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ARTICLE

Preparation of a Kind of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ Magnetic Powder Core with Optimal DC-bias Property

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Abstract: Fe-Si system metal magnetic powder core was prepared from flaky $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy powders instead of the spherical Fe-6.5%Si alloy powders. Firstly, the preparation of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy powders and $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core was studied, and then the influences of annealing temperature and forming pressure on soft magnetic properties of the magnetic powder core were investigated. The result shows that the permeability (μ_e) of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core has optimal stability of frequency in the frequency range of 1~200 kHz. Residual internal stress produced in the pressing process is fully released after annealing at 450 °C for 1 h, which could significantly improve the permeability (μ_e) of the magnetic powder core, and the maximum value is 53 at forming pressure of 1.74 GPa. With the increase of the applied DC magnetic field, the permeability of the magnetic powder core decreases gradually. When the DC magnetic field ($\boldsymbol{H}$) is 4 kA/m, the percentage permeability of the magnetic powder core remains higher than 70%. As $\boldsymbol{H}$ increases to 10.4 kA/m, the permeability of magnetic powder core is still greater than 20. This shows that $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic power core has optimal DC-bias property.

Key words: $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core; permeability (μ_e); DC-bias property; residual internal stress

Fe-Si system magnetic powder core has excellent overall performances of high effective permeability, low loss, good frequency stability and optimal DC-bias property. As the main vibrating core of inductor filter, choke coil and other electric device, it has been widely used in the field of communications, radar, power switch, pulse transformers, etc, which has been become an important part of the soft magnetic materials[1-4]. With the rapid development of information system, the high performance electronic equipment with miniaturization, lightness, high frequency and good DC-bias property were required. Compared with bulk magnetic materials, Fe-Si system magnetic powder core has low loss at high frequency and significant application advantages. However, the DC superposition characteristic still needs to be further improved, which affects the miniaturization of electronic components. So, it is meaningful to study the DC superposition characteristic of the Fe-Si system magnetic powder core under the condition of huge external applied magnetic fields.

The so-called DC superposition characteristic under the condition of huge external applied magnetic fields refers to the change of the inductance, when the magnetic powder core is under the huge external applied magnetic fields that caused by the large DC signal in the circuit. The $\boldsymbol{H}$ values of the huge external applied magnetic fields have been generally reached 8 kA/m. The research on this field has hitherto been barely available, but the market demand is very large.

At present, few measures are taken to improve the DC superposition characteristic under the condition of huge external applied magnetic fields, which focus on the powder composition modification of Fe-Si system metal powder core. Fe-Si system magnetic powder core which was prepared with the addition of elements such as Al, Ni to the Fe-Si alloys can reduce the high frequency loss and increase comprehensive properties, and many studies have been done about it in recent years[5-7]. And FeSiBPCu amorphous/nanocrystalline magnetic powder core was prepared by Tohoku university's Akihiro

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Makinl, Yan Zhang, Parman and Sharma, China's Kong FanLi et al, and its magnetic properties were investigated[8-11]. But, the research on addition of B, P and Cu to Fe-Si alloys to prepare the crystalline structure of FeSiBPCu alloy powders and its magnetic powder core has been reported less at present.

In this paper, to improve the DC superposition characteristic under the condition of the huge external applied magnetic fields, two measures are to be taken: one is to design and modify the powder composition of Fe-Si system metal magnetic powder core, and prepare $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy powders instead of Fe-6.5%Si alloy powders, in order to improve $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic saturation induction B_s. The other is to use flaky powders instead of spherical powders by spinning water atomization methods. This measure is mainly aimed at improving the permeability (μ_e) of magnetic powder core under the condition of low density.

In this work, $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ master alloy ingots were prepared by medium-frequency induction melting, and the flaky powders were prepared by a vibration ball milling method. The alloy powders were then coated with tetraethyl orthosilicate, and used to prepare the magnetic powder core. The influences of annealing temperature and forming pressure on the soft magnetic properties of magnetic powder core were discussed. The relaxation of the internal stress and DC-bias property under the condition of large applied magnetic fields were also studied.

