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Unusual high $B_s$ for Fe-based amorphous powders produced by a gas-atomization technique

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Fe-based alloy powders with a high Fe content of about 81 at. % were produced by a gas-atomization technique. Powders of Fe$_{81}$Si$_{1.9}$B$_{5.7}$P$_{11.4}$ (at.%) alloy showed a good glass forming ability and exhibited unusual high saturation magnetic flux density of 1.57 T. The core-loss property at a frequency of 100 kHz for the compacted core made of the Fe$_{81}$Si$_{1.9}$B$_{5.7}$P$_{11.4}$ powder is evaluated to be less than 500 kW/m$^3$ under a maximum induction of 100 mT. Moreover, good DC-superposition characteristic of the core was also confirmed. These results suggest that the present Fe-based alloy powder is promising for low-loss magnetic-core materials and expected to contribute in miniaturization of electric parts in the near future.

I. INTRODUCTION

It is well known that Fe-based amorphous ribbons with excellent soft magnetic properties have been commercially used for distribution transformers and consumer electronics as magnetic cores.1–5 However, the shape is generally limited only to stacked or wound cores due to their machinability and deformability. On the other hand, an excellent compactability of the amorphous powder is expected to make the core shape more complicated and smaller.6–9 Recently, several production processes such as Spinning-water-atomization process (SWAP$^8$) have been proposed to produce Fe-based amorphous powders. The process have much contribution for producing Fe-based amorphous powder even with a high Fe content because of their high cooling rates of exceeding to $10^6$ K/s. Unfortunately, this production process also have some disadvantages such as oxidation or corrosion due to the contacting powder surface with atomizing medium of water or solutions. To avoid such a disadvantage, compositional modification by adding small amount of element with anti-oxidation or anti-corrosion properties might be useful. However, these additional elements drastically degrade the saturation magnetic flux density ($B_s$) due to the decrease of Fe content or invar effect.

Recently, we have successfully prepared Fe-based hetero-amorphous powder with high Fe content of 81 at. % (i.e., Fe$_{81}$Si$_{1.9}$B$_{5.7}$P$_{11.4}$) following the investigation of optimum addition of metalloid elements. In this work, we intend to clarify the magnetic properties such as $B_s$, core loss ($P_{cv}$) and DC-superposition characteristic of the core made of the hetero-amorphous powders. Industrial feasibility of the core will also be discussed.

II. EXPERIMENTAL PROCEDURE

Fe–Si–B–P quaternary alloy ingots were prepared by an induction melting a mixture of Fe (99.98 mass%), pre-melted Fe$_3$P (99.99%), crystal B (99.5%) and Si (99.999%) in an Ar atmosphere. Molten alloys were rapidly solidified into ribbon forms by a single roller melt-spinning method in ambient atmosphere. To clarify the thermal characteristics, the as-quenched ribbons were examined by using differential scanning calorimeter (DSC) at a heating rate of 0.67 K/s. The glass-forming ability (GFA) was evaluated by comparing the super cooled liquid regions ($\Delta T$) among the alloys.12 $\Delta T$ is
defined as temperature interval between glass transition temperature ($T_g$) and onset of crystallization temperature ($T_x$). $T_g$ was evaluated by reading the temperature at the intersection of the tangent at the inflection point of the baseline. Powders were produced for the selected alloys having the best GFA by confined-type gas-atomization technique in Ar atmosphere with gas atomization pressure ranging from 4 to 5 MPa. Structures and morphologies of the obtained powders were examined using X-ray diffractometry (XRD) and scanning electron microscopy (SEM), respectively.

Toroidal cores with an outer diameter of 13 mm, an inner diameter of 8 mm and a height of 3 mm were then prepared by hot-pressing a mixture of powder and silicon resin under an applied stress of 440 MPa. Bs for the cores was measured by a vibrating sample magnetometer (VSM) under a maximum applied field of 800 kA/m. Coercivity of the core ($H_C$) measured under a $B_{in}$ of 100 mT at a frequency of 100 kHz by B-H analyzer (Iwatsu SY-8217). Initial permeability ($\mu_i$) and $P_{cv}$ were measured by an impedance analyzer (Agilent 4294A) and B-H analyzer (Iwatsu SY-8217), respectively.

