<table>
<thead>
<tr>
<th>Authors</th>
<th>有田 A., 松本 H., 佐藤 S., 坂本 A.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Journal</td>
<td>Journal of Applied Physics</td>
</tr>
<tr>
<td>Volume</td>
<td>105</td>
</tr>
<tr>
<td>Number</td>
<td>7</td>
</tr>
<tr>
<td>Page</td>
<td>07A324</td>
</tr>
<tr>
<td>Year</td>
<td>2009</td>
</tr>
<tr>
<td>URL</td>
<td><a href="http://hdl.handle.net/10097/52382">http://hdl.handle.net/10097/52382</a></td>
</tr>
<tr>
<td>DOI</td>
<td>10.1063/1.3072373</td>
</tr>
</tbody>
</table>
High $B_s$ nanocrystalline alloys with high amorphous-forming ability

A. Urata,1,a) H. Matsumoto, 1 S. Sato, 1 and A. Makino2

1NEC TOKIN Corporation, Sendai 982-8510, Japan
2Institute for Materials Research, Tohoku University, Sendai 982-8577, Japan

(Presented 11 November 2008; received 17 September 2008; accepted 20 November 2008; published online 11 March 2009)

The soft magnetic property and the structure of the new type nanocrystalline Fe-(P,B,Si,Nb,Cu) alloy with high Fe content about 80 at. % have been investigated. In this paper Fe77.9P3B14Nb5Cu0.1 alloy has a supercooled liquid region ($\Delta T_s$) of 33 K defined by the difference between the glass transition temperature ($T_g$) and crystallization temperature ($T_x$), which would be relating to the high glass-forming ability leading a thickness of 140 $\mu$m melt-spun ribbon with a glassy phase. The Fe80.3P2B10Si2Nb5Cu0.1 alloy with the higher Fe content can be melt spun into the thick ribbon with thickness of 65 $\mu$m due to rather large amorphous-forming ability. In addition, a homogeneous nanostructure composed of $\alpha$-Fe grain with around 15 nm in diameter was realized after crystallization, and the nanostructured alloy exhibits the high $B_s$ of 1.55 T and the low coercivity ($H_c$) of 4.6 A/m. The $B_s$ and the glass or amorphous-forming ability of the alloys are superior to those for typical nanocrystalline. Therefore, the miniaturization and the realization of higher efficiency of power supply circuits in electric devices are able to be expected using these nanocrystalline Fe-(P,B,Si,Nb,Cu) alloys. © 2009 American Institute of Physics. [DOI: 10.1063/1.3072373]

I. INTRODUCTION

Further evolution of soft magnetic materials is strongly required in miniaturization and realization of higher efficiency of power supply circuits of electric devices. It is known that magnetic materials such as Fe–Si–Al, 80Ni–Fe, and Co-based amorphous with a lower magnetocrystalline anisotropy and magnetostriction are shown to improve the coercivity ($H_c$) and permeability ($\mu$) of soft magnetic characteristics, in especial. However these materials are not suitable for the power supply circuits of electric devices because of their low saturation magnetic flux density ($B_s$).

Recently, some nanocrystalline soft magnetic materials consisting of $\alpha$-Fe–(Si) grain nanocrystalline phase have been obtained by crystallizing melt-spun amorphous ribbons such as Fe–B–Si–Nb–Cu.1,2 Owing to the strong magnetic coupling between the crystalline grains through the remaining ferromagnetic amorphous phase, the apparent anisotropy is decreased and the alloys exhibit good soft magnetic properties.3,4 Generally, an advanced rapid quenching process is required for production of nanocrystalline alloys. Therefore, the representative nanocrystalline alloy in now practical use with low amorphous-forming ability was limited to the small size form such as ribbon with a thickness of about 20 $\mu$m. In addition, $B_s$ of these alloys is also low because of low Fe content of about 73 at. %. It is necessary to enhance further the amorphous-forming ability and $B_s$ of the nanocrystalline alloy without degrading the soft magnetic property. In the present study, we tried to develop the nanocrystalline Fe-rich Fe–(P,B,Si,Nb,Cu) alloy with high amorphous-forming ability and $B_s$ as well as an excellent soft magnetic properties.

II. EXPERIMENTS

Fe–(P,B,Si,Nb,Cu) ingots were prepared by an induction melting, the mixture of pure metals of Fe, Nb, and Cu and pure metalloid of crystal B, Si, and Fe3P alloy in an Ar atmosphere. Amorphous and glassy alloys were produced in a ribbon form by the single roller melt-spinning method in an Ar atmosphere. Crystallizing treatment was carried out by treating the samples at 873 K for 300 s in an Ar atmosphere. The as-quenched and annealed structures were examined by x-ray diffraction (XRD) with Cu $K\alpha$ radiation and transmission electron microscopy (TEM). The glass transition temperature ($T_g$) and crystallization temperature ($T_x$) were estimated by a differential scanning calorimetry (DSC) at a heating rate of 0.67 K/s. The $B_s$ under an applied field of 400 kA/m was measured by a vibrating sample magnetometer. The $H_c$ was measured under an applied field of 1 kA/m with a dc $B$-$H$ loop tracer.

III. RESULTS AND DISCUSSION

Figure 1 shows the DSC curve of the melt-spun Fe77.9P3B14Nb5Cu0.1 as-quenched ribbon with thickness of about 20 $\mu$m. With increasing temperature, this glassy alloy shows the sequent change in the glass transition, followed by a supercooled liquid region, and then crystallization. A glass transition was observed in low Fe content in the glassy alloy system5,6 and known to be one of the indicators for glass-forming ability. However, it is seen that this alloy which is comparatively high Fe content of 77.9 at. % has an obvious temperature interval of the supercooled liquid region of 33 K and can be expected to have high glass-forming ability.