1 Experiment

$Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ master alloy ingots were prepared by medium-frequency induction melting technique. A mixture of high purity metals (99.9 wt% pure Fe and Cu), and pre-alloyed Fe-Si (99.5 wt%), Fe-B (99.6 wt%) and Fe-P (99.5 wt%) was melted in an nitrogen atmosphere. Then $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy ingots were milled into alloy powders by vibration mill and sieved into different particle size groups: A (75 ~150 μm) and B (45~75 μm). Group A and Group B were mixed according to a certain proportion. The mixed alloy powders were coated with tetraethyl orthosilicate, and then compressed into the toroid core under the pressures of 0.99, 1.24, 1.49 and 1.74 GPa. The heights, inner and outer diameters of the toroid core were 3.5, 9.8, and 20.1 mm, respectively. $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core was subsequently annealed at different temperatures for 1 h under the flowing nitrogen atmosphere, to reduce the internal stress caused by compaction.

The crystalline structure of the alloy powders and the internal stress of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core were analyzed by X-ray diffraction (XRD, PANalytical EMPYREAN) with Cu-Kα and Co-Kα radiation at 40 kV and 40 mA, respectively. The surface morphology and magnetic hysteresis loop of the alloy powders were observed and tested by scanning electron microscopy (SEM) and a vibrating sample magnetometer (VSM, Lake Shore, 7300) in a maximum applied field of 800 kA/m, respectively. The permeability (μ_e) of the magnetic powder core was calculated from the magnetic powder core inductance measured by a Precision LCR meter (TH2816B) and the DC-bias property in the frequency of 200 kHz was measured by a Precision LCR meter (TH2816B) with a DC-bias current source (TH1778). All measurements were performed at room temperature.

2 Results and Discussion

2.1 Powders of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy

Fig.1a and 1b show the SEM images of the alloy powders with different particle sizes. The alloy powders consist of irregular flaky particles with sizes of 75 ~150 μm and 45~75 μm; this is because the particles collide with each other, and impact with extrusion and break into small irregular flaky particles in the process of vibration ball milling.

The XRD patterns of the $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy powders before and after annealing are shown in Fig.2. There is only α-Fe(Si, B, P, Cu) phase in the untreated alloy powders. But the intensity of the α-Fe(Si, B, P, Cu) peak is much stronger for the annealed alloy powders, indicating better crystallinity. Meanwhile, a FeO peak about at 2θ=42° is noticed, which is mainly caused by extra-high annealing temperature, resulting in the oxidation of a small part of alloy powders.

Fig.3 shows the magnetic hysteresis loop of the alloy powders that were smaller than 150 μm measured by VSM (LakeShore 7300). The values of saturation magnetization ($\boldsymbol{M}_s$) and coercive force ($\boldsymbol{H}_c$) are 140 A·m^2/kg and 350 A/m, respectively. This result shows that the alloy powders with

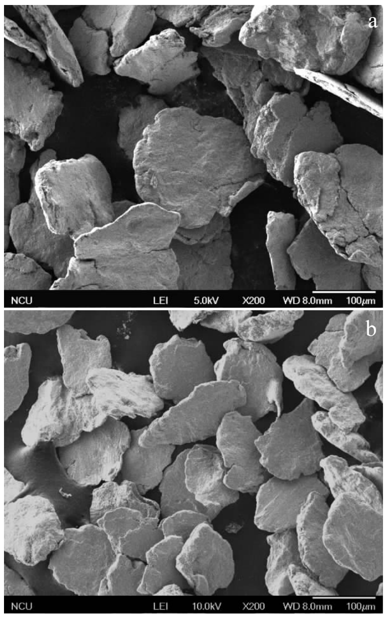

Fig.1 SEM images of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy powders: (a) 75~150 μm and (b) 45~75 μm

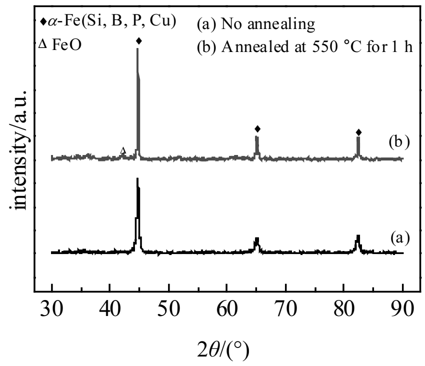

Fig.2 XRD patterns of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy powders

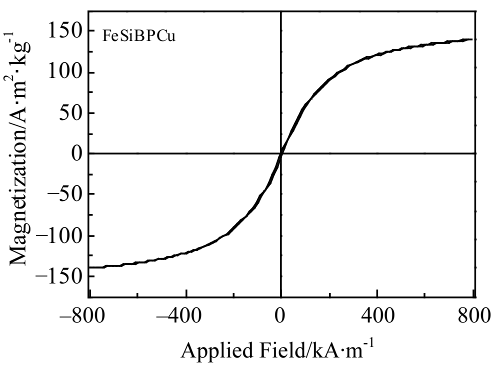

Fig.3 Magnetic hysteresis loop for the $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ alloy powders

good soft magnetic properties can be successfully produced by the medium-frequency induction melting technique.

