/min and then increases to 12.6 A/m when the HR further comes to 500 K/min, indicating that Hc is the crucial factor affecting Pc.

The crystallization behaviors of NPCs annealed at different HR were investigated to further explore the root causes of the changes in soft magnetic properties. The bright-field TEM images of Fe74.75Si8B10P3C2Nb1.5Cu0.75 NPCs annealed at different HR (40, 100, and 500 K/min) are shown in Figure 4a–c, whereas the inset presents the selected area electron diffraction (SAED) patterns. All the NPCs show a unique microstructure where nano-sized grains disperse randomly in the residual amorphous phase, and the grains are defined as the α-Fe(Si) phase by the analysis of the SAED patterns. The high-resolution TEM images are shown in Figure 4d–f. The nano-sized grains of three samples all show noticeable lattice
fringes, whose distances are consistent with the interplanar spacing of the (110) plane of the \( \alpha \)-Fe(Si) phase. Figure 4g shows the grain size distribution of NPCs annealed at different \( HR \), which are curved in the histograms and fitted by a normal distribution function with average grain sizes \((D_{\text{avg}})\) of 25.17, 20.94, and 23.01 nm, respectively.

The grain density of \( \alpha \)-Fe(Si) grains in NPCs annealed at the \( HR \) of 100 K/min is much higher than that of \( HR = 40 \) and 500 K/min (Figure 4a–c). In addition, the NPCs annealed at the \( HR \) of 100 K/min show a minimum \( D_{\text{avg}} \) compared with others, and the grains are concentrated between 15 and 25 nm with a fraction of 55%, while the fraction between 10 and 30 nm is 88%, indicating a concentrated grains size distribution. For NPCs annealed at the \( HR \) of 40 and 500 K/min, \( \alpha \)-Fe(Si) nanocrystals show a less concentrated grain size distribution with more large grains. Therefore, unlike the traditional theory that high \( HR \) is beneficial for forming a finely uniform nanocrystalline structure \([20,21]\), a moderate \( HR \) is better for this alloy composition to obtain \( \alpha \)-Fe(Si) grains with high grain density and a relatively small grain size.

A finely uniform nanocrystalline structure with a high grain density of \( \alpha \)-Fe(Si) grains is beneficial for magnetic softness. According to the single-phase theoretical model by Herzer \([12]\), the exchange coupling between grains shows the easy magnetization direction arranged in parallel when the grains are down on the nanometer scale, and it is no longer the magnetocrystalline anisotropy constant \((K) \) of a single grain that affects the magnetization but the average magnetocrystalline anisotropy \((K_1) \) \([22]\). When the grain size is smaller than the ferromagnetic exchange coupling length \((L_{\text{ex}}) \), which is about 35 nm, \( K_1 \) decreases with the decrease of the grain size. The relationship between \( K_1 \) and grain size can be concluded by Equation (1):

\[
K_1 = \frac{K^4D_{\text{avg}}^6}{A^3}
\]  

(1)
where $D_{\text{avg}}$ is the average grain size, and $A$ denotes the exchange stiffness. $H_c$ and $\mu_e$ are followed by Equations (2) and (3), respectively:

$$H_c \propto \frac{K_4^1 D_{\text{avg}}^6}{J_s A^3}$$

(2)

$$\mu_e \propto \frac{J_s A^3}{\mu_0 K_4^1 D_{\text{avg}}^6}$$

(3)

where $J_s$ is the saturation magnetization, and $\mu_0$ is the vacuum permeability constant. Therefore, the $H_c$ of the nanocrystalline soft magnetic alloy is proportional to $D_{\text{avg}}^6$, and $\mu_e$ is inversely proportional to $D_{\text{avg}}^6$. The refinement of the grains is beneficial to the improvement of the soft magnetic properties, including the decrease of $H_c$ and the increase of $\mu_e$. It can be seen from Figures 3d and 4g that $H_c$ and $D_{\text{avg}}$ both reach the lowest value when $HR$ is 100 K/min, namely 10.8 A/m and 20.94 nm, respectively, which is consistent with the above conclusion; therefore, the $P_{cv}$ of NPCs reaches to the minimum, since $H_c$ is the crucial factor affecting $P_{cv}$. In addition, the increase of $\mu_e$ shown in Figure 3a is also attributed to the refinement of grain size. In this regard, we concluded that, in this alloy system, a moderate heating rate can make it obtain a finely uniform nanocrystalline structure, thereby optimizing soft magnetic properties.

