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In-situ Lorentz microscopy of $Fe_{85}Si_2B_8P_4Cu_1$ nanocrystalline soft magnetic alloys

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ABSTRACT

Microstructure dependence of magnetic properties of soft magnetic Fe–Si–B–P–Cu nanocrystalline alloys were studied by using in-situ Lorentz microscopy in a transmission electron microscope equipped with a magnetizing system. In particular, we investigated in detail motion of magnetic domain walls in heat-treated $Fe_{85}Si_{12}B_6P_4Cu_1$ amorphous ribbons. Smooth motion of domain walls was observed for the optimally heat-treated (at 430 °C) nano-crystalline alloy. Pinning of domain walls was observed for higher-temperature-heat-treated (470 °C) ribbons. Both ribbons showed a nanocrystalline structure containing α-Fe crystallites of about 15 nm in size. Electron diffraction patterns indicated that the higher-temperature-heat-treated samples contained boride precipitates, which is considered to cause less smooth domain wall motion.

1. Introduction

Soft magnetic materials are used as magnetic cores in a variety of applications such as motors, transformers, inductors, sensors, actuators, power circuits, and electronic communication devices [1,2]. Electrical energy loss, which manifests as heat, is inherent in materials used in these applications. Metallic alloys in amorphous and/or nano-crystalline states exhibit low magnetic core loss (W) [1,2]. In addition to low W, soft magnetic materials are required to have a high saturation magnetic flux density (B_s) for device miniaturization. Unfortunately, there are very few amorphous/nanocrystalline materials that have at the same time W lower than that of grain oriented steel and B_s close to that of the grain oriented steel. Recently, Fe-rich Fe–Si–B–P–Cu nanocrystalline alloys have been reported to have these necessary characteristics [3–5]. Therefore, these alloys have large application potential in saving electrical energy in various devices.

The Fe–Si–B–P–Cu alloys have a uniform nanocrystalline structure composed of high density α-Fe grains with the size less than 20 nm surrounded by remaining amorphous matrix. A uniform nanocrystalline structure is obtained after an optimum heat-treatment of an as-quenched amorphous alloy. Sharma et al. [5] reported that heat-treatment at temperatures lower than the optimum temperature results in a significantly larger volume of remaining amorphous phase, whereas higher temperature heat-treatment leads to crystallization of iron-boride phases.

Magnetic properties of nano-crystalline alloys strongly depend on the type of phases (crystalline and amorphous), distribution, and their volume fraction. High density α-Fe nanocrystals are required for strong magnetic exchange coupling, which is necessary to average out the magnetocrystalline anisotropy of α-Fe [1]. A larger separation between α-Fe grains i.e. relatively more volume fraction of amorphous phase, leads to indirect exchange coupling among α-Fe grains (through the amorphous matrix), and affect the magnetic properties. Similarly appearance of a minor amount of iron-boride phase can significantly increase the coercivity (H_c) [6]. In soft magnetic materials coercivity is mainly governed by the motion of domain walls, and their propagation is affected by the presence of inhomogeneities, which can be amorphous/iron boride phases. Therefore, it is important to study motion of domain walls in the newly developed Fe–Si–B–P–Cu alloys heat-treated at different temperatures. The $Fe_{85}Si_2B_8P_4Cu_1$ alloys were selected because of good magnetic properties ($H_c \sim 6$ A/m and $B_s \sim 1.85$ T) and an ability to produce as-quenched X-ray amorphous ribbons [5]. Motion of magnetic

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domain walls was studied by using in-situ Lorentz microscopy in a transmission electron microscope (TEM) equipped with a magnetizing system [7–9] in order to clarify the dependence of domain walls motion on microstructure.

2. Materials and methods

The $Fe_{85}Si_2B_8P_4Cu_1$ ribbons (width ~4–6 mm and thickness ~20 μm) were made by high frequency induction melting (in vacuum) and melt-spinning (in air) techniques. Ribbons were heat-treated in an infra-red furnace under Ar-gas flow condition. Heating rate to achieve the required heat-treatment temperature was 400 °C/min. A magnetic flux density (B) versus magnetic field (H) loop analyzer was used to measure the magnetic properties. Three samples were selected based on their heat-treatment temperature and magnetic properties: Sample 1 is heat-treated at 330 °C, which is lower than the optimum heat-treatment temperature. Sample 2 is heat-treated under the optimum conditions, i.e., at 430 °C. Sample 3 is heat-treated at a higher temperature (~470 °C) than the optimum temperature. The coercivities (H_c) of these samples were as follows: Sample 1: 21.2 A/m; Sample 2: 6.29 A/m; Sample 3: 259 A/m. The H_c of the as-quenched amorphous ribbon was ~17 A/m. The TEM samples were prepared using a JIB-4500-Multibeam focused ion beam (FIB) system. They were cross-sectioned in the direction of the thickness of the ribbons with dimensions of 10 μm wide, 2–5 μm long and 0.1 μm thick. These rectangular samples were attached to a collodion film of TEM mesh-grids by using a micro-manipulator with a glass probe. For conventional TEM observations and Lorentz microscope observations, we used HF-3300S (Hitachi High-Technologies). For in-situ Lorentz microscopy observations with a static or alternating external magnetic fields, we used JEM-3000F (JEOL) equipped with a magnetizing specimen holder, which can apply a magnetic field horizontal to the sample [7–9].

