Improvement of soft magnetic properties of FeSiBPNb amorphous powder cores by addition of FeSi powder

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Abstract
The effect of FeSi powder with volume fraction of 0–40% on the soft magnetic properties of the FeSiBPNb amorphous magnetic powder cores has been systematically investigated in this study. The results show that the amorphous magnetic powder core mixed with 10% FeSi powders in volume fraction exhibits superior DC-bias properties, constant permeability, low core loss up to a higher frequency range, and excellent soft magnetic properties after appropriate heating treatment. The DC-bias properties of the present amorphous magnetic cores decrease by only 20% as the external field increases to 100 Oe. Meanwhile, it also exhibits a high permeability of 60 at 10 MHz and a low core loss of 159 W/kg at $B_m = 0.1$ T and $f = 50$ kHz. The present Fe-based amorphous magnetic powder cores mixed FeSi powders with superior DC-bias properties are a potential candidate for a variety of industrial applications.

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1. Introduction

Magnetic powder cores (MPCs) are widely used as components for electric and electronic systems, such as switching power supplies, various DC/DC converter circuits, active filters, choke coils, etc. [1–7]. These types of equipment are making significant progress, and their power consumption keeps increasing. With the development of the electric power industry, electronics are developing towards miniaturization, high frequency, and high current. Following this trend, the supply of magnetic powder cores with high permeability, low loss, and excellent performance of DC-bias at high frequencies has become a key issue that accompanies this progress [8]. Fe-based amorphous alloys are considered to be ideal materials for producing magnetic powder cores due to their excellent magnetic properties, such as low coercivity, high resistance and superior good DC-bias performance. However, compared with Fe and Fe–Si magnetic cores, Fe-based amorphous magnetic powder cores have a lower saturation magnetization ($B_s$), resulting in them not being able to realize high magnetic flux density and excellent magnetic softness simultaneously. Before the emergence of core materials with excellent soft magnetic properties, it is very difficult to improve magnetic properties of the cores using traditional methods. Meanwhile, it is inevitable to develop new methods to improve magnetic properties, especially saturation magnetization ($B_s$) of the cores. In previous studies, the authors have developed a new Fe-based bulk glassy magnetic alloy (Fe0.76Si–0.09B0.1P0.05)99Nb1, which exhibits relatively high $B_s$ (1.4 T) and excellent soft magnetic properties [9]. We have fabricated amorphous powder cores by water atomization of (Fe0.76Si–0.09B0.1P0.05)99Nb1 powders mixed with organic and inorganic binders under the condition of cold pressing [10]. To further improve the performance of the (Fe0.76Si–0.09B0.1P0.05)99Nb1 amorphous magnetic powder cores, we intend to mix the amorphous powders with different volume fractions (0%–40%) of FeSi powders, thus amorphous (Fe0.76Si–0.09B0.1P0.05)99Nb1/FeSi compound powder cores could be fabricated and the high-frequency magnetic properties of the resulted cores are studied in the present study.

2. Experimental details

Multicomponent alloy ingot with a nominal composition of (Fe0.76Si–0.09B0.1P0.05)99Nb1 was prepared by induction melting...
mixtures of industrial Fe and Si and Fe-B, Fe-P, Fe-Nb pre-alloys. Metallic glassy powders were prepared by water atomization with a water pressure of 60 MPa. The (Fe0.76Si0.09B0.1P0.05)99Nb1 amorphous powders with the size below 75 μm and FeSi powders with the size below 10 μm were selected for the compaction of magnetic powder cores.

The (Fe0.76Si0.09B0.1P0.05)99Nb1 amorphous alloy powders mixed with volume fractions of 0–40% of FeSi powders were passivated with 2 wt% phosphating solution to form a suitable inorganic oxide layer for insulation on the surface of powders, and then 1 wt% of NH2CH2CH2CH2Si(OC2H5)3 KH550 as a coupling agent, 2 wt% of epoxy resin as an organic binder were added and uniformly mixed with the powders at room temperature. The mixture then was dried at 353 K for 1 h within a vacuum dryer. Finally, the toroidal powder cores with dimensions of 27.0 mm in outer diameter, 14.8 mm in inner diameter and 6.50 mm in thickness (Φ 27.0 × Φ 14.8 × 6.50 mm) were prepared through the compaction of the powder mixture by cold pressing under a pressure of 2000 MPa at room temperature.

