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Low core loss combined with high permeability for Fe-based amorphous powder cores produced by gas atomization powders

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Toroid-shaped Fe-based soft magnetic amorphous powder cores were prepared from $Fe_{76}Si_9B_{10}P_5$ (at.%) amorphous powder by gas atomization and subsequent cold pressing with epoxy resin as a binder and insulation. The influence of epoxy resin content and annealing temperature on the magnetic properties of the amorphous soft magnetic powder cores were analyzed. The core loss at a frequency of 100 kHz for the samples with 2 wt% epoxy resin and annealed at 480 °C for 1 h is evaluated to be less than 800 mW/cm^3 under a maximum induction of 100 mT. Meanwhile, the cores also exhibit high permeability of 86 at a frequency up to 20 MHz. These results suggest that Fe-based amorphous powder cores provide great potential for expanding the application of high frequency electronic elements used in rectifiers, inverters, transformers and inductance, which requires soft magnetic cores with high saturation magnetization, high permeability, and low core loss.

1. Introduction

It is well known that Fe-based amorphous materials with excellent soft magnetic properties have been commercially used for distribution transformers and consumer electronics as soft magnetic composites (SMCs) [1–5]. However, the SMCs using Fe-based amorphous alloys are generally produced by using melt-spun ribbons in the limited forms of toroidal wound and stacked type, and they often can only be applied at low and medium frequencies. As the operating frequencies of electronic equipment continue to increase, the SMCs using Fe-based amorphous ribbons cannot meet the current development trend. The SMCs fabricated by ferromagnetic powder particles surrounded by an electrical insulating film may exhibit unique properties including very low eddy current loss, relatively low total core loss at medium and high frequencies, and a prospect of greatly reduced weight and production costs. However, for the amorphous SMCs based on crushed amorphous ribbons, the edges and corners of the crushed powders are hard to be insulated and compacted perfectly, which will result in the increase of core loss [6–9]. Thus, the development of production of spherical amorphous alloy powders is essential.

Recently, several Fe-based bulk amorphous alloys have been developed, and great effort has been devoted to fabricate the SMCs by powder metallurgical process using various consolidation techniques [10–14]. Among them, $Fe_{76}Si_9B_{10}P_5$ (at.%) amorphous alloy possesses high saturation induction density and excellent soft magnetic properties [15–17], which is promising to be used for preparing novel high-performance SMCs. In this work, in order to expand the application of SMCs at high frequencies, we have prepared spherical Fe-based amorphous powders by gas atomization and studied the preparation of SMCs based on the spherical $Fe_{76}Si_9B_{10}P_5$ amorphous powders. The influences of epoxy resin content and annealing temperature on the permeability (μ), DC-bias performance, and the core loss (P_{cv}) for the amorphous SMCs were investigated in details and excellent soft magnetic properties were achieved finally.

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2. Materials and methods

Multicomponent alloy ingots with a nominal composition of $Fe_{76}Si_9B_{10}P_5$ (at.%) were prepared by induction melting the mixtures of pure Fe (99.99 wt %), B (99 wt %) and Si (99.99 wt %), and pre-alloyed Fe-P (99 wt%) alloy in a purified argon atmosphere. The ribbons of $Fe_{76}Si_9B_{10}P_5$ were obtained using melt-spinning method at a linear speed of 40 m/s for copper wheel. Spherical $Fe_{76}Si_9B_{10}P_5$ amorphous powders were prepared by gas atomization. The $Fe_{76}Si_9B_{10}P_5$ ingots were remelted under vacuum in a quartz tube using an induction-heating coil, injected through a nozzle with a diameter of 0.8 mm and atomized by high-pressure Ar gas. Then the obtained powders were screened by standard sieve. The sieved $Fe_{76}Si_9B_{10}P_5$ amorphous powders with particle size below 75 μm were used to prepare amorphous SMCs. The epoxy resin was dissolved in acetone in ultrasonic cleaner for 10 min. Subsequently, the amorphous powders were added to the acetone solution with epoxy resin and continuously stirring with a glass rod until the acetone volatilized completely. The composite powders were then dried in an electric thermostatic drying oven for 30 min. Toroid-shaped $Fe_{76}Si_9B_{10}P_5$ SMCs with an outer diameter of 20.3 mm, an inner diameter of 12.7 mm and a height of 5.3 mm were fabricated by cold pressing under a pressure of 1600 MPa at room temperature. Then the compacted cores were annealed at temperatures in the range from 350 to 510 °C for 1 h in vacuum to improve the soft magnetic properties of the cores. The characteristics of the gas-atomized powder were analyzed by X-ray diffraction (XRD) using Cu Kα-radiation, differential scanning calorimetry (DSC) and scanning electron microscopy (SEM). Permeability (μ) spectra from 1 kHz to 20 MHz were measured by an impedance analyzer (HP 4294A) with twined 20 turns of copper wire tightly around the amorphous SMCs. The DC-bias performance was measured by a wide frequency LCR meter (TH2828A) and the core loss of the SMCs was measured by an AC B–H loop tracer.

