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# Ternary Fe-B-C and quaternary Fe-B-C-Si amorphous alloys with glass transition and high magnetization

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The ternary Fe-B-C amorphous alloys exhibit good soft magnetic properties and high saturation magnetization. The high magnetization is favorable to reduce the size of the magnetic devices. However, mass production of the Fe-B-C amorphous alloys is difficult due to their low glass-forming ability (GFA). In the present study, the various properties of the ternary Fe-B-C and quaternary Fe-B-C-Si amorphous alloys have been investigated. It has been discovered for the first time that some ternary Fe-B-C amorphous alloys with the Fe content close to that of the $Cr_{23}C_6$-type metastable $Fe_{23}(B, C)_6$ phase exhibit a glass transition prior to crystallization on heating. The alloys with the glass transition also have high mass magnetization of 176–178 A m$^2$/kg at room temperature in as as-quenched state. As a result of attempts to improve GFA by Si addition to the Fe-B-C alloys, it has been discovered that the quaternary $(Fe_{0.780}B_{0.152}C_{0.068})_{96}Si_4$ and $(Fe_{0.767}B_{0.148}C_{0.085})_{96}Si_4$ amorphous alloys exhibit the large GFA. The large GFA of the quaternary Fe-B-C-Si alloys enables to produce thick amorphous specimens with 120 μm in thickness and with high mass magnetization of approximately 170 A m$^2$/kg at room temperature in as as-quenched state. The ternary Fe-B-C and quaternary Fe-B-C-Si amorphous alloys with high GFA as well as high magnetization are suitable for a core material of various magnetic components.



## 1. Introduction

Recently, many kinds of portable electronic devices, *e.g.*, laptop computers, smart phones and tablets are rapidly widespread. It causes a strong demand for the miniaturization and the improvement of energy efficiency of these devices. Many magnetic components such as inductors and transformers are used in the electric devices, especially their power supply unit. Of course, the same requests as described above are also required to the magnetic components.

Soft magnetic materials are used as a core material of the magnetic components. The high saturation magnetic induction ($\mu_0 M_s$, where $\mu_0$ is the permeability of vacuum and $M_s$ is the saturation magnetization) and the low magnetic loss are demanded for the soft magnetic materials to reduce the size and the energy loss of the magnetic components. In addition, in order to enable high-density mounting, the components having the electromagnetic compatibility (EMC) and the low profile are favored. Thus, in recent years, the coil embedded inductors with no leakage magnetic flux have attracted attention in the market. Usually, the soft magnetic powder material is used as a core material for the embedded inductors. Therefore, the development of a new soft magnetic material with high $M_s$, good soft magnetic properties and good powder productivity is strongly demanded.

The Fe-based soft magnetic amorphous alloys had been developed in the 1970's and used in various fields [1]. However, because of their low glass-forming ability (GFA), the mass production of the amorphous powders by gas or water atomization is difficult. In the 1990's, the Fe-based bulk metallic glasses (BMGs) with large GFA were developed [2–4]. Since the glassy powder can be easily prepared by the atomization, some BMGs such as Fe-Cr-P-C-B-Si [5] and Fe-P-B-Nb-Cr [6] have been commercialized. However, the Fe-based soft magnetic BMGs have lower $M_s$ (typical $\mu_0 M_s$ is 1.2–1.3 T) compared with the ordinary Fe-based amorphous alloys because they include the large amount of the solute elements to enhance GFA. Especially, it is well known that the addition of transition metal elements (such as Zr, Nb, Mo, Hf, Ta and W) enhance the GFA of the Fe-based BMGs [3]. However, it also brings the remarkable decrease in $M_s$ and the increase of the material cost.

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The ternary Fe-P-C alloy system shows the very low eutectic temperature (1225 K) in Fe-rich composition [7]. The existence of deep eutectic is highly beneficial for glass formation [8]. Therefore, many Fe-based BMGs have been developed based on the Fe-P-C alloy system. However, the vapor pressure of P is quite high, and the pollution of crucibles and of furnaces by P cause various problems in the manufacturing premises.

The Fe-B-C amorphous alloys are attractive because they exhibit the high $\mu_0 M_s$ approximately 1.7 T at room temperature [9–11]. However, GFA of the Fe-B-C is much smaller than that of the commercial amorphous alloys such as Fe-B-Si [12]. If GFA of the Fe-B-C alloys can be enhanced while maintaining the high $\mu_0 M_s$, a new and very attractive soft magnetic material will be developed. In the present study, the composition dependence of the thermal and magnetic properties of the ternary Fe-B-C alloy has been studied. It has been found that the Fe-B-C amorphous alloys with the Fe content of approximately 79.3 at%, which is the value of $Fe_{23}(B, C)_6$ phase, exhibit a small endothermic reaction prior to crystallization on heating. The endothermic reaction prior to the crystallization is due to the glass transition. The addition of Si to the Fe-B-C alloys makes the glass transition more clearly, and GFA of the alloys is significantly enhanced.