3. Results and discussion

3.1. Microstructures

Fig. 1 shows bright field images (a–c) and selected area electron diffraction patterns (d–f) for $Fe_{85}Si_2B_8P_4Cu_1$ ribbons: Sample 1 (330 °C heat-treated), Sample 2 (430 °C heat-treated), and Sample 3 (470 °C heat-treated). Sample 1 has randomly oriented α-Fe grains of size ~20–40 nm sparsely distributed in the remaining amorphous matrix (Fig. 1a and d). The α-Fe grain size decreases significantly at higher-temperature heat-treatment. Dense and uniform distributions of α-Fe grains with the size of 15 nm are noticeable in the TEM images of Sample 2 and Sample 3 (Fig. 1b and c). Such a large difference in grain sizes is due to changes in the mechanism of crystallization at different heat-treatment temperatures. Grain nucleation density, which is dependent on heat-treatment temperature, governs the grain size. The diffraction patterns of all the heat-treated ribbons (Figs. 1d–f) show Debye-Scherrer rings corresponding to α-Fe nano-crystallites. Additional diffraction spots can be noticed for Sample 3 (Fig. 1f, marked with red circles). These spots can be indexed as 112, 202 and 310 reflections from Fe_2B crystals.

Low H_c (~6 A/m) for Sample 2 (430 °C heat-treated) is due to suppression of magneto-crystalline anisotropy (K_1) of α-Fe grains through magnetic exchange interactions [6]. The necessary requirement for this to happen is that the grain size should be smaller than the ferromagnetic exchange length, which is about

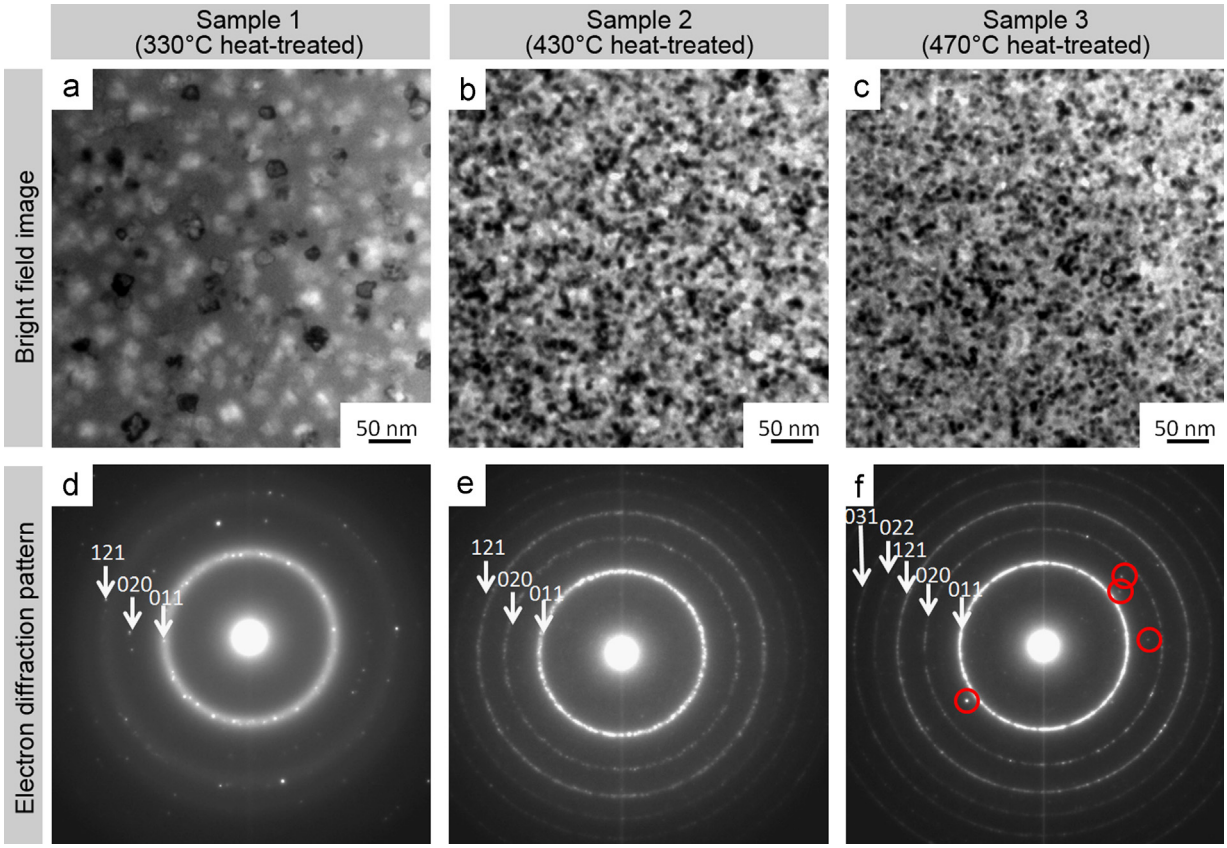

Fig. 1. Bright field images (a–c) and selected area diffraction patterns (d–f) obtained from heat-treated $Fe_{85}Si_{12}B_6P_4Cu_1$ amorphous-ribbons. The heat-treatment temperatures are 330 °C, 430 °C, and 470 °C. (For interpretation of the references to color in this figure the reader is referred to the web version of this article.)