![Figure 4](image-url)

Figure 4. (a–c) TEM images and SAED patterns, (d–f) corresponding high-resolution TEM images, and (g) grain size distribution of Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs annealed at different $HR$. 
3.3. Magnetic Properties and Microstructures of Fe\textsubscript{74.75}Si\textsubscript{8}B\textsubscript{10}P\textsubscript{3}C\textsubscript{2}Nb\textsubscript{1.5}Cu\textsubscript{0.75} NPCs Annealed for Different HT

The HT for NPCs was subsequently adjusted to further optimize the structure and performance; annealing processes with different HT (0, 10, 30, and 90 min) and the optimized HR of 100 K/min were applied for Fe\textsubscript{74.75}Si\textsubscript{8}B\textsubscript{10}P\textsubscript{3}C\textsubscript{2}Nb\textsubscript{1.5}Cu\textsubscript{0.75} NPCs. The $\mu_e$ of the NPCs annealed for different HT is shown in Figure 5a. The NPCs show a low $\mu_e$ of 39.7 at the frequency of 100 kHz when HT is 0 min, and as HT is extended from 0 to 10 min, $\mu_e$ rises sharply from 39.7 to 60.9, and then, the amplitude of the increase tends to be flat, and the $\mu_e$ of NPCs reaches the maximum value of 64.7 with an HT of 30 min. As HT further reaches 90 min, the $\mu_e$ gradually decreases from the maximum to 62.3.

The dependence of $P_{cv}$ on the frequency for NPCs is shown in Figure 5b, and the applied field is 50 mT. NPCs annealed for the HT of 10, 30, and 60 min all show extremely low $P_{cv}$, which are very close. The comparison of $P_{cv}$ tested at 500 kHz is shown in Figure 5c. NPCs annealed for 0 min show a high $P_{cv}$ of 2429 mW/cm\textsuperscript{3}. As HT extends from 0 min to 10 min, $P_{cv}$ decreases sharply from 2429 to 756 mW/cm\textsuperscript{3}. When HT comes to 30 min, $P_{cv}$ reaches the minimum value of 751 mW/cm\textsuperscript{3}. When HT further extends to 90 min, $P_{cv}$ gradually increases from 751 to 939 mW/cm\textsuperscript{3}. The $H_c$ change curve is shown in Figure 5d, which has the same trend as $P_{cv}$. The $H_c$ of NPCs decreases from 33.5 A/m to 10.5 A/m as HT rises from 0 to 30 min and then increases to 13.2 A/m when HT further comes to 90 min. The $P_{cv}$ and $H_c$ both reach the lowest value when the HT is 30 min, indicating that $H_c$ is the key factor affecting $P_{cv}$, which is consistent with the above conclusion.

The crystallization behaviors of NPCs annealed for different HT were further analyzed by TEM. Figure 6a,b show the bright-field TEM images and SAED patterns of Fe\textsubscript{74.75}Si\textsubscript{8}B\textsubscript{10}P\textsubscript{3}C\textsubscript{2}Nb\textsubscript{1.5}Cu\textsubscript{0.75} NPCs annealed for different HT (10 and 30 min). The NPCs annealed for 10 min mainly show an amorphous structure where only a few crystal grains can be observed, and no obvious diffraction spots can be seen from the SAED pattern. NPCs annealed for 30 min show a unique microstructure where nano-sized grains are densely distributed in the amorphous matrix, and the crystalline phase of these nano-sized grains can be defined as the $\alpha$-Fe(Si) phase by the analysis of the SAED patterns. The grain size distribution of the NPCs annealed for 30 min is shown in Figure 6c, which is curved in a histogram and fitted by a normal distribution function with $D_{avg}$ of 18.71 nm.

![Figure 5](image-url)\(^{(a)}\) $\mu_e$, (b) $P_{cv}$, (c) histogram of $P_{cv}$, and (d) $H_c$ change curve of Fe\textsubscript{74.75}Si\textsubscript{8}B\textsubscript{10}P\textsubscript{3}C\textsubscript{2}Nb\textsubscript{1.5}Cu\textsubscript{0.75} NPCs annealed for different HT.
The differences in the crystallization structures of Fe_{74.75}Si_{8}B_{10}P_{3}C_{2}Nb_{1.5}Cu_{0.75} NPCs annealed for different HT are mainly controlled by the growth of α-Fe(Si) grains. The XRD patterns of NPCs annealed for different HT (10 min, 30 min, and 60 min) are shown in Figure 7. Only a broad peak at around 2θ = 45° can be observed in the pattern for the NPCs annealed for 10 min, indicating that the structure is still mainly amorphous, which is consistent with the TEM image and SAED pattern (Figure 6a). It is reasonable to speculate that the interiors of NPCs are completely amorphous when annealed for 0 min. NPCs annealed for 30 and 60 min both exhibit three peaks corresponding to the α-Fe(Si) phase, according to the standard PDF card (PDF#35-0519@PDF2-2004), and the intensity increases gradually with the rise of HT, indicating the growth of α-Fe(Si) grains, which is consistent with the trend of the particle size variation shown in Figures 4g and 6c.

