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Novel Fe-based nanocrystalline powder cores with high performance prepared by using industrial materials

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ABSTRACT

The FeSiBPNbCu amorphous magnetic powder with partial crystallization is successfully synthesized using low purity industrial raw materials by gas atomization. The magnetic powder cores (MPCs) are then produced from the mixture of the FeSiBPNbCu powders with 2 wt % epoxy resin as insulation and bonding materials by cold pressing under a compact pressure of 1800 MPa. Evolution of the high-frequency properties for the MPCs with respect to the annealing temperature is systematically studied. The results show that upon annealing at the optimum temperature, the cores exhibit excellent magnetic properties, including high initial permeability of 86 with a high frequency stability up to 10 MHz, high quality factor of 110 at 1 MHz, and low core loss of 1290 mW/cm^3 at 100 kHz for B_m = 0.1 T. It is emphasized that the MPCs prepared by partially crystallized magnetic powders can also achieve lower core losses and higher magnetic permeability, which is of great significance in industrial production.

1. Introduction

The soft magnetic materials of powder cores have been widely used as electromagnetic components to meet the specialized requirements for practical applications [1–4]. As magnetic components, magnetic powder cores (MPCs) have been widely used in noise suppressors, inductor, and other reactors in electromagnetic devices [5–8]. It is known that electronic components are being gradually miniaturized and made highly sensitive, which requires the MPCs to have excellent soft magnetic properties such as high permeability, high saturation magnetization and low core loss [9]. In recent years, Fe-based amorphous and nanocrystalline alloys have been of great interest as they exhibit excellent soft magnetic properties and numerous studies on the fabrication of Fe-based amorphous and nanocrystalline alloy powder cores have been carried out [10,11]. Compared to amorphous powder cores, nanocrystalline powder cores (NPCs) show superior performance at high frequencies and can reduce the audible load/no-load noise significantly as their saturation magnetostriction is nearly zero. However, NPCs are generally fabricated using the flake powders produced by ribbon pulverization, as the conventional nanocrystalline alloys have low glass forming ability (GFA) [12–14]. During the high pressure cold compaction process, the insulation layer at the irregular corners and sharp edges of the flake powder may breakdown, which leads to an increase in core loss and unstable soft magnetic performance [15,16]. In contrast, it is easier to form a uniform insulating coating on the surface of the spherical amorphous powder, which can reduce the core loss of the soft magnetic amorphous composite material. In order to prepare spherical amorphous alloy powder by gas atomization, the alloy must show sufficiently high GFA.

In our previous work, we have successfully developed a novel Fe-based nanocrystalline alloy with the composition of $(Fe_{0.76}Si_{0.09}B_{0.1}P_{0.05})_{98.5}Nb_1Cu_{0.5}$, which exhibits high GFA (d_{max} = 2.5 mm), high B_s (1.46 T) and low H_c (2.8 A/m) [16], and high performance NPCs can be fabricated with spherical powder produced using high-purity materials by gas atomization [17]. However, given the high price of the high purity raw materials, it is not suitable for industrial production and unavoidably limits their industrial applications [18]. In this study, to reduce the production cost and broaden the application field, we tried to prepare the FeSiBPNbCu amorphous powder by gas atomization using low purity industrial raw materials, and the corresponding FeSiBPNbCu MPCs were fabricated by cold compaction. Then, the NPCs were obtained by annealing the MPCs under the appropriate conditions. The magnetic properties of the resulting FeSiBPNbCu NPCs were investigated systematically.

