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Optimization of crystallization, microstructure and soft magnetic properties of Fe-B-Cu alloys by rapid cyclic annealing

X.B. Zhai [a], L. Zhu [a], H. Zheng [a], Y.D. Dai [a], J.K. Chen [a], Y.G. Wang [a,*], F.M. Pan [b]

[a] College of Materials Science and Technology, Nanjing University of Aeronautics and Astronautics, Nanjing 211106, PR China
[b] College of Science, Nanjing University of Aeronautics and Astronautics, Nanjing 211106, PR China

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ABSTRACT

$Fe_{83.5}B_{16.5-x}Cu_x$ ($x = 0$, 0.7, 1.2, 1.5) amorphous alloys were rapidly annealed under various cyclic numbers (C). The crystallization behavior and its effects on the microstructure and soft magnetic properties of nanocrystalline alloys have been investigated. It is found that the cyclic annealing is more effective to refine and uniform α-Fe grains by introducing multiple rapid heating than the non-cyclic annealing. The saturation magnetic flux density of the $Fe_{83.5}B_{15}Cu_{1.5}$ nanocrystalline alloy increases from 1.79 T to 1.82 T and the coercivity decreases from 13.1 A/m to 7.5 A/m simultaneously when the cyclic numbers change from $C = 1$ to 6. Moreover, the rapid cyclic annealing with $C = 6$ enlarges the annealing temperature ranges for the $Fe_{83.5}B_{16.5}$ and $Fe_{83.5}B_{15}Cu_{1.5}$ nanocrystalline alloys with low H_C by ~20 K compared to that with $C = 1$. The Mössbauer spectroscopy confirms that the rapid cyclic annealing can inhibit the aggregation of B atoms in the amorphous matrix, which is beneficial to enhance the magnetic interaction between α-Fe nanocrystals. However, when the cyclic numbers further rise to $C = 10$, the mean grain size of the $Fe_{83.5}B_{15}Cu_{1.5}$ nanocrystalline alloy begins to increase, resulting in the deterioration of soft magnetic properties.

1. Introduction

Since the FeSiBNbCu (FINEMET) nanocrystalline alloy with a structure consisting of nanosized α-Fe grains embedded in an amorphous matrix was first reported in 1988 [1], it has been widely used in various electronic devices [2,3]. However, the relative low saturation magnetic flux density (B_S) of ~1.2 T makes it hard to meet the needs of development due to its low Fe content [1,4]. The uniform and fine nanostructure is the key to obtain excellent soft magnetic properties for nanocrystalline alloys. In the past several decades, FeBCu [5], FeBPCu [6,7] and FeSiBPCu [8] nanocrystalline alloys with high Fe content and a high B_S of ~1.8 T were successively developed. The high Fe content usually leads to the formation of pre-existing α-Fe nuclei in amorphous precursors due to the limit of cooling rate [9,10] and these nuclei will grow in size during low heating rate annealing, which results in a non-uniform precipitation of α-Fe grains [11]. In addition to the development of new Fe-based nanocrystalline soft magnetic materials, some improved nanocrystallization processes, such as rapid annealing [12–14] and magnetic field annealing [15,16], are also used to optimize the microstructure and magnetic properties of nanocrystalline alloys. Among them, the rapid annealing is believed to be capable of inhibiting the growth of the pre-existing nuclei during heating and promote the precipitation of high-density α-Fe nuclei during the crystallization [11]. It has been reported for $Fe_{82}Cu_1Nb_1Si_4B_{12}$ nanocrystalline alloy that the coercivity (H_C) can be decreased from 120 A/m to 3.2 A/m by increasing the heating rate from 0.3 K/s to 3 K/s [17].

The annealing with high heating rate is usually carried out under high annealing temperature, a pivotal temperature for the formation of a massive amount of new α-Fe nuclei, because the crystallization temperature is positively related with the heating rate [18,19]. However, in this case, the annealing temperature will be much higher than the actual crystallization temperature if the ribbons are held for long time in the heat preservation stage, which causes the coarsening of α-Fe grains. Therefore, a short holding time is necessary for the annealing with high heating rates [13]. Recently, $Fe_{85}Nb_1B_{13}Cu_1$ [20] and $Fe_{85}B_{13}Ni_2$ [21] nanocrystalline alloys which possess good soft magnetic properties have been successfully developed by annealing their corresponding amorphous precursors with an extremely high heating rate (~150 K/s)

* Corresponding author.
E-mail address: yingang.wang@nuaa.edu.cn (Y.G. Wang).

