High permeability and low loss of Ni-Zn-Fe ferrite/metal composite cores in high frequency region

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High permeability and low loss of Ni-Zn-Fe ferrite/metal composite cores in high frequency region

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A Ni-Zn-Fe ferrite was deposited on the surface of Fe-Si and three kinds of Fe-Si-Cr powders as an insulating material using an ultrasonic-enhanced ferrite plating method. All ferrite-coated powders exhibited a high magnetization of about 180 emu/g derived from their soft magnetic metal and ferrite composition. The three different ferrite-coated Fe-Si-Cr cores annealed at 973 K exhibited a constant permeability $\mu'$ of 55–60 in the frequency range up to 50 MHz owing to the insulating effect of the ferrite layer. In contrast, the permeability $\mu'$ of the ferrite-coated Fe-3.5Si mass % core decreased in the MHz range. The Cr component acted as a passive layer that made it possible to maintain the Ni-Zn-Fe ferrite layer after annealing. The core loss of the ferrite-coated Fe-3.5Si-4.5Cr mass % core was 448 kW/m$^3$ at 100 kHz and 50 mT, which was half of that observed for the Fe-3.5Si-4.5Cr core without ferrite layer. Additionally, the eddy-current loss of the ferrite-coated Fe-3.5Si-4.5Cr core was strongly decreased compared with the non-coated Fe-3.5Si-4.5Cr core owing to the insulating properties of the ferrite layer. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). [http://dx.doi.org/10.1063/1.4976945]

INTRODUCTION

Recently, soft magnetic materials with high saturation magnetization and low total core loss ($P_{cv}$), which are used in devices such as coils, motors, and transformers, have been improved their magnetic properties in high frequency region to meet demands for downsizing, and high-frequency applications. Magnetic powder cores consisting of magnetic metal powders coated with electrically insulating materials are one particularly attractive class of materials expected to meet these demands.1,2 Powders are easily shaped into complex structures by press shaping.3,4 The use of insulating materials in their coatings could reduce eddy current loss ($P_e$) in the high frequency region. However, insulating layers such as resin generally do not have a magnetic moment and so cause a decrease in magnetization and permeability per unit volume.5 Furthermore, cores are typically annealed at high temperature to reduce their hysteresis loss ($P_h$) due to residual stress. An insulating layer such as resin may be damaged by such annealing processes, which would lead to an increase in $P_h$. Therefore, many efforts have been made to improve the magnetic properties of powder cores using more appropriate insulating materials such as epoxy, silicon, phosphate, SiO$_2$, and Al$_2$O$_3$.6–8

In this work, a Ni-Zn-Fe ferrite layer was used as an insulating material because of its high resistivity, thermal stability, and soft magnetic properties.9 The surface of a Fe-based metal

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powder was covered with a Ni-Zn-Fe ferrite layer using an ultrasonic-enhanced ferrite plating method. This technique can easily fabricate ferrite layers on the surface of various types of particle, including metals, silica, and polymer powders. Additionally, a Ni-Zn-Fe ferrite layer was deposited on Fe-Si and Fe-Si-Cr metal powders to create high magnetization, high permeability, and low core-shell powders. These materials were expected to exhibit not only magnetic properties but also thermal stability derived from the ferrite layer.

**EXPERIMENTAL PROCEDURE**

Fe-3.5Si mass % powder and three kinds of Fe-Si-Cr powder (Fe-3.5Si-4.5Cr, Fe-3.0Si-6.5Cr, and Fe-5.5Si-4.0Cr mass %) were used in this experiment. The Fe-3.5Si-4.5Cr powder exhibited the highest magnetization of the three Fe-Si-Cr powders, while Fe-3.0Si-6.5Cr (Cr rich) and Fe-5.5Si-4.0Cr (Si rich) were expected to have high stability against rusting or corrosion. The average particle size of the Fe-3.5Si powder and three Fe-Si-Cr powders was 10 μm. The powders were dispersed in a buffer solution maintained at 343 K and pH = 11 under bubbling N₂ gas. The pH of the buffer solution was adjusted with KOH solution. A reaction solution containing FeCl₂, NiCl₂, and ZnCl₂ and an oxidation solution containing KNO₂ were mixed in the buffer solution under powerful sonication (600 W, 19.5 kHz) for 25 min. After the reaction, the ferrite-coated powders were dried and sieved.