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2. Experiments

Multicomponent alloy ingots with the nominal composition of $(Fe_{0.76}Si_{0.09}B_{0.1}P_{0.05})_{98.5}Nb_1Cu_{0.5}$ were prepared by induction melting mixtures of industrial raw materials of pure Fe (99.6 wt%), Si (99.7 wt %), and Cu (99.9 wt%), premelted Fe-P (98.8 wt%), Fe-B (99.4 wt%), and Fe-Nb (99.3 wt%) ingots in a purity argon atmosphere. Spherical $(Fe_{0.76}Si_{0.09}B_{0.1}P_{0.05})_{98.5}Nb_1Cu_{0.5}$ magnetic powder was prepared by gas atomization. Then the powder was sieved and used to prepare the MPCs with particle sizes below 30 μm. The powder was mixed uniformly with 2 wt% epoxy resin as an organic binder, which was dissolved with acetone in ultrasonic cleaner for 10 min. Subsequently, the obtained amorphous powder was added to the previous solution and stirred continuously with a glass rod until the acetone was volatilized completely. The composite powder was then dried for 30 min in an electric thermostatic drying oven. The composite magnetic powder was then pressed into a toroidal magnetic core with an outer diameter of 20.3 mm, an inner diameter of 12.7 mm, and a thickness of 5.7 mm (Φ 20.3 × φ 12.7 × t 5.7 mm) under a pressure of 1800 MPa at room temperature. The MPCs were annealed at different temperatures for 1 h in vacuum to eliminate the stress generated during pressing and to improve the soft magnetic properties.

The particle size, morphology, and surface composition of the powder were characterized by scanning electron microscopy (SEM). Phase identification of the magnetic powder and MPCs was evaluated by X-ray diffraction (XRD) analysis in the 2θ range from 30° to 90°. The frequency dependences of effective permeability (μ_e) and quality factor (Q) were measured using an impedance analyzer. The DC-bias performance was measured by a wide frequency LCR meter and the magnetic core loss (P_{cv}) was measured by an AC B–H loop analyzer. All the measurements were performed at room temperature.

3. Results and discussion

Fig. 1 shows the X-ray diffraction (XRD) pattern of the FeSiBPNbCu gas-atomized powder. The diffraction pattern consisted of three crystalline peak involving the (110), (200), and (211) lattice planes of the α-Fe(Si) phase. It is worth pointing that there is nearly a halo pattern between 2θ values of 40°–50°, indicating that although it is partially crystallized, the main constituent of the powder is still a glassy phase. The inset in Fig. 1 shows the SEM morphology of the gas-atomized FeSiBPNbCu powder. It was observed that most of the gas-atomized FeSiBPNbCu powders were spherical, and although some were irregular in shape and non-uniform, they had little consequence on the insulating effect. Compared with the powder prepared by crushing the corresponding ribbons, the gas-atomized powder is more conducive to uniform insulation, thereby reducing the eddy current loss between the powders.

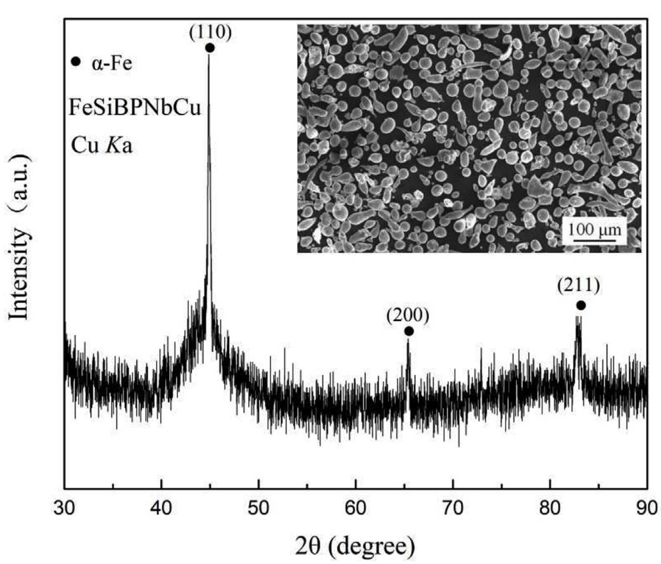

Fig. 1. XRD pattern of the FeSiBPNbCu gas-atomized powder; the inset shows the morphology of the FeSiBPNbCu powder.