The saturation magnetization of those powders were measured by vibrating sample magnetometer (VSM, Lakeshore 7410). The characteristics of the water-atomized powders were analyzed by X-ray diffraction (XRD, Bruker D8 Advance) with Cu Kα radiation and scanning electron microscopy (SEM, EVO 18, Carl Zeiss), and thermal stability associated with glass transition temperature (Tg), crystallization temperature (Tc), the supercooled liquid region (ΔTc = Tc − Tg) was examined by differential scanning calorimetry (DSC, NETZSCH 404C) at a heating rate of 0.67 K/s in an Ar atmosphere. The effective permeability of the compacted cores were calculated from core inductance measured by the Agilent 4294A Impedance Analyzer, the DC-bias field performance was measured by Agilent 4980 A, and the magnetic core loss was measured by an AC B-H loop analyzer (Riken ACBH-100K). All of the compacted cores were annealed at 673 K for 1 h to release the residual stress caused by the compaction and improve the magnetic properties. All the measurements were performed at room temperature.

3. Results and discussion

Fig. 1 shows the typical DSC curve of water-atomized (Fe0.76Si0.09B0.1P0.05)99Nb1 powders with a particle size below 75 μm. The DSC curve exhibits a small endothermic peak due to the Curie transition, the glass transition, followed by an extended supercooled liquid region, and almost a single stage of the crystallization process, indicating simultaneous precipitation of many kinds of crystalline phases, which is in agreement with that of the corresponding glassy alloy ribbon [9]. The Curie temperature (Tc), the glass transition temperature (Tg), and the onset crystallization temperature (Tc) of the water-atomized (Fe0.76Si0.09B0.1P0.05)99Nb1 powders determined from the DSC curve are 627 K, 773 K, and 833 K, respectively. The shape is due to the surface tension of liquid alloy, the different interparticle insulation is a key factor for reducing the eddy current in the magnetic powders cores working at high frequency. In this study, we attempted to synthesize an oxide layer for insulation on the surface of the amorphous and FeSi powder by the passivation [11,12]. The interparticle insulation observed by SEM of (a) water-atomized (Fe0.76Si0.09B0.1P0.05)99Nb1 powders and (b) mixture of amorphous powder with 10% volume fraction of gas-atomized FeSi powders after passivation. The average particle size of (Fe0.76Si0.09B0.1P0.05)99Nb1 powders was approximately 50 μm and that of FeSi powders was about 5 μm. The amorphous powders were rather flat with round edges, suitable for forming consolidated powder cores with high density. The shape is due to the surface tension of liquid alloy, the different disintegrating and cooling conditions. All these results indicate clearly that the particles of the water atomization powders almost had a fully glassy structure. Additionally, most of the gas-atomized FeSi powders were in spherical form. When the two kinds of powders were mixed, the smaller FeSi powders will enter the gap between (Fe0.76Si0.09B0.1P0.05)99Nb1 amorphous powders, which is suitable for forming consolidated powder cores with high density.

Fig. 3 shows the hysteresis loops of the water-atomized (Fe0.76Si0.09B0.1P0.05)99Nb1 amorphous powders with the size below 75 μm, the gas-atomized FeSi powders with the size below 10 μm, and the mixed powders with different FeSi content measured by VSM. The gas-atomized FeSi powders show good magnetic properties with saturation magnetization (Ms) of 200 emu/g, and the water-atomized (Fe0.76Si0.09B0.1P0.05)99Nb1 powders.
amorphous powders exhibit $M_s$ of 140 emu/g. As expected, a higher Fe-Si content will lead to a larger $M_s$ in mixed powders, the $M_s$ of the mixed powders increases to about 146 emu/g, with further increasing the FeSi contents to 30%, the $M_s$ of the mixed powders increases to about 156 emu/g. This demonstrates that FeSi powders addition can raise the $M_s$ of magnetic powder cores. Here, it can be emphasized that FeSi powders with high $M_s$ are expected to be markedly available for downsizing of choking coils or transformers operating at high frequency and high power.