Organic binder decomposed to carbon after annealing at 773 K, which decreased the strength of the cores. Therefore, the ferrite-coated and non-coated powders were mixed with as little as 0.3 wt. % of water glass as a heat stable binder. The powders were compacted at room temperature into toroidal (ϕ8 mm × ϕ4.5 mm × 1 mm) and cylindrical (ϕ8 mm × 1 mm)-shaped specimens for permeability and resistivity measurements, respectively. The non-coated powders were compacted at 800 MPa and the ferrite-coated powders were compacted at 1600 MPa to obtain a density greater than 6.0 g/cm³ because the density was affected by the surface morphology of the powders. The specimens were annealed at 773 K and 973 K in an Ar atmosphere for 15 min.

The structure and crystal features of the powder samples were observed and analyzed by field emission scanning microscopy (FE-SEM), transmission electron microscopy (TEM), and X-ray diffraction (XRD) analysis. The chemical composition of the powders was analyzed by X-ray photoelectron spectroscopy (XPS). The resistivity (ρ) of the cylindrical specimens was measured by a 4-point method. Magnetization (M) was measured with a vibrating sample magnetometer (VSM). Permeability μ ′ was measured with an impedance analyzer at 1 MHz to 1 GHz. The core loss P_cv was measured with a B-H analyzer at 50 kHz to 100 kHz under a magnetization of 50 mT. The values of P_h and the P_e were calculated from the equation below and the obtained P_cv data. Here, the residual loss (P_r) was approximated as zero because of low ferrite volume fraction as described later.\(^1\)

\[
P_{cv} = P_h + P_e, \quad (1)
\]

\[
P_h = f \oint H \, dB, \quad (2)
\]

\[
P_e = \frac{C B_m^2 f^2 d^2}{\rho}, \quad (3)
\]

where f is the frequency, \(H\) is the magnetic field strength, \(B\) is the magnetic induction, \(C\) is the proportionality constant, \(B_m\) is the flux density, \(d\) is the thickness of the material, and \(\rho\) is the resistivity.\(^1\)

**RESULTS AND DISCUSSION**

Figure 1(a)–(h) shows the surface morphology of the non-coated (a)–(d) and ferrite-coated (e)–(h) Fe-Si and Fe-Si-Cr powders observed by FE-SEM. And Figure 1(i) shows the cross-sectional...
image of Fe-Si powder observed by TEM. Ferrite nanoparticles of 10 nm to 100 nm covered the whole surface of the coated powders. The thickness of the ferrite layer was about 50–100 nm, therefore the volume fraction of ferrite was estimated to be about 4 vol. %.

Figure 2 shows X-ray diffraction (XRD) patterns of the ferrite-coated Fe-Si and ferrite-coated Fe-Si-Cr powders. Metallic and spinel phases derived from the metal core and deposited ferrite layer were observed in all of the patterns.

Table I shows the magnetization of the powders and the density of the compacted toroidal cores. Both the ferrite-coated Fe-Si and ferrite-coated Fe-Si-Cr powders exhibited a high magnetization because of their ferromagnetic ferrite layer. The Fe-3.5Si-4.5Cr powder that contained 4 vol. % of paramagnetic water glass exhibited a magnetization of only 176 emu/g.

Figure 3 shows the frequency dependence of the permeability $\mu'$ and $\mu''$ of the compacted cores before and after annealing at 773 K [Fig. 3(a) and (b)], and after annealing at 973 K [Fig. 3(c) and (d)]. In Fig. 3(a), the ferrite-coated Fe-3.5Si-4.5Cr cores exhibited a constant permeability $\mu'$ from 1 MHz to 50 MHz before and after annealing. This indicated that the ferrite layer on the surface of the coated powder worked as an insulating layer. In contrast, the permeability $\mu'$ of the ferrite-coated Fe-3.5Si cores started to decrease at 10 MHz after annealing. After this result, high
temperature annealing (973 K) of the ferrite-coated Fe-Si-Cr cores was attempted to determine their heat stability.

The three different types of ferrite-coated Fe-Si-Cr cores were annealed at 973 K for 15 min [Fig. 3(c) and (d)]. After the annealing, all three samples exhibited a constant permeability $\mu'$ of 55–60 in the frequency range up to 50 MHz, which was higher than that observed after annealing at 773 K [Fig. 3(a)]. These results indicated that the cores had a high heat stability, which is a desirable property for core materials.

Table II shows the electrical resistivity of the ferrite-coated and non-coated cores before and after annealing at 773 K for 15 min. The resistivity of the non-coated cores was higher than that of the bulk metal because of the insulating effect of the water glass. For the non-annealed samples, the ferrite-coated cores had a higher resistivity than the non-coated cores because the ferrite layer acted as an insulating layer. After annealing, the resistivity of both the non-coated cores and ferrite-coated Fe-3.5Si cores decreased, whereas the ferrite-coated Fe-Si-Cr cores maintained a high resistivity.