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and a very short holding time (~3 s). However, such tough annealing conditions make the process of crystallization uncontrollable and irreproducible. Although cyclic annealing as a method of thermal treatment has been used for some alloys [22], little is known about the crystallization process in Fe-based amorphous alloys during rapid cyclic annealing. In this work, the controlled rapid cyclic annealing, successfully implemented, achieves better magnetic properties than the non-cyclic annealing with high heating rate. Meanwhile, we also investigate the effect of cyclic numbers on the crystallization and nanostructure of Fe-B-Cu alloys.

2. Experimental details

Alloy ingots with nominal composition $Fe_{83.5}B_{16.5-x}Cu_x$ ($x = 0$, 0.7, 1.2, 1.5) were prepared by arc-melting of industrial raw materials Fe (99.9%), B-Fe (B: 17%, Fe: 82.9%), Cu (99.99%) in a Ti-gettered Ar atmosphere. The ingots were remelted four times to ensure a uniform chemical composition. Then amorphous ribbons were fabricated under an Ar atmosphere by the rapid quenching method onto a copper wheel with a tangential velocity of 40 m/s. To obtain a two phase composite structure, the melt spun ribbons were annealed at 603 K–743 K for 180 s with various thermal cyclic numbers ($C = 1$, 2, 4, 6 and 10) under an Ar flow, where $C = 1$ represents an isothermal annealing treatment without cycle. A RTP-1000D4 rapid annealing furnace is used for the heat treatment. The annealing process is controlled by the program. Fig. 1 shows the schematic diagram of the rapid cyclic annealing. T_a and T_R represent annealing temperature and room temperature, respectively. The total holding time at T_a of all the alloy ribbons is 180 s. The heating and cooling rates between T_a and T_R in the program of the heat treatment are both set to 10 K/s. At the end of each heat preservation stage, the argon flow rate is increased in order that the cooling rate of the ribbons satisfies the set value and then the ribbons stay at T_R for 60 s.

The thermal characteristics of the Fe-B-Cu alloy ribbons were evaluated by differential scanning calorimetry (DSC) at a constant heating rate of 20 K/min. The microstructure of the annealed ribbons was identified by X-ray diffractometry (XRD) with Cu-Kα radiation, transmission Mössbauer spectroscopy and transmission electron microscopy (TEM). Mössbauer spectra were recorded at room temperature using a ^{57}Co(Rh) source. All experimental spectra were analyzed using NORMOS program [23] to get more information about the microstructure of the nanocrystallized ribbons. The B_S and H_C of the melt spun and annealed ribbons were measured by a DC B-H loop tracer under a maximum applied field of 8000 A/m.

3. Results and discussion

As shown in Fig. 2, the thermal behavior of the amorphous $Fe_{83.5}B_{16.5-x}Cu_x$ ($x = 0$, 0.7, 1.2, 1.5) alloys is characterized by DSC. All the DSC curves show two distinct exothermic peaks. The first and second crystallization temperatures, T_{X1} and T_{X2}, correspond to the onset temperature of α-Fe phase and Fe-metalloid phase precipitated from the amorphous matrix, respectively [18]. The T_{X1} decreases with the increase of Cu content while the T_{X2} remains almost unchanged. Therefore, the largest interval between the T_{X1} and T_{X2} is can be seen in the $Fe_{83.5}B_{15}Cu_{1.5}$ alloy, which is beneficial for the formation of a two-phase composite structure without Fe-metalloid compounds precipitation. The effect of Cu on crystallization temperature can be easily understood by the fact that Cu clusters are generated prior to crystallization and stimulate nucleation of α-Fe phase [24,25].

