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Effect of Si addition on crystallization behavior, thermal ability and magnetic properties in high Fe content Fe-Si-B-P-Cu-C alloy



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ABSTRACT

Dependence on the crystallization behavior, thermal ability and soft magnetic properties of the $Fe_{84.7}Si_xB_{10.5-x}P_4Cu_{0.5}C_{0.3}$ (x = 0–2.0 at.%) alloys with high Fe content were investigated. The results show that the addition of minor Si significantly weakens the crystallization peaks and increases the amorphous forming ability of the alloys. With the increase of Si content, it improves the second crystallization peak temperature and greatly enlarges the temperature interval between the two crystallization peaks, which broadens the annealing range. When Si content is up to 1.5 at.%, the alloy can effectively inhibit the precipitation of Fe-B compounds during the process of annealing. The addition of Si contributes to the formation of stable nano-structure, leading to a good soft magnetic properties for the amorphous alloy. Through the optimum annealing treatments, the nanocrystalline alloy exhibits the high Ms of 206.5 emu/g (Bs \approx 1.92 T) and low Hc of 12.5 A/m.

1. Introduction

Fe-based amorphous/nanocrystalline soft magnetic alloys were widely used in magnetic cores in the last decades due to their excellent soft magnetic properties [1-3]. Among them, the representative FI-NEMET (Fe73.5Si13.5B9Cu1Nb3) nanocrystalline alloy with high permeability (μ) and low coercivity (Hc), is becoming a trend to replace the silicon steel [4]. Through the optimum annealing treatments, the nanocrystals (particle size is about 10 nm) of the Fe₃Si phase can be formed in the remaining amorphous matrix then lead to excellent magnetic properties [1]. However, a large amount of metalloid and non-magnetic metal elements leads to the low saturation flux density (Bs ~ 1.25 T) of the alloy [5-7]. With the aim of obtaining a competitively high Bs with Si-steel, some of authors reported that partial substitution of Co for Fe and the increase of Fe content could effectively achieve a high Bs in amorphous alloys [8,9]. However, the above two types of the alloys contain expensive metal elements (i.e. Nb, Co, etc.), which increases the cost of materials [10]. Therefore, a new type of amorphous alloys with high Bs, low core loss and low cost need to be explored. Recently, a new system nanocrystalline FeSiBCuP soft magnetic alloys have attracted much attention to researchers [11-14]. Compared with the previous alloys, the difference is that the Nb/Co element is replaced by P element and at the same time the iron content greatly increases in these alloys system. According to the previous report [15], it is considered that the P element is advantageous to increase the amorphous forming ability and obtain uniform and fine

nanocrystalline structure during the process of annealing. It is worth noting that the enhancement of Fe content could increase magnetic properties and reduce the cost of materials. The Bs of the nanocrystalline alloys is up to 1.85 T [16,17]. Ribbons are usually subjected to annealing which converts from amorphous to nanocrystalline structure [18]. In addition, whether an alloy can obtain the excellent soft magnetic properties, it mainly depends on its crystallization behavior after annealing. In the choice of annealing temperatures, we try to control the nano-structure without precipitation of the secondary phase which deteriorates the soft magnetic properties, such as Fe-B phase [19-21]. However, the effect of elements on the magnetic properties after annealing and the sensitivity to temperatures for amorphous alloys is still relatively unexplored. The purpose of this paper is to provide the effect of Si content on the temperatures sensitivity for these alloys, especially in the annealed state. Thus, we aim to investigate that the crystallization behavior, thermal ability and magnetic properties in high Fe content of the newly alloy $Fe_{84.7}Si_xB_{10.5-x}P_4Cu_{0.5}C_{0.3}$ (x = 0, 0.5, 1.0, 1.5, 2.0 at.%) with industrial raw materials.

2. Experimental details

The Fe_{84.7}Si_xB_{10.5-x}P₄Cu_{0.5}C_{0.3} (x = 0, 0.5, 1.0, 1.5, 2.0) alloy ingots were prepared by melting the mixtures of industrial raw materials Fe (99.9 wt.%), Si (99.9 wt.%), Cu (99.9 wt.%), C (99.9 wt.%), pre-alloyed Fe-B ingot (17.4 wt.% B) and pre-alloyed Fe-P ingot (17.0 wt.% P) by Liaoyang International Co., LTD under a high-purity argon atmosphere.

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Fig. 1. XRD patterns of as-quenched $Fe_{84.7}Si_xB_{10.5-x}P_4Cu_{0.5}C_{0.3}\ (x=0,\,0.5,\,1.0,\,1.5,\,2.0)$ alloy ribbons.

Ingots were remelted four times to enhance homogeneity. Melt-spun ribbons were produced by a single roller melt spinning method with a speed of 50 m/s in Ar atmosphere. The width and thickness of about 1 mm and 25 μ m, respectively. The as-quenched ribbons were annealed at various temperatures with a constant heat rating of 20 °C/min to the specified temperature by using a tubular furnace in vacuum atmosphere, and heated for 10 min then cooled to room temperature in the furnace. According to the previous report [20], the rapid thermal processing could effectively prevent the excessive growth of crystals and the precipitation of secondary phase. So, the alloy is annealed for short time (i.e. 10 min) contributes to a good magnetic properties. Phase structure of melt-spun ribbons was examined by X-ray diffractometer (XRD) with Cu Ka radiation. A detailed atom distribution in an asquenched specimen was investigated by Scanning electron microscope-Energy dispersive X-ray spectroscopy (SEM-EDX). Crystallization onset temperature (T_x) and peak temperature (T_p) was examined by differential scanning calorimeter (DSC) at a heating rate of 20 °C/min, under nitrogen atmosphere. Saturation magnetization (Ms) was measured with a vibrating sample magnetometer (VSM) under the maximum applied field of 15000 Oe. The density of the ribbons was obtained by the Archimedes method, and the density is about 7.42 g/cm³. A DC-BH



Fig. 2. SEM EDX spectra for Fe, Si, B, P, Cu and C elements for the as-quenched $Fe_{84.7}Si_{1.5}B_9P_4Cu_{0.5}C_{0.3}$ alloy.

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