Fig. 4 shows the aspect of the toroidal shape surface of powder core ($\Phi$ 27.0 × $\Phi$ 14.8 × t 6.50 mm) fabricated by cold compressing the mixture of (Fe$_{0.76}$Si$_{0.09}$B$_{0.1}$P$_{0.05}$)$_{99}$Nb$_{1}$ amorphous powders and FeSi powders under a pressure of 2000 MPa for 5 s at room temperature. The density of the core increases from 5.3 g/cm$^3$ to 6.2 g/cm$^3$ with increasing the volume fraction of FeSi powder from 10% to 40%. The powder cores are annealed at 673 K for 1 h to improve the soft magnetic properties. Fig. 5 shows the frequency dependence of the effective permeability ($\mu_e$) of the cores. In general, the magnetic cores used as components for the electronic system are often required to have a constant permeability up to the high-frequency region. It can be seen that the $\mu_e$ keeps almost a constant value in the frequency range of up to 10 MHz when the volume fraction of FeSi powders is no more than 20%, while a larger content of Fe-Si powders results in the significant drop of $\mu_e$ with increasing frequency. Meanwhile, the $\mu_e$ has a tendency to increase with increasing the volume fraction of FeSi powders from 0% to 20%, and the $\mu_e$ of the cores are 60 and 67 for 10% and 20% volume fractions of FeSi powders, respectively. Although the high content of FeSi powders elevates the permeability of the cores at low frequency, those mixed with a 30% or 40% volume fraction of FeSi powders shows a plummeting tendency with the increase of frequency. This behaviour is ascribed to the effect of ferromagnetic resonance occurring [13]. The $\mu_e$ of the cores has a tendency to increase with an increasing content of the FeSi powder at lower frequency, and the initial permeability of the cores mixed with 30% and 40% volume fractions of FeSi powders are the highest than those of the cores with a less volume fraction of FeSi powders. The core with no addition of FeSi powders exhibits a lower permeability of 56, whereas the effective permeability of the cores with a volume fraction of 40% FeSi powders is up to 71. It is considered that the reduction of $\mu_e$ may be due to the decrease of the core density. These powders insulating materials contain inorganic, organic top coating, and a gap between amorphous powders decreases the volume fraction of the amorphous powder fillers in the composite, provides the equivalent of a distributed air gap, and thus decrease the $\mu_e$ of materials with respect to powder cores [14,15]. As we know, the permeability of the magnetic core can be calculated by:

$$\mu = \frac{3 + (\varphi - 1)(3 - 3g)}{3 + g(\varphi - 1)};$$  (1)

where $\varphi$ is the permeability of the magnetic powders, $g$ is the content of the non-magnetic materials [16]. From equation (1), one can infer that the permeability of the magnetic cores increases with the increasing content of FeSi, which has a higher $M_s$. By contrast, the smaller FeSi powders fill the gap between amorphous powders and thus improve the density of the magnetic cores. Therefore, the $M_s$ also reaps the benefit to some extent. The core is of high advantage to be used as components for electronic system, which requires a constant permeability up to the high frequency region. Fig. 6 shows the frequency dependence of the core loss ($P_{c/v}$) of the magnetic cores fabricated from (Fe$_{0.76}$Si$_{0.09}$B$_{0.1}$P$_{0.05}$)$_{99}$Nb$_{1}$

Fig. 3. Hysteresis loops of the (Fe$_{0.76}$Si$_{0.09}$B$_{0.1}$P$_{0.05}$)$_{99}$Nb$_{1}$/FeSi composite powders with different FeSi contents.