The changes in B_S and H_C of the $Fe_{83.5}B_{15}Cu_{1.5}$ alloy as a function of annealing temperature T_a with $C = 1$ and 6 are shown in Fig. 3(a) and (b), respectively. The change trends in magnetic properties with T_a are generally the same regardless of the cyclic numbers. In Fig. 3(a), the B_S first increases remarkably with the increase of annealing temperature until 663 K and then remains nearly a constant at $T_a = 663$ K–743 K. From Fig. 3(b), the H_C slightly changes at $T_a = 603$ K–693 K, and then increases obviously when the annealing temperature increases further. One can see that the H_C of the non-cyclic annealed ribbons increases abruptly to more than 800 A/m at 713 K, which can be attributed to the excessive growth of α-Fe crystals. However, when the ribbons are cyclically annealed with $C = 6$, the H_C remains below 28 A/m in a large T_a range of 603 K–733 K. It is worth noting that, when the T_a exceeds 643 K, the B_S of the ribbons after rapid cyclic annealing is higher and H_C is lower than those of ribbons without cyclic annealing, which may be due to the assignable effect of cyclic annealing on the structural evolution of the $Fe_{83.5}B_{15}Cu_{1.5}$ alloy. Fig. 4(a) and (b) show the dependence of B_S and H_C of the $Fe_{83.5}B_{16.5}$ alloy on T_a with $C = 1$ and 6, respectively. A resemblance that the rapid cyclic annealing will optimize the soft magnetic properties and enlarge

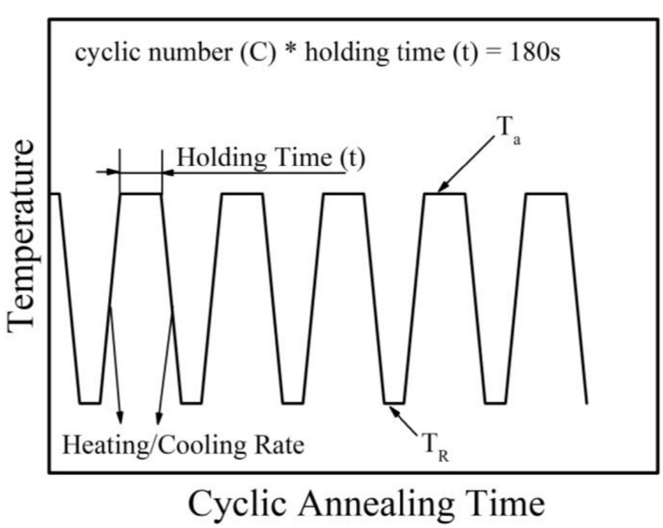

Fig. 1. Schematic diagram of rapid cyclic annealing.

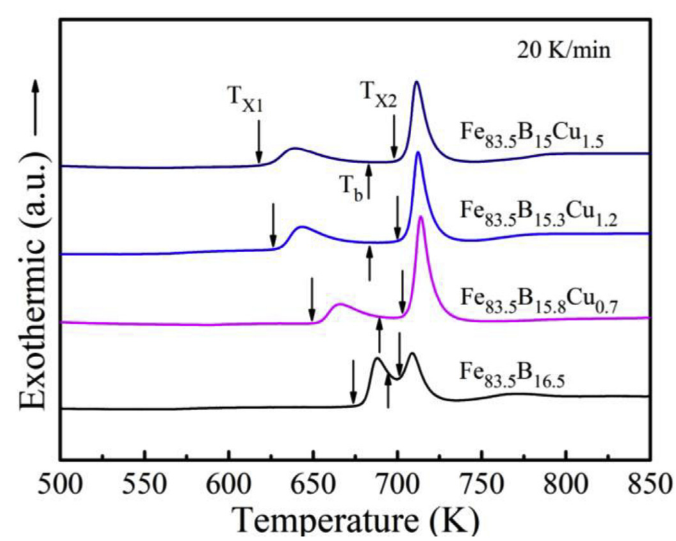

Fig. 2. DSC curves of the melt spun $Fe_{83.5}B_{16.5-x}Cu_x$ ($x = 0$, 0.7, 1.2, 1.5) alloy ribbons.