3. Results and discussion

The XRD pattern of the $Fe_{76}Si_9B_{10}P_5$ powders with particle size below 75 μm is shown in Fig. 1. As seen in this figure, only a diffuse halo typical for an amorphous phase is seen, and no peak of a crystalline phase is observed. It indicates that a glassy phase without crystallinity is formed in the powders. The inset in Fig. 1 shows the DSC curves of the $Fe_{76}Si_9B_{10}P_5$ powder and ribbon at a heating rate of 0.67 K/s under a flowing Ar atmosphere. The curves show an endothermic peak due to the Curie transition, and a glass transition, followed by an extended supercooled liquid region and almost a single-stage of crystallization process. No appreciable difference in Curie transition (T_c), glass transition temperature (T_g), on-set temperature of crystallization (T_x), supercooled liquid region (ΔT_x), and crystallization process between the melt-spun ribbon and the powders is observed. Thus, these behaviors indicate that the $Fe_{76}Si_9B_{10}P_5$ powders made by gas atomization are completely amorphous.

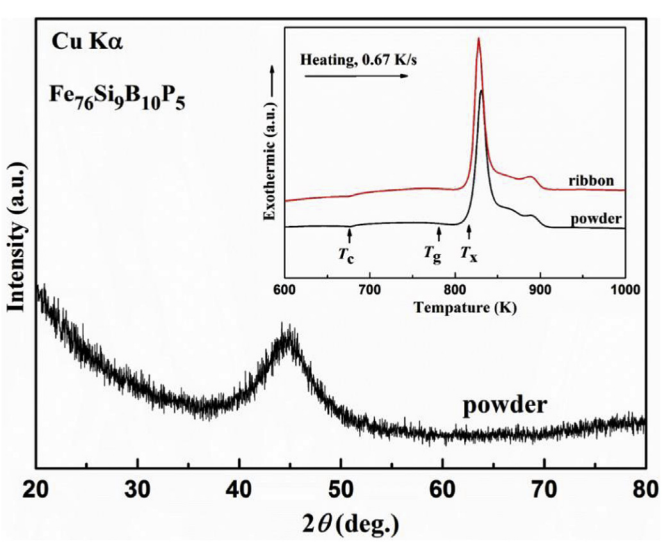

Fig. 1. XRD pattern of the gas-atomized $Fe_{76}Si_9B_{10}P_5$ amorphous powder. The inset shows the DSC curves of the $Fe_{76}Si_9B_{10}P_5$ powder and corresponding ribbon.

Fig. 2(a) shows the morphology of the $Fe_{76}Si_9B_{10}P_5$ amorphous powders. The powders are spherical and no crystalline precipitation feature could be found on the powder surface. Fig. 2(b) shows the surface morphology of the $Fe_{76}Si_9B_{10}P_5$ amorphous powders coated with 2 wt% epoxy resin. Apparently, the epoxy resin has completely and uniformly covered the surface of the $Fe_{76}Si_9B_{10}P_5$ powders, which is the key to get the relatively low eddy current loss of the SMCs. SMC was prepared by pressing the $Fe_{76}Si_9B_{10}P_5$ amorphous powders coated by 2 wt% epoxy resin at a pressure of 1600 MPa, and cross-sectional view of the SMC is shown in Fig. 2(c). Due to the different particle size distribution of the $Fe_{76}Si_9B_{10}P_5$ amorphous powders, the smaller powders are filled in the gap between the larger powders, which will increases the density of the magnetic powder core and benefit the μ of the amorphous SMCs.