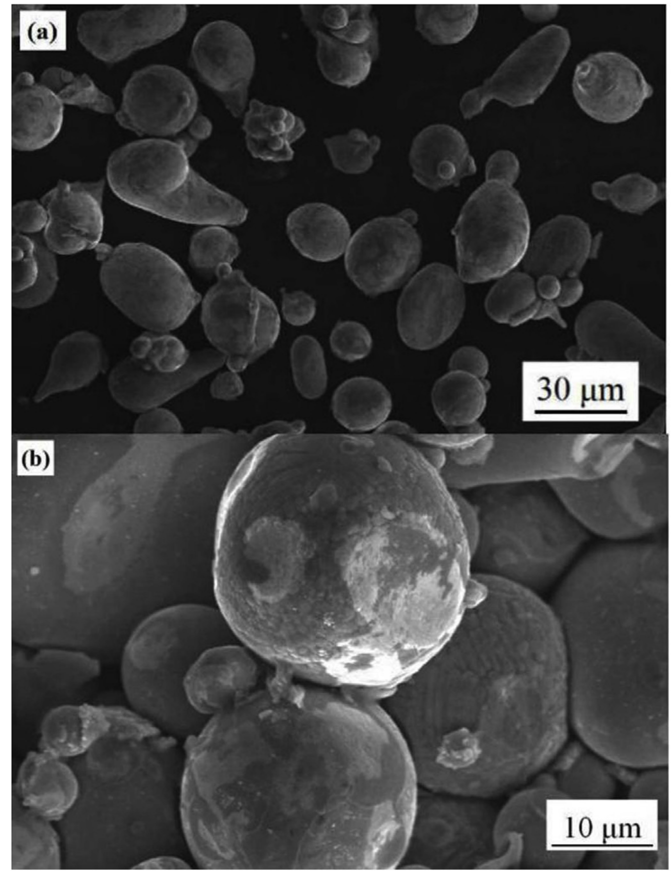

Fig. 2. The SEM image of the morphology of the coated FeSiBPNbCu powder and the cross-section of the magnetic powder core.

In the fabrication of MPCs, the inter-particle insulation is crucial to improve the high-frequency magnetic properties and reduce the core loss of the cores. Fig. 2 shows the SEM image of the morphology of the coated FeSiBPNbCu powder and the cross-section of the MPC. It can be seen from Fig. 2(a) that the surface of the composite powder coated with 2 wt % epoxy resin tends to become rougher and the particle size increases slightly, indicating that a thin coating layer has formed on the surface of the powder. Some voids and gaps can be observed in the cross-section of the MPC, as shown in Fig. 2(b), which will reduce the permeability [19].

The XRD patterns of the FeSiBPNbCu MPCs annealed at different temperatures for 1 h are shown in Fig. 3. Three distinguishable diffraction peaks representing the (110), (200) and (211) lattice planes of the α-Fe(Si) phase are observed for the cores annealed below 520 °C. The XRD patterns exhibit sharp diffraction peaks superimposed on a halo pattern, indicating the coexistence of amorphous and crystalline phase [20]. As the annealing temperature rises to 500 °C, it can be seen that the halo pattern becomes narrower and narrower, indicating that the amorphous phase transition is scarce. When the annealing temperature rises to 520 °C, in addition to the soft magnetic phase of a-Fe (Si), there are also have some diffraction peaks corresponding to $Fe_3(B,P)$ and some unknown phase, which serve as pinning centers retarding the movement of the domain walls, deteriorating the soft magnetic properties of the powder cores [21,22].

Fig. 4 shows the frequency dependence of effective permeability (μ_e)