From the results of Fig. 3 and Table II, it was thought that the Cr component of the core metal and the existence of the ferrite layer were important factors for maintaining a high permeability $\mu'$ in the MHz range. Cr containing metals can form a passive layer ($\text{Cr}_2\text{O}_3$) of several nanometers in thickness on their surface that can protect the underlying metal from chemical reactions such as rusting or corrosion. Figure 4 shows the Cr 2p XPS spectrum of the Fe-3.0Si-6.5Cr powder. Chromium oxide ($\text{Cr}_2\text{O}_3$) was detected on the surface of the Fe-3.0Si-6.5Cr powder. It can be assumed that the passive layer on the Fe-Si-Cr metal surface prevented diffusion between the Ni-Zn-Fe ferrite layer and the metal surface during the annealing step.

Table III shows the core loss ($P_{cv}$) properties of the non-coated and ferrite-coated Fe-3.5Si-4.5Cr cores measured at 50 mT in the 50–100 kHz range. The $P_{cv}$ of the ferrite-coated Fe-3.5Si-4.5Cr cores was particularly small, only half that of the non-coated core at 100 kHz. Moreover, the $P_e$ of the ferrite-coated Fe-3.5Si-4.5Cr cores was strongly decreased in the 50–100 kHz range. In contrast, the $P_h$ of the ferrite-coated Fe-3.5Si-4.5Cr cores was higher than that of the non-coated cores because of the formation of the ferrite layer.

| TABLE I. Magnetization ($M$) of powders (emu/g) and density of compacted cores (g/cm$^3$). |
|---------------------------------------------------------------|
| Fe-3.5Si  | Fe-3.5Si-4.5Cr  | Fe-5.5Si-4.0Cr  | Fe-3.0Si-6.5Cr  |
| Density  | $M$   | Density  | $M$   | Density  | $M$   | Density  | $M$   |
| Non-coated | 6.42  | 209   | 6.46  | 188   | 6.30  | 179   | 6.65  | 183   |
| Ferrite-coated | 6.19  | 185   | 6.59  | 183   | 6.42  | 175   | 6.74  | 179   |
FIG. 3. Frequency dependence of permeability $\mu'$ and $\mu''$ for compacted cores (a), (b) before and after annealing at 773 K for 15 min; (c), (d) after annealing at 973 K for 15 min.

| Ferrite layer | Fe-3.5Si | Fe-3.5Si-4.5Cr | Fe-5.5Si-4.0Cr | Fe-3.0Si-6.5Cr |
|---------------|----------|----------------|----------------|----------------|
| Without       | 1.8 $\times 10^3$ | 4.6 $\times 10^{-1}$ | 2.2 $\times 10^{-1}$ | 1.8 $\times 10^{-1}$ | 2.1 $\times 10^{-1}$ | 8.8 |
| With          | 9.0 $\times 10^3$ | 4.1 $\times 10^3$ | 5.2 $\times 10^3$ | 2.5 $\times 10^3$ | 5.2 |
CONCLUSIONS

In this work, a Ni-Zn-Fe ferrite was deposited on the surface of Fe-3.5Si mass % and three kinds of Fe-Si-Cr (Fe-3.5Si-4.5Cr, Fe-5.5Si-4.0Cr, and Fe-3.0Si-6.5Cr mass %) powders to obtain high magnetization, high $\mu'$, low $P_{cv}$, and heat stable materials. The volume fraction of ferrite was estimated to be about 4 vol. %. The magnetization of the ferrite-coated powders was about 180 emu/g, almost the same as that of the metal powders and higher than that of Fe-3.5Si-4.5Cr powder contained 4 vol. % of paramagnetic water glass (176 emu/g). The ferrite-coated Fe-Si-Cr cores exhibited good heat stability, and Cr$_2$O$_3$ passivation was necessary to prevent diffusion between the ferrite and metal during annealing. Cores annealed at 973 K exhibited a constant permeability $\mu'$ of 55 to 60 in the 1–50 MHz range, while conventional organic insulators start to break down below 773 K. The $P_{cv}$ of the ferrite-coated Fe-3.5Si-4.5Cr was 448 kW/m$^3$ at 50 mT and 100 kHz, and its $P_e$ was strongly decreased. However, its $P_h$ was increased by the formation of the ferrite layer. Optimization of the thickness of the ferrite layer is necessary to decrease $P_h$ and make such ferrite-coated materials more advantageous.

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