Amorphous SMCs with different amounts of epoxy resin in the range of 1–5 wt% were also prepared, and the soft magnetic properties of the SMCs are summarized in Table 1. According to Table 1, with the increase of the epoxy resin content, the saturation magnetization (M_s) of the composites decreased gradually. The presence of the non-magnetic epoxy resin insulating layer reduced the value of M_s dues to the lower weight fraction of the magnetic phases in the SMCs. When the epoxy resin content is 2 wt %, M_s of the SMC still remains at 150 emu/g, which will provide a superior DC-bias property for the amorphous SMC. The μ of the amorphous SMCs decreases from 82 to 55 with increasing the amount of epoxy resin. It is considered that amorphous powders coated by nonmagnetic insulating materials decrease the volume fraction of the amorphous powders in the composites, providing the equivalent of a distributed air gap, and thus decrease μ of the materials. Moreover, the μ of the magnetic powder cores can be defined as equation (1) [18].

$$\mu = \frac{3 + (\mu' - 1)(3 - 3g)}{3 + g(\mu' - 1)} \approx 3/g \tag{1}$$

where μ' is the permeability of the magnetic powders, g is the content of the non-magnetic materials. According to Eq. (1), it is clearly that the μ of the soft magnetic powder cores is inversely related to the content of non-magnetic materials. Consequently, the μ of the $Fe_{76}Si_9B_{10}P_5$ SMCs tends to decline with increasing the amount of epoxy resin. The DC-bias field dependence of the percent permeability for the $Fe_{76}Si_9B_{10}P_5$ SMCs at 100 Oe with different epoxy resin content is also shown in Table 1. The $Fe_{76}Si_9B_{10}P_5$ SMCs show superior DC-bias properties higher than 50% permeability. The higher percent permeability of the $Fe_{76}Si_9B_{10}P_5$ SMCs was achieved in more concentrated insulator, implying that the amorphous SMCs are not easily saturated under low applied fields. It is clearly seen that the core loss exhibit an increase tendency with decreasing the amount of epoxy resin. The decrease of the epoxy resin content means that the thickness of the insulating layer formed on the amorphous surface was thinned and increased the probability of amorphous powders contact, which results in the increase of the eddy current loss and the P_{cv} of the amorphous SMCs.

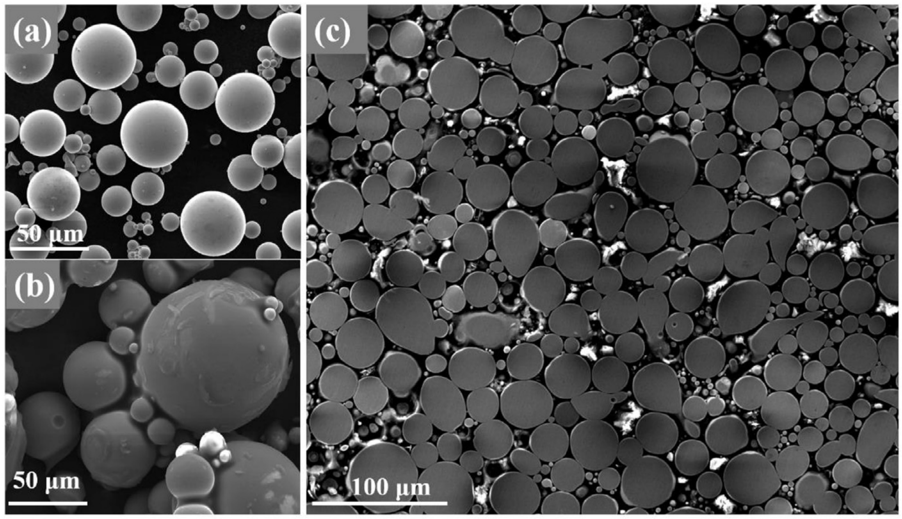

Fig. 2. Morphology of $Fe_{76}Si_9B_{10}P_5$ powder (a) obtained by gas atomization, (b) after coating with 2 wt % epoxy resin and (c) cross-sectional view of the amorphous SMCs with 2 wt % epoxy resin.