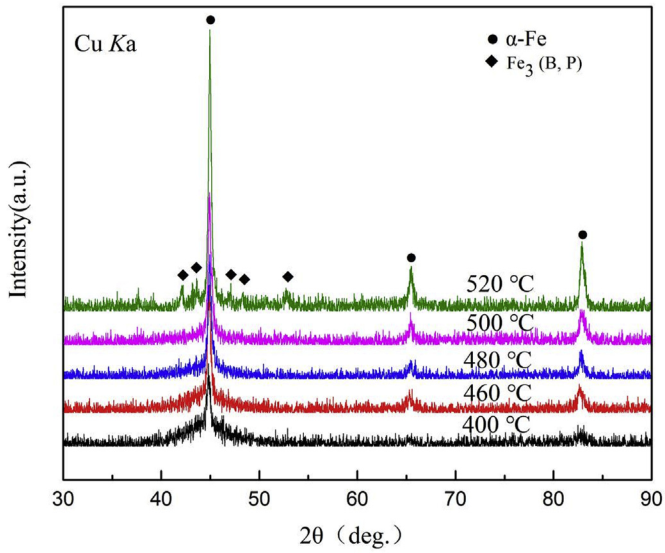

Fig. 3. The XRD patterns of the FeSiBPNbCu MPCs annealed at different temperatures.

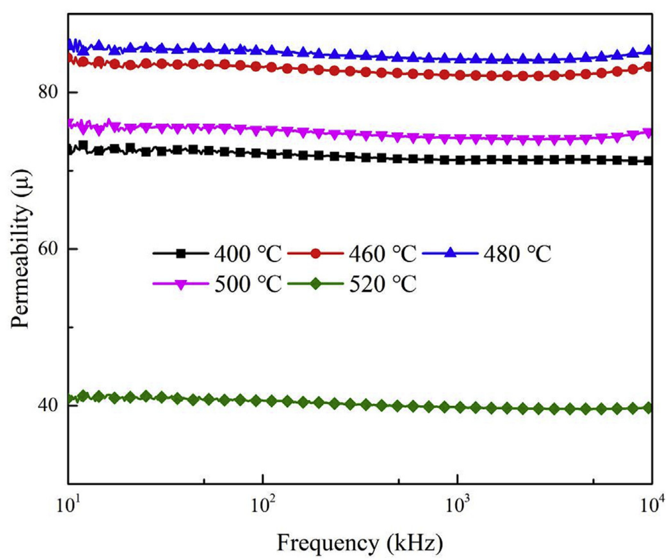

Fig. 4. The frequency dependence of permeability of the FeSiBPNbCu MPCs annealed at different temperatures.

of the FeSiBPNbCu MPCs annealed at different temperatures. It can be seen that μ_e in the frequency range up to 10 MHz is approximately constant, thus the core is suitable to be used as components for electronic systems that require a constant permeability up to the high frequency region. In general, amorphous and nanocrystalline alloys have their optimum annealing temperature range to eliminate the residual stress and get the best magnetic properties; exorbitant temperature and time will make materials to generate new phase or the existing phase to grow bigger, which will worsen the soft magnetic properties [23]. With increasing the annealing temperature from 400 to 480 °C, the permeability at 100 kHz increases from 72 to 86, showing a significant improvement in permeability with an increase of about 20%. When the annealing temperature was further raised to 500 °C, the permeability decreased to 75. As the annealing temperature was increased to 520 °C, the magnetic permeability dropped drastically to 41, significantly deteriorating the performance of the magnetic powder core. For the MPCs annealed at 400 °C, the residual stress cannot be completely removed, so the permeability remains at a low level. For the MPCs annealed at

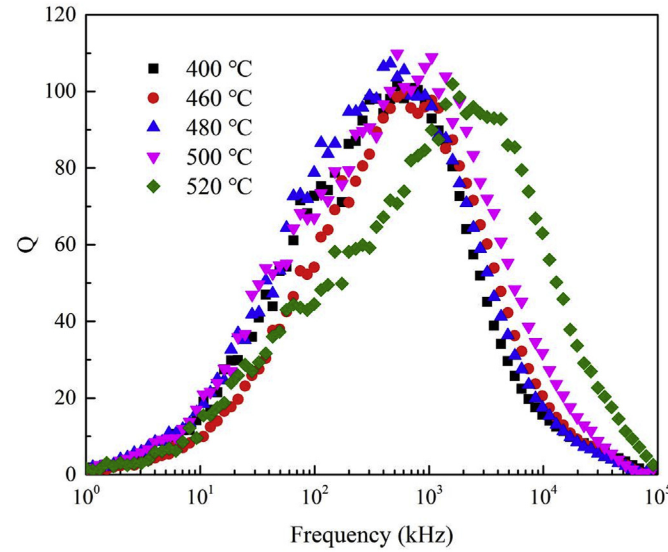

Fig. 5. The variation of the quality factor with frequency for the FeSiBPNbCu MPCs annealed at different temperatures.