2.2 Effect of annealing temperature on magnetic properties of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core

Fig.4 shows the effect of annealing temperature on the permeability (μ_e) of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core with forming pressure of 1.49 GPa, and the magnetic powder

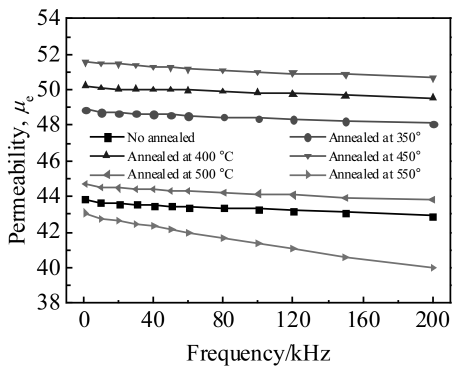

Fig.4 Permeability (μ_e) of the $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core annealed at different temperatures for 1 h

core was annealed under the flowing nitrogen atmosphere for 1 h. The annealing temperature was chosen from 350 °C to 550 °C at 50 °C intervals.

We have noticed that permeability (μ_e) for magnetic powder core arises firstly and then decreases with the increasing annealing temperature, and the best annealing temperature is 450 °C. But when the annealing temperature increases to 500 °C, the permeability (μ_e) for magnetic powder core decreases obviously compared to the annealed treatment at 450 °C. Because annealing treatment can release the internal stress in the pressing process (it can be seen from Fig.5), the structure of magnetic powder core is more uniform, which can promote the movement of the magnetic domain, and thus the permeability (μ_e) can improve[12]. The permeability (μ_e) for magnetic powder core falls and does not have good frequency stability when the annealing temperature continues to rise to 550 °C. It might be that the cladding layer is ablated or carbonized because of the higher annealing temperature, which due to more interspaces are formed between the powder particles. Due to the carbonization of the cladding layer, the magnetic powder core might be oxidized, leading to decrease of the permeability (μ_e) of magnetic powder core[13], which can be seen in Fig.2. In this figure, there is a diffraction peak of FeO.

Therefore, the best annealing temperature of magnetic powder core is 450 °C.

The residual internal stress refers to the balance of the internal elastic stress that still exists in the material without external forces. According to the scope of its function and balance, it can be divided into the macro-stress and micro-stress. While the internal stress of the magnetic powder core is mainly from the pressing process, that is, the macro-stress. Taking d_ψ and $\sin^2\psi$ as ordinate and abscissa axis, respectively, the XRD data are plotted in Fig.5a and 5b.

The d_ψ as a function of $\sin^2\psi$ can be derived from the following equation[14]:

When the crystal plane parallels to the stress direction (ψ= 0°):

$$\varepsilon_n = \frac{d_n - d_0}{d_0} = \frac{-\upsilon}{E}\sigma_\varphi \tag{1}$$

When the crystal plane is perpendicular to the stress direction (ψ=90°):

$$\sigma_\varphi = \frac{d_\sigma - d_0}{d_0} = \frac{\sigma_\varphi}{E} \tag{2}$$

When the included angle between the crystal plane and the stress is ψ:

$$\varepsilon_\psi = \frac{d_\psi - d_0}{d_0} = \varepsilon_\varphi \sin^2\psi + \varepsilon_n \cos^2\psi \tag{3}$$

Considering the strain (ε_ψ) of the grain with ψ angle cooperated the strain (ε_n) of the grain with ψ=0°, according to the equation (1) and (3), it can be obtained:

$$\varepsilon_\psi - \varepsilon_n = (\varepsilon_\varphi - \varepsilon_n)\sin^2\psi = \frac{1+\upsilon}{E}\varepsilon_\varphi \sin^2\psi \tag{4}$$