Table 1
Properties of amorphous SMCs with different amount of epoxy resin.

Epoxy resin	M_s	Permeability	Core loss (P_{cv}, mW/cm^3)		DC-bias (%)
wt. %	(emu/g)	(μ)	100 kHz/0.05 T	100 kHz/0.1 T	100 Oe
1	156	82	350	1252	53
2	150	73	300	1130	58
3	140	65	250	1041	63
4	133	60	210	850	69
5	127	55	180	800	72

Heat treatment played an important role in improving the soft magnetic properties of the SMCs. Apparently when the epoxy resin content is 2 wt %, the amorphous SMC has relatively high B_s and μ. So the effect of annealing temperature on the magnetic properties and the core loss of amorphous SMC with 2 wt % epoxy resin were further investigated in order to obtain a soft magnetic core with excellent comprehensive performance finally. Fig. 3 shows frequency dependence of the μ for the $Fe_{76}Si_9B_{10}P_5$/2 wt% epoxy resin SMCs after annealing at different temperatures. No obvious decrease in μ of the amorphous SMCs can be found even increasing the frequency up to ~ 20 MHz, which is due to completely insulation between the amorphous powders. The permeability increases from 65 to 86 as the annealing temperature increases from 350 to 480 °C, followed by an abrupt decrease to 39 with further elevated temperature to 510 °C. The heat treatment provided a highly uniform film as well as a low-volume fraction of defects, and reduced the distortion within particles, leading to the increase in the μ of amorphous magnetic powder cores. However, the deterioration in the μ is seen at 510 °C. As seen in the inset of Fig. 3, in addition to soft magnetic phase of a-Fe, the XRD pattern for the core annealed at 510 °C also exhibits some diffraction peaks corresponding to the Fe_3(B,P) and Fe_2B phase, which serve as pinning centers retarding movement of the domain walls, deteriorating the soft magnetic properties of the powder cores [19].

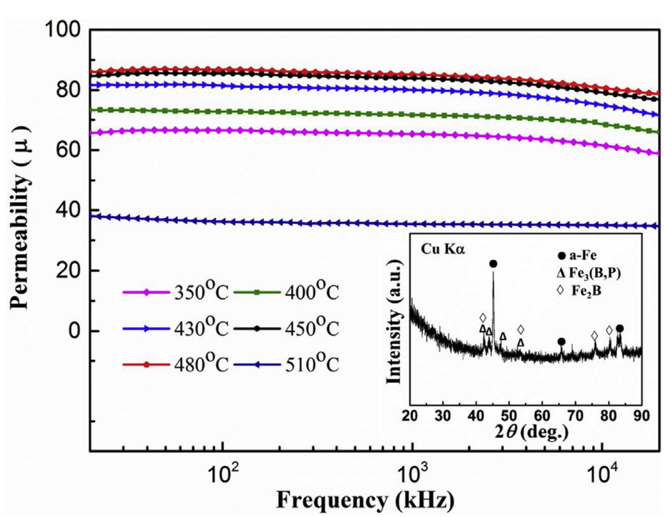

Fig. 3. Effects of the annealing temperature on the permeability of $Fe_{76}Si_9B_{10}P_5$ amorphous soft magnetic cores with 2 wt % epoxy resin. The inset shows the XRD pattern of the $Fe_{76}Si_9B_{10}P_5$ SMCs annealed at 510 °C for 1 h.

Fig. 4 shows the effect of the annealing temperature on the core loss of $Fe_{76}Si_9B_{10}P_5$ SMCs with 2 wt % epoxy resin. The P_{cv} of the amorphous SMCs mainly consists of hysteresis loss (P_h) and eddy current loss (P_e) [19,20]. To investigate the contribution of each component, separation of the total core loss P_{cv} has been carried out using the least square method [21].

$$P_{cv} = P_h + P_e = f \oint HdB + \frac{CB^2 f^2 d^2}{\rho} \quad (2)$$

where f is the frequency of the varying magnetic field, H is magnetic field intensity, C is proportionality constants, B is the flux density, ρ is the electrical resistivity of the ferromagnetic material, and d is