The amorphous matrix in Fe-based nanocrystalline alloy has a large positive magnetostriction coefficient (λ_s), which leads to a high H_c. Thus, NPCs annealed for 0 min show a large H_c of 33.5 A/m and P_cv of 2429 mW/cm^3 due to the completely amorphous structure. The precipitation of α-Fe(Si) grains, which show negative λ_s, can effectively neutralize the positive λ_s of the amorphous matrix and leads to the decrease of H_c. However, H_c also has a great relationship with the grain size, which is shown in Equation (2); the excessively grown grains would cause a sharp rise in H_c, thereby deteriorating the soft magnetic properties of NPCs. The optimization effect of α-Fe(Si) grain precipitation on λ_s and the deterioration effect on H_c reach equilibrium when HT is 30 min, making the H_c of NPCs reach the minimum value of 10.5 A/m (Figure 5d). Therefore, the P_cv of NPCs reaches the minimum, since H_c is the crucial factor affecting P_cv. The μ_e of NPCs also reaches the maximum value of 64.7 (Figure 5a) due to the optimized nanocrystalline structure.
3.4. Magnetic Domain Motion Behavior of Fe\textsubscript{74.75}Si\textsubscript{8}B\textsubscript{10}P\textsubscript{3}C\textsubscript{2}Nb\textsubscript{1.5}Cu\textsubscript{0.75} NPCs

The magnetic properties of the NPCs are strongly related to the magnetic domain structure. $H_c$ represents the strength of the reverse magnetic field that needs to be applied after NPCs are magnetized to saturation and then completely demagnetized (magnetization $M = 0$) [23], which is closely related to the motion behavior of the magnetic domains [24–27]. Therefore, revealing the relationship between magnetism and the microstructure is crucial for controlling magnetic behavior in NPCs.

The Lorentz transmission electron microscope (LTEM) was applied to observe the magnetic domain structure. In order to explore the effect of the microstructure on the movement of the magnetic domain walls, NPCs with an HR of 40 K/min, HT of 60 min and HR of 100 K/min, HT of 30 min are selected for comparison, which has the largest and smallest average grain sizes, respectively. The in-focus, overfocused, and under-focused LTEM images are shown in Figures 8a–c and 8d–e, respectively. The overfocused sample is superimposed with black lines (indicating insufficient electrons) and white lines (indicating excess electrons), directly showing the magnetic domain boundaries in the sample. As the sample is in the under-focused position, the fringe contrast observed at this time is just the opposite of that of the overfocused. As indicated by the red circles in Figure 8b,c, a few light spots can be observed inside the NPCs annealed with the HR of 40 K/min and HT of 60 min, where the domain walls show finer branches and further increase the complexity of the magnetic domain structure. Meanwhile, the NPCs annealed under the HR of 100 K/min and HT of 30 min show a simpler magnetic domain structure; the magnetic domain walls are clearer without obvious branches.

The external magnetic field applied to the NPCs would lead to the rotation of magnetic moments and displacement of domain walls [28–30]; the dynamic processes of the magnetic domain movement for NPCs annealed under different conditions (HR = 40 K/min, HT = 60 min and HR = 100 K/min, HT = 30 min) are shown in Figure 9a,b and Figure 9c,d, respectively. By observing the magnetic domain wall perpendicular to the direction of the external field, It can be seen that the magnetic domain wall of NPCs annealed under the HR of 40 K/min and HT of 60 min starts to move when the applied field is 83 Oe, and the moving distance is about 0.24 μm, while the magnetic domain wall of NPCs annealed under the HR of 100 K/min and HT of 30 min starts to move when the applied field is 25 Oe, and the moving distance is about 0.75 μm.
The dynamic processes of the magnetic domain movement for NPCs annealed under different conditions, (a,d) in focus, (b,e) overfocused, and (c,f) under-focused.

Figure 8. LTEM images of Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs annealed under different conditions: (a,d) 40 K/min, 60 min; (b,e) 100 K/min, 30 min; (c,f) 40 K/min, 60 min; (d) 100 K/min, 30 min; (e,f) under-focused.