Fig. 4. The aspect of the toroidal shape surface of the (Fe$_{0.76}$Si$_{0.09}$B$_{0.1}$P$_{0.05}$)$_{99}$Nb$_{1}$ compound with FeSi amorphous powder cores.

Fig. 5. Frequency dependence of the effective permeability for the (Fe$_{0.76}$Si$_{0.09}$B$_{0.1}$P$_{0.05}$)$_{99}$Nb$_{1}$ amorphous powder mixed with different contents of FeSi powder cores.
amorphous powders mixed with different volume fractions of FeSi powders for the maximum induction ($B_m$) of 50–125 mT at 50 kHz. Generally, $P_{cv}$ includes hysteresis loss ($P_h$), eddy current loss ($P_e$) and residual loss ($P_r$). The $P_r$ is a combination of relaxation and resonant losses of the system, which is important only at very low induction levels and very high frequencies and can be ignored in power applications. Then, the core loss of MPCs can be expressed by Eq. (2) [17,18]:

$$P_{cv} = P_h + P_e$$ (2)

The eddy current loss is dominant in the high-frequency range and can be dramatically reduced by the addition of appropriate insulation in the magnet powders. It is also proportional to the particle size when the shape of particles is spherical [19]. As can be seen in Fig. 6, the core loss increases greatly with the maximum induction ($B_m$). Additionally, the core loss at 50 kHz for $B_m = 0.1$ T of the core mixed with 10% volume fraction of FeSi powders exhibits the lowest core loss of 159 W/kg. It is well known that the smaller FeSi powders in the magnetic powder cores reduces the hysteresis loss ($P_h$) by filling the gap between larger amorphous powders more than the smaller size FeSi powders, resulting in a higher eddy current loss ($P_e$) [20], so the influence of particle size on $P_{cv}$ is much greater than that of low resistivity for the smaller FeSi powders. Meanwhile, separating the magnet powders electrically from each other, which provide a relatively high resistivity and can effectively reduce the $P_r$ results in the improvement of high frequency characteristics of the cores. The ultra-low core loss of the amorphous powder cores is attributed to the combination of relatively high resistivity and the amorphous structure.

The DC-bias field dependence of the percent permeability of the cores with different FeSi powder contents at 100 kHz, which is defined by the percentage of the permeability upon the DC-bias field to the permeability in a no-DC-bias field [14], is shown in Fig. 7. These composite powder cores show permeability change performance over the bias field range higher than 100 Oe. They exhibit superior DC-bias properties higher than 75% permeability at $H = 100$ Oe. As the $B_m$ of the composite powder cores increased by composing an appropriate mass fraction of FeSi powders, the DC-bias performance can be improved. The permeability of the present powder cores was kept stable, implying that the (Fe$_{0.76}$Si$_{0.09}$B$_{0.1}$P$_{0.05}$)$_{99}$Nb$_1$ amorphous powder cores mixed with FeSi powders are not easily saturated under low applied fields and might be applied to process the high output current in the power supply.

4. Conclusions

(Fe$_{0.76}$Si$_{0.09}$B$_{0.1}$P$_{0.05}$)$_{99}$Nb$_1$ glassy alloy powders below 75 µm were successfully prepared by water atomization and FeSi powders below 10 µm were done by gas atomization. The Fe-based magnetic cores were prepared by cold compressing the mixture of the two kinds of magnetic powders with binder at room temperature. The cores were annealed for 1 h under 673 K. The resulting amorphous magnetic core mixed with a volume fraction of 10% FeSi powders exhibits superior magnetic properties, including high permeability of 60 up to 10 MHz, superior DC-bias properties higher than 80% of percent permeability at an external field of 100 Oe and low core loss of approximately 159 W/kg at 50 kHz for $B_m = 0.1$ T. The synthesis of the Fe-based composite powder cores with good magnetic properties is encouraging for future applications as functional materials.

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