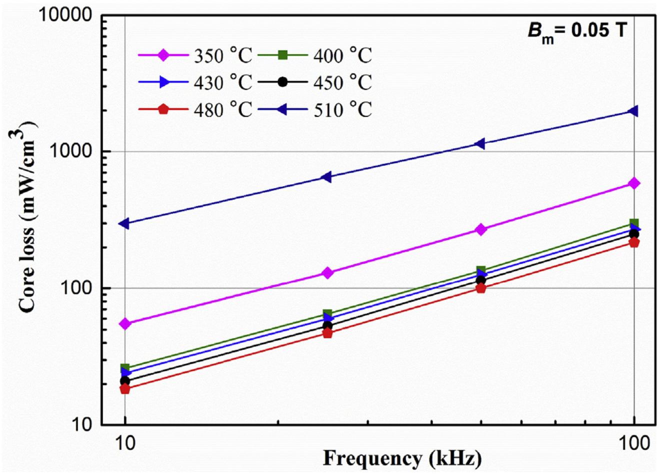

Fig. 4. Effect of the annealing temperature on the core loss of $Fe_{76}Si_9B_{10}P_5$ SMCs with 2 wt % epoxy resin at $B_m = 0.05$ T.

Table 2
Comparison of permeability, core loss and DC-bias between our newly developed amorphous SMCs and the typical SMCs reported previously.

Samples	Permeability	Core loss (P_{cv}, mW/cm^3)		DC-bias (%)
	(μ)	100 kHz/0.05 T	100 kHz/0.1 T	100 Oe
Amorphous SMCs/480 °C	86	200	780	56
Fe-Si-Al [22]	55	–	600	55
Fe-Si-Ni [23]	60	620	–	50
Fe-Si-B-C [9]	54	–	900	82
Fe-Si-B [24]	60		901	65

effective dimension (for the material prepared by casting, d is thickness of the sample, for laminated material d is thickness of the sheet, for SMCs d corresponds to the particle diameter of the insulated soft magnetic powder). At low frequencies the hysteresis loss is the main core loss part and can be reduced by large particle size, higher purity of the soft materials in the particles and stress relieving heat treatment. Eddy current loss is dominant in high-frequency range and can be effectively reduced by appropriate insulation of the soft magnet powders. The P_{cv} in Fig. 4 follows an initial decreasing trend and reaches a minimum of approx. 200 mW/cm^3 at 480 °C ($B_m = 0.05$ T, $f = 100$ kHz). The initial reduction of the P_h is attributed to the stress relaxation and internal defects removal by annealing treatments, which resulted in the decrease of P_{cv} finally. The P_{cv} then increases rapidly to 1820 mW/cm^3 after annealing at 510 °C. Firstly, the P_{cv} zoomed up at 510 °C owing to crystallization of amorphous powder. On the other hand, high treatment temperature caused the degradation of the coating layer and resulted in the growing of P_e between the amorphous powders. Table 2 summarizes the soft magnetic properties of the present amorphous SMC after annealing at 480 °C and the typical SMCs reported previously [9,22–24]. It is seen that the $Fe_{76}Si_9B_{10}P_5$ amorphous SMC fabricated in this work exhibits rather better comprehensive properties than those of the previously reported SMCs. The present $Fe_{76}Si_9B_{10}P_5$ amorphous SMC has a higher permeability of 86, lower core loss of 780 mW/m^3 and better dc-bias property of 56%, which can better meet the development direction of miniaturization and high frequency of electronic components in the near future.

4. Conclusions

Spherical $Fe_{76}Si_9B_{10}P_5$ amorphous powders with particle size below 75 μm were fabricated by gas atomization. The permeability of the corresponding Fe-based amorphous SMCs tends to decline when the amount of the epoxy resin increases, whereas the core loss and dc-bias properties are improved. The amorphous SMCs with 2 wt % epoxy resin show good soft magnetic properties and further optimized by heat treatment. Optimum magnetic performances of the permeability and the core loss are 86, 200 mW/cm^3 ($B_m = 0.05$ T, $f = 100$ kHz) when annealing at 480 °C for 1 h. The $Fe_{76}Si_9B_{10}P_5$ amorphous SMCs with excellent comprehensive properties may be used in the large current application field and contribute to the miniaturization of electrical parts with higher efficiency.

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