Figure 9. LTEM images for the field dependence of domain wall evolution in Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs annealed under different conditions: (a,b) 40 K/min, 60 min; (c,d) 100 K/min, 30 min.
After the optimizing procedure, the movement of the magnetic domain walls of NPCs becomes easier and shows a longer moving distance even in a smaller external field, indicating a sharp drop of $H_c$. During the magnetization of NPCs, these branches observed on the domain walls of NPCs annealed under the $HR$ of 40 K/min and $HT$ of 60 min severely hinder the progress of magnetization, resulting in a larger $H_c$ [31]. After the optimization of heat treatment processes, no obvious branches can be observed; thus, the movement of the magnetic domain walls becomes easier.

The dynamic processes of the magnetic domain movement of NPCs strongly reveal the relationship between magnetism and microstructure: A nanocrystalline structure with a less-concentrated grain size distribution and large grain size leads to a complex magnetic domain structure, which deteriorates the magnetic properties, while a finely uniform nanocrystalline structure leads to a simple and clear magnetic domain structure with no pinning points that significantly reduce the $H_c$ and $P_{cv}$ of NPCs.

3.5. Nanocrystallization Processes of Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs

The probable schematics of nanocrystallization processes in Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs are reasonably assumed in Figure 10. The NPCs show a disordered amorphous structure before annealing. When the annealing temperature gradually increases, the Fe element in the NPCs exhibits a concentration difference due to the Cu clusters [32,33], and the enriched Fe element precipitates and grows in the form of $\alpha$-Fe(Si) grains.

$HR$ is a decisive factor affecting the crystallization structure during the crystallization process. When NPCs are crystallized at a low $HR$, although a high-density $\alpha$-Fe(Si) core can be formed, a part of the grains are absorbed during the nucleation and growth process due to its slow $HR$, resulting in the discrete grain size distribution and relatively large grain sizes (Figure 4g). When NPCs are crystallized at a high $HR$, the amorphous matrix fails to form a uniform and dense nucleation. Subsequent grain growth also unfolds based on this, eventually leading to an uneven crystalline structure. The moderate $HR$ can avoid the mutual engulfment of grains and the less dense nucleation, and finally, form a fine and uniform nanocrystalline structure with a relatively small grain size.

![Figure 10. Schematic of the crystallization behavior of Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs.](image)

$HT$ further regulates the growth of grains. The NPCs basically maintain the amorphous structure, and only a few $\alpha$-Fe(Si) grains can be precipitated in the amorphous matrix when...
the HT is too short, which leads to a low crystallization volume fraction. However, although a high crystallization volume fraction can be obtained when the HT is long, it would cause the excessive growth of α-Fe(Si) grains, resulting in an increase of $H_c$ and a decrease of $\mu_e$. A moderate HR is beneficial to obtain an optimal nanocrystalline structure and optimize the performance of the NPCs to prepare electronic components with an excellent performance suitable for high-frequency fields.

4. Conclusions

In this work, the effects of HR and HT on the microstructure, crystallization behavior, and soft magnetic properties of Fe$_{74.75}$Si$_8$B$_{10}$P$_3$C$_2$Nb$_{1.5}$Cu$_{0.75}$ NPCs were investigated systematically. The main conclusions are as follows:

1. The moderate HR and HT of 100 K/min and 30 min lead to a finely uniform nanocrystalline structure, which shows a concentrated grain size distribution and a fine grain size of 18.71 nm.

2. The NPCs annealed under the HR and HT of 100 K/min and 30 min show a modified magnetic domain structure, where no obvious pinning points can be observed, and the domain wall moves easier than that of the NPCs annealed under other annealing conditions, which indicates the reduction of $H_c$.

3. The NPCs annealed under the HR and HT of 100 K/min and 30 min show excellent comprehensive soft magnetic properties, including a low $H_c$ of 10.5 A/m 7.0 (50 mT, 500 kHz), high $\mu_e$ of 64.7 (100 kHz), and low $P_{cv}$ of 751 mW/cm$^3$ (50 mT, 500 kHz).

Author Contributions: Conceptualization, Y.W. and Y.D.; methodology, Y.W., Y.D. and X.J.; validation, Z.L., H.L. and H.Z.; data curation, Y.W. and X.J.; writing—original draft preparation, Y.W.; writing—review and editing, Y.W., Y.D. and M.Y.; supervision, A.H. and J.L.; project administration, Y.W.; and funding acquisition, Y.D. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the S&T Innovation 2025 Major Special Program (Grant No. 2021Z38), Youth Innovation Promotion Association CAS (Grant No. 2021294), Science and Technology Service Network Initiative of the Chinese Academy of Sciences (Grant No. KJFSTS-QYZD-2021-07-002), and the National Natural Science Foundation of China (Grant No. U1809216), Jiangxi Province Science and Technology Cooperation Key Project (No. 20212BDH80007), Science and Technology Major Project of Ganzhou (No. 202101064871).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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