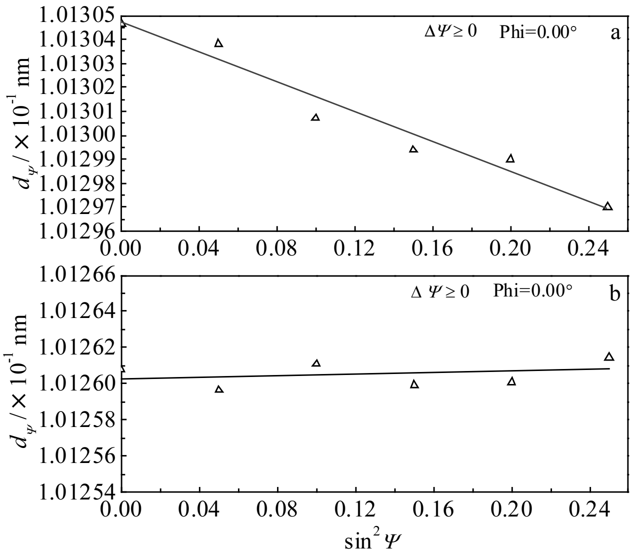

Fig.5 Plots of d_ψ-$\sin^2\psi$ for $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core before (a) and after (b) annealing measured by XRD

$$\varepsilon_\psi - \varepsilon_n = \frac{d_\psi - d_n}{d_0} \approx \frac{d_\psi - d_n}{d_n} \tag{5}$$

According to Eqs. (4) and (5), it can be obtained:

$$\frac{d_\psi - d_n}{d_n} \approx \frac{1+\upsilon}{E}\sigma_\varphi \sin^2\psi \tag{6}$$

where E is the Young's modulus, υ is the Poisson ration, d is the interplanar spacing, ψ is the tilt angle (the included angle between the crystal face normal and the sample surface normal with ψ representation), σ is the internal stress and ε is the internal strain.

Because the values of d_ψ–d_n are far less than d_0, and the difference between d_n and d_0 is very small, d_n can be approximated as the denominator. The interplanar spacing variation is proportional to the $\sin^2\psi$. We can calculate their ratio to measure the stress size.

The magnitude of the residual internal stress can be reflected indirectly by the slope of the fitting linear correlation between d_ψ and $\sin^2\psi$. The larger the slope of the fitting line, the stronger the internal stress.

If the internal stress is equal to zero, the fitting line is a horizontal line. From Fig.5a, the sample without annealing treatment shows a greater slope, which means that the residual stress of the sample is larger. According to Eq.(6), d_ψ is less than d_n that leads to $\frac{d_\psi - d_n}{d_n}$ is less than zero, so the internal stress is negative, that is, the pressure stress. When magnetic powder core was annealed at 450 °C for 1 h, the result of the internal stress measure by XRD is shown Fig.5b. From Fig.5b, we have found the fitting line is basically close to the horizontal line. It shows that the internal stress of magnetic powder core in the pressing process is fully released after annealing at 450 °C for 1 h, and the inner of the sample attain new balance, which can further improve the magnetic properties of the magnetic powder core.

2.3 Effect of forming pressure on magnetic properties of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core

Fig.6 shows the influence of forming pressure on the permeability (μ_e) of the magnetic powder core after annealing at 450 °C for 1 h.

It can be seen that the permeability (μ_e) of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core has optimal stability of frequency in the frequency range of 1~200 kHz. The permeability (μ_e) of magnetic powder core increases from about 43 to 48 with the forming pressure rises from 0.99 GPa to 1.24 GPa. Compared with the permeability (μ_e) under 0.99 GPa, when the forming pressure increases to 1.49 GPa, the permeability (μ_e) of magnetic powder core is about 51, which increases by nearly 20%, and the permeability (μ_e) of magnetic powder core is about 53, with forming pressure up to 1.74 GPa, which increases only by 23%. So we can see that the permeability (μ_e) of the magnetic powder core can be improved significantly with increasing the forming pressure. This is because the interspace among the particles decreases and the filling rate of particles in the magnetic powder core increases with the forming pressure increasing, which increases the density of the magnetic powder core.

But the magnitude of increase of the permeability (μ_e) is limited, when the forming pressure reaches a certain value of 1.49 GPa. Taking into comprehensive consideration the effect of forming pressure on the permeability (μ_e), the mold service life and other factors, we think the forming pressure of magnetic powder core is better in 1.49 GPa.