Figure 2 shows the XRD pattern taken from the free surface of melt-spun (a) Fe77.9P3B14Nb5Cu0.1, (b) Fe80.3P2B10Si2Nb5Cu0.1, and (c) Fe73.5B9Si13.5Nb3Cu1 rib-

---

a)Electronic mail: urata@nec-tokin.com.
bons with critical thickness of 140, 65, and 45 μm for glass or amorphous formation, respectively. The XRD patterns reveal only typical halos, and no sharp diffraction peak corresponding to crystalline phase is visible. Based on the above result, we can see that Fe_{77.9}P_{3}B_{14}Nb_{5}Cu_{0.1} alloys have high glass or amorphous-forming ability compared with representative Fe_{73.5}B_{9}Si_{13.5}Nb_{5}Cu_{1} nanocrystalline alloy with the lower Fe content of 73.5 at.%. Particularly, the Fe_{77.9}P_{3}B_{14}Nb_{5}Cu_{0.1} alloy with the supercooled liquid region of 33 K has much higher glass or amorphous-forming ability than the alloys without glass transition.

Figure 3 shows the TEM images and the selected-area electron diffraction (SAED) patterns of the (a) as-quenched and (b) crystallized Fe_{80.9}P_{2}B_{10}Si_{2}Nb_{5}Cu_{0.1} alloy ribbons with thickness of about 20 μm. Figure 3(a) indicates that the as-quenched Fe_{80.9}P_{2}B_{10}Si_{2}Nb_{5}Cu_{0.1} alloy has a single amorphous structure. On the other hand, Fig. 3(b) shows a homogeneous nanocrystallized structure composed of α-Fe grains from 10 to 20 nm in diameter and the amorphous phase. In addition, the crystallized Fe_{80.9}P_{2}B_{10}Si_{2}Nb_{5}Cu_{0.1} alloy ribbon exhibits a good soft magnetic characteristic, $H_c$ of 4.6 A/m, because of low magnetocrystalline anisotropy due to the nanocrystalline structure and low magnetostriction due to compensation of negative and positive magnetostrictions generated in the α-Fe grains and the amorphous phase, respectively. Meanwhile, the nanostructure of the Fe_{77.9}P_{3}B_{14}Nb_{5}Cu_{0.1} alloy has the relatively large and non-uniform α-Fe grains with 10–50 nm in size. Therefore, the $H_c$ of Fe_{77.9}P_{3}B_{14}Nb_{5}Cu_{0.1} alloy ranging from 30 to 70 A/m is higher than that of Fe_{80.9}P_{2}B_{10}Si_{2}Nb_{5}Cu_{0.1} alloy.

Figure 4 shows the $B_s$ of the melt-spun Fe_{77.9}P_{3}B_{14}Nb_{5}Cu_{0.1}, Fe_{80.9}P_{2}B_{10}Si_{2}Nb_{5}Cu_{0.1}, and Fe_{73.5}B_{8}Si_{13.5}Nb_{5}Cu_{1} ribbons with thickness of about 20 μm after crystallization. It is seen that $B_s$ of crystallized Fe_{77.9}P_{3}B_{14}Nb_{5}Cu_{0.1} and that of Fe_{80.9}P_{2}B_{10}Si_{2}Nb_{5}Cu_{0.1} alloys are 1.50 and 1.55 T, respectively, which are considerably higher than 1.3 T of Fe_{73.5}B_{8}Si_{13.5}Nb_{5}Cu_{1} alloy due to the high Fe content.

Table 1 summarizes the thermal stability, the critical thickness ($t_{\text{max}}$) for glass or amorphous formation, and the magnetic properties of the nanocrystalline Fe–(P,B,Sn,Cu)
alloy and the representative nanocrystalline Fe–(B,Si,Nb,Cu) alloy. These nanocrystalline Fe–(P,B,Si,Nb,Cu) alloys realize the coexistence of high $B_s$ and high glass or amorphous-forming ability, which is superior to the nanocrystalline Fe$_{73.5}$B$_9$Si$_{13.5}$Nb$_3$Cu$_1$ alloy. Therefore, the nanocrystalline Fe–(P,B,Si,Nb,Cu) alloys should have great advantage as engineering materials and are expected to be used for many kinds of power supply circuits of electric devices.

IV. CONCLUSIONS

We have successfully developed a Fe–(P,B,Si,Nb,Cu) nanocrystalline alloy with combination of high glass or amorphous-forming ability and excellent magnetic properties such as high $B_s$ and low $H_c$. The results obtained are summarized as follows:

1. As-quenched Fe$_{77.9}$P$_3$B$_{14}$Nb$_5$Cu$_{0.1}$ ribbon alloys have a supercooled liquid region of approximately 33 K, which is possibly relating to the high glass-forming ability leading the large thickness of 140 $\mu$m of melt-spun ribbon with a glassy phase.
2. Fe$_{80.9}$P$_2$B$_{10}$Si$_2$Nb$_5$Cu$_{0.1}$ alloy can be melt spun into the thick ribbon with thickness of 65 $\mu$m due to rather large amorphous-forming ability in spite of the higher Fe content of 80.9 at. % than 73.5 at. % of the representative Fe–(B,Si,Nb,Cu) alloy in now practical use.
3. The nanocrystalline Fe$_{80.9}$P$_2$B$_{10}$Si$_2$Nb$_5$Cu$_{0.1}$ exhibits the high $B_s$ of 1.55 T and the low $H_c$ of 4.6 A/m due to the high Fe content and the homogeneous nanostructure composed of $\alpha$-Fe grains with diameter of 10–20 nm.