III. RESULTS AND DISCUSSION

Compositional dependence on $\Delta T$ of Fe$_{81}$Si$_x$B$_y$P$_z$$_{19}$ ($x + y + z = 1$) ribbons was investigated. Distinct $T_g$ could be found for the ribbons with the composition of around Fe$_{81}$Si$_{1.5}$B$_5$P$_{12.5}$, suggesting that alloys in the vicinity of this nominal composition may have relatively high GFA. A wide $\Delta T$ of 38K was obtained for the Fe$_{81}$Si$_{1.5}$B$_5$P$_{12.5}$ ribbon shown Figure 1. A distinctive $T_g$ was also exhibited by the powders with the same nominal composition. Figure 2(a) shows the XRD profiles of three kinds of atomized powders with the composition around Fe$_{81}$Si$_{1.5}$B$_5$P$_{12.5}$. The Fe$_{81}$Si$_{3}$B$_6$P$_{10}$ and Fe$_{81}$Si$_{1.5}$B$_3$P$_{12.5}$ powders show a mixed pattern of halo and Bragg-peaks of into metallic compounds, such as Fe$_3$B, in their XRD profiles. On the other hand, Fe$_{81}$Si$_{1.9}$B$_5$P$_{11.4}$ powder shows broader halo pattern and smaller Bragg-peak of $\alpha$-Fe phases as compared to the other alloys, suggesting that the Fe$_{81}$Si$_{1.9}$B$_5$P$_{11.4}$ alloy has higher GFA. Figure 2(b) shows the DSC curve taken from the Fe$_{81}$Si$_{1.9}$B$_5$P$_{11.4}$ alloy powder. $T_g$ and $T_x$ is evaluated to be 699 K and 731 K, respectively, and therefore $\Delta T$ is 32 K. This $\Delta T$ value is almost comparable to that of the Fe$_{81}$Si$_{1.5}$B$_5$P$_{12.5}$ ribbon. This small difference in $\Delta T$ value is attributed to the precipitation of crystalline phases.

Figure 3(a) shows the morphology of the Fe$_{81}$Si$_{1.9}$B$_5$P$_{11.4}$ alloy powder. The powders are spherical and no crystalline precipitation feature could be found on the powder surface. A magnetic core made of the powders is shown in Figure 3(b).

**FIG. 1.** DSC curve taken from the Fe$_{81}$(Si$_x$B$_y$P$_z$)$_{19}$ ribbons.
FIG. 2. (a) XRD profiles of Fe-based alloy powders and (b) DSC curve taken from the Fe$_{81}$Si$_{19}$B$_{5.7}$P$_{11.4}$ powder.

FIG. 3. (a) Morphology of Fe$_{81}$Si$_{19}$B$_{5.7}$P$_{11.4}$ powder produced by gas-atomization and (b) outer appearance of compacted core using Fe$_{81}$Si$_{19}$B$_{5.7}$P$_{11.4}$ powder.
Table I summarizes coercivity ($H_C'$), core loss ($P_{CV}$) at a frequency of 100 kHz, hysteresis loss coefficient ($K_h$) and eddy current loss coefficient ($K_e$) for the magnetic cores of the three alloy compositions with Fe content of 81 at.%. The data for the core of alloy with a Fe content of 76 at.% is also shown for comparison. Even for the same Fe content of 81 at.% each core shows different $B_s$.
FIG. 5. (a) $P_{cv}$ as a function of frequency at $B_m = 0.5$ T and 0.1 T and (b) frequency dependence of the $\mu'$. 

value. The $B_s$ value increases with increasing the volume fraction of $\alpha$-Fe phase. It is worth noting that the Fe$_{81}$Si$_{10}$B$_6$P$_{10}$ core exhibited remarkable high $B_s$ value of 1.64 T.

Figure 4 shows the TEM image for the (a) Fe$_{81}$Si$_{10}$B$_5$P$_{11}$ and (b) Fe$_{81}$Si$_{10}$B$_5$P$_{12}$ powder cores. Fe$_{81}$Si$_{10}$B$_5$P$_{11}$ alloy powder is not amorphous but hetero-amorphous including a large number of $\alpha$-Fe grains with extremely small size of less than about 3 nm, while the Fe$_{81}$Si$_{10}$B$_5$P$_{12}$ alloy powder shows clear crystalline structure. The small amount of $\alpha$-Fe phase precipitation for the Fe$_{81}$Si$_{10}$B$_5$P$_{11}$ powder core as shown in the Figure 2(a) is due to the local ordering of $\alpha$-Fe similar to the previous report. The relatively higher $H_c'$ for the Fe$_{81}$Si$_{10}$B$_5$P$_{12}$ powder core is attributed to the precipitation of Fe$_3$B shown in Figure 2. The other reason for the degradation of magnetic softness is the grain size ($D$). $H_c$ steeply increases with increasing $D$ (up to $D \approx 50$ nm) according to the $D^6$ law.