According to engineering practice application, magnetic core devices often need to work under DC superposition state[15]. DC superposition characteristic is not only an important performance parameter of soft magnetic materials, but also one of the dynamic characteristics of soft magnetic materials. In this paper, the permeability of magnetic powder

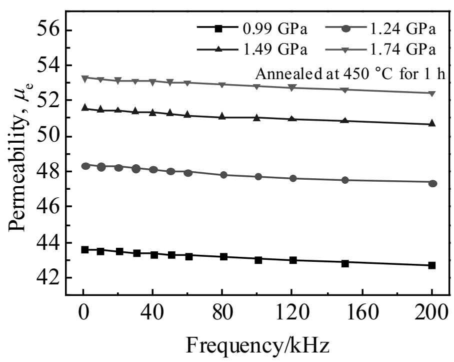

Fig.6 Permeability (μ_e) of the $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core formed at different pressures

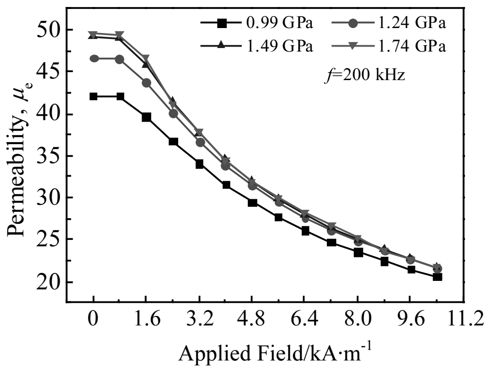

Fig.7 Curves of DC-bias property for $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core

core under the superposition of DC magnetic field is used to directly reflect the current superposition characteristic. The dependence of the permeability on the applied DC-bias field for magnetic powder core is shown in Fig.7.

From Fig.7, the permeability of magnetic powder core decreases gradually with the increase of DC magnetic field. Because the motion of magnetic domain walls and the size of domains become smaller with the increase of DC-bias, the permeability decreases. However, the percentage permeability of magnetic powder core remains higher than 70% when DC magnetic field (***H***) is 4 kA/m. As ***H*** continues to increase to 10.4 kA/m, the permeability decreases by about 55%, but it is still greater than 20. By comparison with Fe-Si metal magnetic powder core, there are two reasons for the improvement in DC superposition characteristic of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core. The first is that the B_s value of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ master alloy powder is significantly larger than that of Fe-6.5%Si powder, which greatly improves the anti-saturation ability of the magnetic powder core. The second is that the flaky alloy powders are used instead of spherical powder by spinning water atomization methods, although the density of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core is not high while the permeability of the magnetic powder core still remains a larger value due to the "bridging" effect of the flake powders. So, the $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic powder core has optimal DC-bias property.

3 Conclusions

1) Flaky powders of $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ can be designed and prepared. The values of saturation magnetization (***M***$_s$) and coercive force (***H***$_c$) are 140 A·m^2/kg and 350 A/m, respectively, which show that the alloy powders has good soft magnetic properties.

2) After annealing at 450 °C for 1 h, the residual internal stress of the $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic power core produced in the pressing process is fully released. At the same time, the permeability (μ_e) of magnetic powder core has optimal stability of frequency in the frequency range of 1~200 kHz, and the value of μ_e is about 53.

3) $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ magnetic power core has optimal DC-bias property after annealing at 450 °C for 1 h. With the increase of the applied DC magnetic field, the permeability (μ_e) of magnetic powder core decreases gradually. When the DC magnetic field (***H***) is 4 kA/m, the percentage permeability of the magnetic powder core remains higher than 70%. As ***H*** increases to 10.4 kA/m, the permeability of the magnetic powder core is still greater than 20.

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一种具有良好直流叠加特性的 $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ 磁粉芯的制备

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摘　要：采用片状 $Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ 合金粉体替代球形 Fe-6.5%Si 合金粉体，制备了 FeSi 系金属磁粉芯。研究了合金粉及磁粉芯的制备，退火温度和成型压力对磁粉芯的软磁性能的影响。结果表明，经 450 ℃/1 h 退火处理后，$Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ 磁粉芯的磁导率在 1~200 kHz 频率范围内具有良好的频率稳定性，磁粉芯内部残余内应力得到充分的释放，其有效磁导率 μ_e 得到大幅的提升，成型压力为 1.74 GPa 时，最大值达到 53。随着外加直流磁场强度的逐渐增大，磁粉芯的有效磁导率逐渐下降，当外加直流磁场强度 $\boldsymbol{H}$=4 kA/m 时，有效磁导率 μ_e 维持在 70%以上，当 $\boldsymbol{H}$ 继续增大到 10.4 kA/m 时，μ_e 仍大于 20，表明磁粉芯具有良好的直流叠加特性。

关键词：$Fe_{95}Si_1B_2P_{0.5}Cu_{1.5}$ 磁粉芯；有效磁导率（μ_e）；直流叠加特性；残余内应力

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