## 2. Experimental procedure

The ternary Fe-B-C alloys having the low liquidus temperature ($T_l$) less than 1473 K were selected according to the equilibrium phase diagram of the Fe-B-C ternary alloys [13]. Then, the effect of the Si addition to some ternary alloys has been investigated. The mixtures of pure Fe (99.99%), B (99.5%), C (99.5%) and Si (99.999%) were melted by an arc furnace in an Ar atmosphere. The weight loss during arc melting was controlled 0.1% or less. This means that the compositional error is within 0.4 at% even in the worst case, the alloy has high Fe content of approximately 80 at% and only B with the smallest atomic mass was lost. The rapidly-solidified ribbons with approximately 1 mm in width were prepared by a single-roller melt-spinning apparatus with a Cu wheel in an Ar atmosphere. The structure of the specimens was examined by X-ray diffractometry (XRD) with Cu $K_\alpha$ incident radiation. The thermal properties of the alloys were investigated using a differential scanning calorimetry (DSC) during heating. The Curie temperature ($T_C$), glass-transition ($T_g$) temperature and onset temperature of crystallization ($T_x$) were measured by using Pt-Rh pans with a thin $Al_2O_3$ liner and NETZSCH STA 449 F3 Jupiter with a heating rate of 0.67 K/s. The liquidus temperature was measured by using $Al_2O_3$ pans and NETZSCH DSC 404 C Pegasus with a heating rate of 0.33 K/s. The saturation mass magnetization ($\sigma_s$) was measured with a magnetic balance in an applied magnetic field ($H$) up to 800 kA/m at room temperature. The hysteresis loops of the approximately 70 mm long straight specimens were measured by a hysteresis loop tracer with a compensation coil under a maximum magnetic field of 10 kA/m at room temperature.

## 3. Results and discussion

Fig. 1 shows examples of the XRD profile of the as-quenched specimens approximately 20 μm in thickness taken from the free surface. The profile of the $Fe_{79.3}B_{14.3}C_{6.4}$ alloy consist only of a halo which originates from an amorphous phase. On the other hand, the XRD profile of the other alloys consist of a halo and some diffraction lines. It was reported that the primary phase of the alloys in the compositional range of 77–81 at% Fe and 6–15 at% B is the metastable $Cr_{23}C_6$-type $Fe_{23}(B, C)_6$ phase when the alloys are cast on a Cu plate and quenched in water [14]. Therefore, the precipitated crystalline phase in the $Fe_{83.1}B_{8.9}C_{8.0}$ and $Fe_{82.9}B_{11.1}C_{6.0}$ alloys is probably $Fe_{23}(B, C)_6$. On the other hand, the $Fe_{71.0}B_{25.0}C_{4.0}$ alloy probably forms an orthorhombic $Fe_3(B, C)$ phase during quenching.

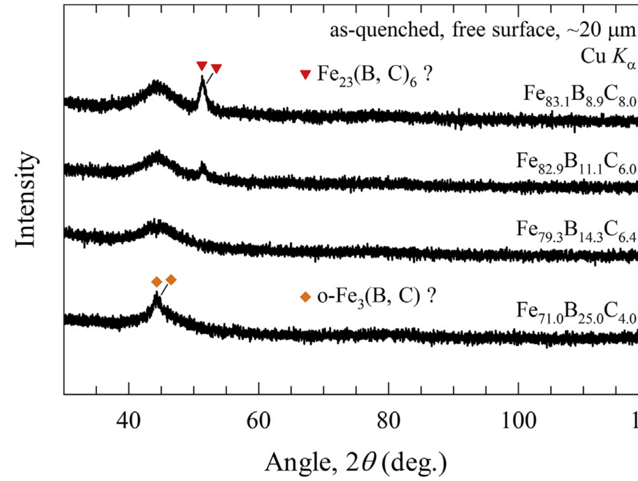


**Fig. 1.** Examples of x-ray diffraction profiles of Fe-B-C ternary alloys with thickness of approximately 20 μm taken from free surface in an as-quenched state.

The examples of the DSC traces are shown in Fig. 2. All specimens shown in Fig. 2 have a thickness of approximately 20 μm and

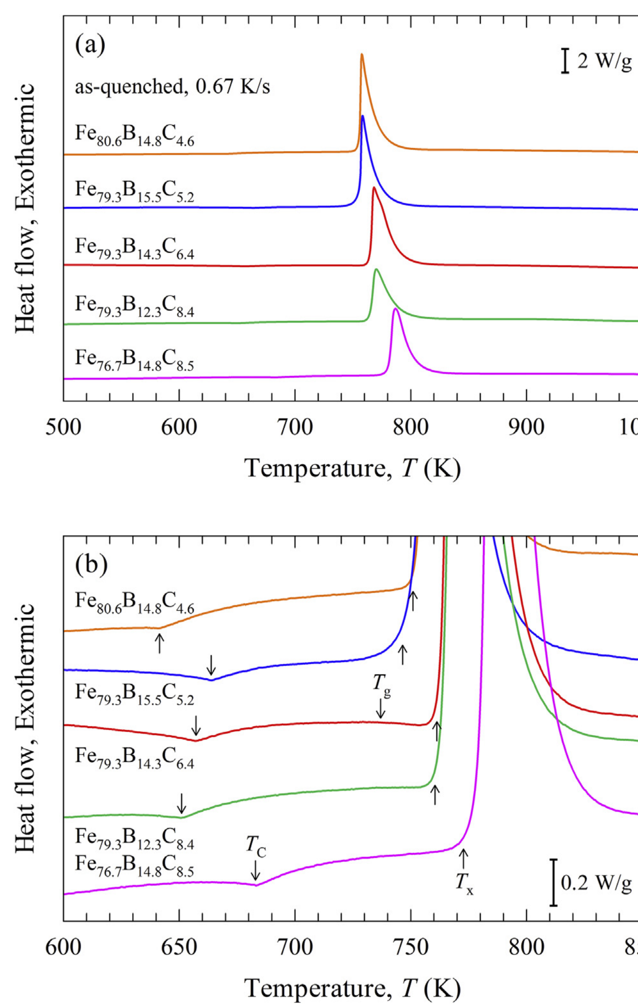


**Fig. 2.** Examples of DSC traces of ternary Fe-B-C fully amorphous alloys with thickness of approximately 20 μm. (b) is the enlarged view to show Curie temperature ($T_C$), glass-transition temperature ($T_g$) and onset temperature of crystallization ($T_x$).

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