Figure 5 shows frequency dependence of the $P_{cv}$ for the three kinds of cores measured at a maximum flux density ($B_m$) of 50 mT and 100 mT. The $P_{cv}$ for the Fe$_{76}$Si$_{9}$B$_{10}$P$_5$ core is also shown for comparison. Traditionally, core loss is composed of classical eddy current loss and hysteresis loss and defined as:

$$P_{cv} = K_h f + K_e f^2$$  \hspace{1cm} (1) 

where $f$ is the frequency, $K_h$ and $K_e$ are hysteresis and eddy current loss coefficients, respectively. $K_h$ and $K_e$ are frequency dependent and the coercive force $H_c'$ (measured under a $B_m$ of 100 mT at a
frequency of 100 kHz by B-H analyzer) is proportional to $K_h$. $K_e$ decreases by increasing electrical resistivity of core, because core loss at high frequency range is dominated by eddy-current loss. Figure 5(b) shows frequency dependence of permeability ($\mu_i$). No decreasing in $\mu_i$ can be found for all the cores even with increasing frequency up to $\sim 10$ MHz because of complete insulation between the powder surfaces by silicone resin binder. The $P_{cv}$ for Fe$_{81}$Si$_{19}$B$_5$P$_{11.4}$ core is evaluated to be 372 kW/m$^3$ under a $B_m$ of 100 mT at a frequency of 100 kHz.

To investigate industrial feasibility, $B_s$ and $P_{cv}$ for the present core are compared to those for commercial cores made of Fe-based amorphous, Fe-Ni, Fe-6.5%Si and Mn-Zn ferrite. $B_s$ and $P_{cv}$ for the Fe-based amorphous, Fe-Ni, Fe-6.5%Si and Mn-Zn ferrite cores are 1.3 T and 300-400 kW/m$^3$,$^{16}$ 1.5 T and 1300-1800 kW/m$^3$,$^{17}$ 1.6 T and -2000 kW/m$^3$,$^{16}$ 0.5 T and 200-500 kW/m$^3$,$^{16}$ respectively. The present core has good magnetic properties that are low $P_{cv}$ and high $B_s$, as compared to those of the commercial cores. In particular, for switching device application, the $P_{cv}$ at 100 kHz as the target frequency for the present core is only 25% of the commercial dust cores. In addition, $B_s$ for the present core is three times larger than that for Mn-Zn ferrite core. These beneficial properties suggest that the present powder is potential to develop lighter, smaller and large-current-capable switching devices.

Figure 6 shows the relative permeability ($\mu_r$) and the reduction ratio of $\mu_r$ with increasing magnetic field measured at 100 kHz under a DC-bias field ($H_{DC}$). As seen in the figure, the Fe$_{81}$Si$_{19}$B$_5$P$_{11.4}$ core exhibited superior characteristics when compared with the other cores as the reduction ratio of $\mu_r$ was only 20% even at $H_{DC}$ of 8000 A/m. Degradation of permeability is mainly caused by magnetic saturation. Therefore inductance drops suddenly at high frequency region, if $B_s$ of a core material is low. The Fe$_{81}$Si$_{19}$B$_5$P$_{11.4}$ core with high $B_s$ and low $P_{cv}$ can be used for large current application field, and may contribute the miniaturization of electrical parts with higher efficiency.
IV. CONCLUSIONS

Optimum composition of Fe-based hetero-amorphous alloy powders with high Fe content of 81 at.% exhibiting high saturation magnetic flux density was investigated. The results are summarized as follows.

1. Fe$_{81}$Si$_{1.9}$B$_{5.7}$P$_{11.4}$ (at.%) showed relatively high glass-forming ability, and the powders made of the alloy exhibited high $B_s$ of 1.57 T.

2. The $P_{cr}$ for the Fe$_{81}$Si$_{1.9}$B$_{5.7}$P$_{11.4}$ powder core is evaluated to be 372 kW/m$^3$ under a $B_m$ of 100 mT, while is comparable to Mn-Zn ferrite core.

3. The Fe$_{81}$Si$_{1.9}$B$_{5.7}$P$_{11.4}$ core exhibited good DC-superposition characteristic. Because of its high $B_s$ and low $P_{cr}$, it may be used for large current application field and contribute for miniaturization of electrical parts with higher efficiency.

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