520 °C, secondary crystallization occurs and the grains suddenly grow, resulting in the dramatically deterioration of the soft magnetic properties. Annealing at a temperature of 480 °C not only led to elimination of the residual stress, but also provided uniformly distributed nanocrystalline grains, which is beneficial to achieve the excellent soft magnetic properties.

Fig. 5 shows the variation of the quality factor (Q) with frequency for the FeSiBPNbCu MPCs annealed at different temperatures. In physics and engineering applications, Q is a dimensionless parameter that describes the under-damped performance of an oscillator or a resonator [24] and also characterizes a resonator's bandwidth relative to its center frequency [25]. For soft magnetic materials, Q is an important parameter; the higher the Q value, the lower is the energy loss rate. It can be seen that from 100 kHz to 2 MHz, the MPCs annealed between 400 and 500 °C have a high Q value that is larger than 50 and the maximum value of over 110 can be obtained at about 1 MHz, which suggests that the MPCs exhibit excellent performance.

Fig. 6 shows the frequency dependence of total core loss (P_{cv}) for the FeSiBPNbCu MPCs annealed at different temperatures. For the MPCs

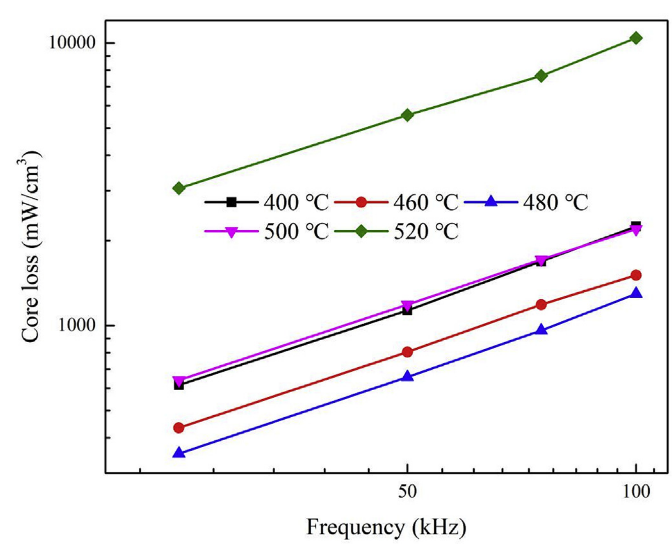

Fig. 6. The frequency dependence of total core loss for the FeSiBPNbCu powder cores annealed at different temperature.

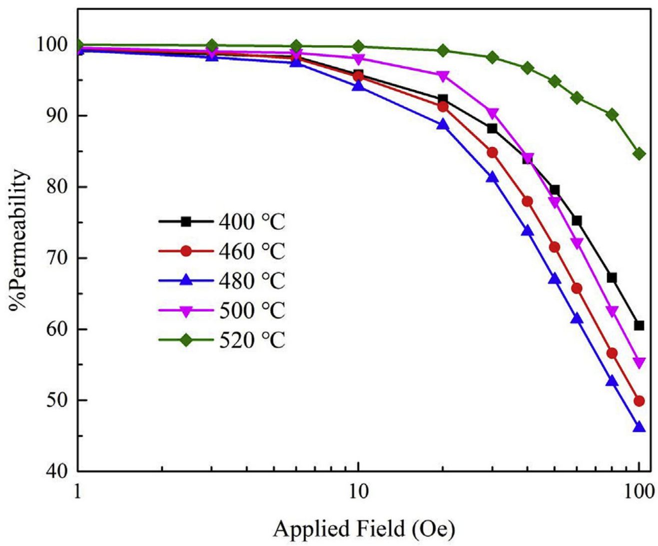

Fig. 7. The DC-bias field dependence of the permeability for the FeSiBPNbCu powder cores annealed at different temperature.

annealed at 400 °C, the P_{cv} is 2060 mW/cm^3 (B_m = 0.1 T, f = 100 kHz). As the annealing temperature increases, the magnetic core loss tends to decrease. The lowest P_{cv} is 1290 mW/cm^3 for the MPCs annealed at 480 °C. With further increasing the annealing temperature to 500 °C, the value of P_{cv} increases to 2190 mW/cm^3. The P_{cv} of the MPCs mainly consists of the hysteresis loss (P_h), eddy current loss (P_e) and residual loss (P_r), which can be expressed by Eq. (1) [26–28]:

$$P_{cv} = P_h + P_e + P_r \quad (1)$$

P_r is a combination of relaxation and resonant losses. It is only important at very low induction levels and very high frequencies and can be ignored in power applications [29]. Hence, P_{cv} of a magnetic device is the sum of P_e and P_h. The P_h value is mainly influenced by the coercive force [7], which is a structure sensitive soft magnetic property and depends on the micro-strain accompanied by structural defects [23]. During the annealing process at higher temperatures, the micro-strain and internal stress can be effectively eliminated, resulting in the reduction of coercivity and the value of P_h. That is why the loss decreases as the annealing temperature increases from 400 to 480 °C. When the annealing temperature is further increased to 500 °C, the insulating layer begins to decompose rapidly, resulting in the increase of coercivity and P_h. In addition the P_e value between the powders also increases, so the total P_{cv} increases correspondingly. However, when the annealing temperature increases to 520 °C, the P_{cv} value experiences a substantial increase to 10420 mW/cm^3. The increase in temperature causes more volatilization of the epoxy resin, and the decomposition of the dielectric layer of the powder is more complete, which give rise to an increase in P_e. As mentioned above, the secondary phase precipitates when the MPCs are annealed at 520 °C as shown in Fig. 3. This retards the movement of the domain walls as pinning centers, resulting in an increase in core loss and poor performance.

Fig. 7 shows the DC-bias field dependence of the permeability, defined by the percentage of the permeability under DC-bias field to the permeability on without DC-bias field, for the annealed FeSiBPNbCu MPCs at 100 kHz [30]. The DC-bias property is important to our materials because almost all the powder cores are used in a DC-bias field. When the applied DC field is small, the attenuation of the magnetic core inductance is not obvious. At this time, the magnetic induction intensity of the powder core has not yet reached saturation. However, when the applied magnetic field continues to increase, there is significant decrease of magnetic core inductance [14,16]. The larger the decrease of μ is, the more the DC-bias of the MPCs finally appears to be poorer. With the increase of annealing temperature, the DC bias characteristics of the magnetic core change significantly. At 100 Oe, the MPC shows superior DC-bias properties of higher than 60% permeability, the reason is that the voids and gaps in the powder cores can pin the domain wall in the magnetizing process and prevent the propagation of domain movement between particles, then suppressing the decrease of permeability, which is beneficial in achieving a higher percent permeability [20]. With an annealing temperature of 480 °C, a lower value of 46% for percent permeability was obtained due to its higher effective permeability. In contrast, the percent permeability of the core annealed at 520 °C was found to be 85%, implying that the Fe-based MPCs are not easily saturated under the applied magnetic field, and can be used to deal with the high output current in electrical power supplies.

4. Conclusions

In this study, the magnetic properties of FeSiBPNbCu powder cores prepared from gas-atomized powders with low-purity raw materials were investigated. When annealed at 480 °C for 1 h, the MPCs exhibited excellent magnetic properties including a high initial permeability of 86 with a high frequency stability up to 10 MHz, high quality factor of 110 at 1 MHz, and low core loss of 1290 mW/cm^3 at 100 kHz for B_m = 0.1 T. The excellent magnetic performance of the NPCs considered in this study is mainly due to the precipitation of the ultrafine α-Fe(Si) phase with a homogeneous grain size distribution in the residual amorphous matrix and presence of the uniform insulation layer on the gas-atomized powder.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.intermet.2018.09.001.

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