Magnetic Properties of $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ As-Quenched Ribbons

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$\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ nanocrystalline ribbons with appropriate Cu contents can be prepared without annealing processes. The $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons with lower Cu contents ($x \leq 1.5$) consist of amorphous structure. The nucleation of $\alpha\text{-Fe}(\text{Si})$ enhances with the increase of Cu content. An optimal Cu addition for obtaining the largest permeability, smallest saturation magnetostriction and largest magnetoimpedance occurs at $x \approx 3$. For as-quenched ribbons with high Cu contents ($x \geq 3.5$), soft magnetic properties deteriorate due to the drop of nucleation of $\alpha\text{-Fe}(\text{Si})$ and the precipitation of Fe-B phases. For as melt-spun $\text{Fe}_{71.5}\text{Cu}_3\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ ($x = 3$) nanocrystalline ribbon prepared with wheel speed of 40 ms^{-1} , the average grain-size of $\alpha\text{-Fe}(\text{Si})$ is about 10.5 nm, the initial permeability μ' is about 35000, the saturation magnetostriction λ_s is less than 1×10^{-6} , and the maximum value of magnetoimpedance $\Delta Z/Z_0$ under a field of 7162 A/m is -12.88% at 2 MHz. [doi:10.2320/matertrans.MRA2008058]

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1. Introduction

Fe-Cu-Nb-Si-B nanocrystalline ribbons are usually prepared by crystallization of the amorphous ribbons with annealing processes. It has been found that Fe-Cu-Nb-Si-B annealed ribbons show excellent soft magnetic properties, due to the microstructure consisting of nanometer-ordered $\alpha\text{-Fe}(\text{Si})$.¹⁾ Recently, we found that Fe-Cu-Nb-Si-B nanocrystalline ribbons with appropriate Cu contents can also be prepared by melt-spinning technique without annealing processes.²⁻⁴⁾ The Cu addition in as-quenched ribbons improves the nucleation of $\alpha\text{-Fe}(\text{Si})$ during rapid solidification,²⁻⁴⁾ similar to the case of annealed ribbons.⁵⁻⁷⁾ It was shown that Fe-Cu-T-Si-B (T = Nb, Mo, Ta, etc.) annealed ribbons have analogous microstructures with nanometer-ordered $\alpha\text{-Fe}(\text{Si})$, where the grain size was reduced most by Nb = Ta > Mo = W > V > Cr.⁸⁾ We have reported that as-quenched Fe-Cu-(Nb,V)-Si-B⁹⁾ and Fe-Cu-Mo-Si-B¹⁰⁾ nanocrystalline ribbons can be prepared by melt-spinning technique. In this way, it can be expected that Fe-Cu-Ta-Si-B nanocrystalline ribbons with appropriate Cu contents may be prepared without thermal-treatment. In order to confirm this assumption and explore new materials, in the present work, we prepared $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons and investigated their structure as well as magnetic properties.

2. Experiments

The alloys $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ ($x = 1, 1.5, 2, 2.5, 3, 3.5$ and 4) were obtained by arc-melting in an argon atmosphere. The alloy buttons were melted four times to ensure homogeneity. $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ ribbons of approximately 20–25 μm thickness were prepared by the single wheel melt-spinning technique with wheel speed of 40 ms^{-1} in an argon atmosphere. The X-ray diffraction (XRD) measurement of $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons was performed with Cu $K\alpha$ radiation. The perme-

ability of soft magnetic materials is usually measured with the toroidal shape sample. However, as-quenched nanocrystalline ribbons can't be rolled into toroids, as they are very brittle. The initial permeability of as-quenched ribbons was measured at 40 Hz and 0.8 A/m by the equivalent-impedance method using an impedance analyzer at room temperature, where four pieces of ribbons were used to form a closed magnetic path. The saturation magnetostriction λ_s at room temperature was measured in an applied magnetic field up to 1035 kA/m by the strain-gauge method. As-quenched ribbons with 2 mm width, 20 mm length were used in the magnetoimpedance measurement with a HP 4294A impedance analyzer at room temperature. The ac currents with an amplitude of 10 mA and dc magnetic fields were applied in the direction along the ribbon length for longitudinal magnetoimpedance measurement.

3. Results and Discussion

Figure 1 shows X-ray diffraction patterns for the $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons. The $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons with $x \leq 1.5$ consist of amorphous structure. With increasing Cu content in ribbons, nanometer-ordered $\alpha\text{-Fe}(\text{Si})$ appears. For $x = 2.5$, only (110) peak of $\alpha\text{-Fe}(\text{Si})$ appears, while for $x = 3$, the (110), (200) and (211) peaks of $\alpha\text{-Fe}(\text{Si})$ are evident. It is evident that an appropriate addition of Cu in $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ alloys can improve the nucleation of $\alpha\text{-Fe}(\text{Si})$ in as-quenched ribbons. The volume fraction of precipitated $\alpha\text{-Fe}(\text{Si})$ reaches a maximum at $x \approx 3$. Based on Scherrer's equation,¹¹⁾ the average grain-size of $\alpha\text{-Fe}(\text{Si})$ for as-quenched ribbon $x = 3$ was estimated as about 10.5 nm. As shown in Fig. 1, the (200) peak becomes weak for $x = 3.5$, the (200) and (211) peaks almost disappear for $x = 4$. The excess Cu addition reduces the volume fraction of precipitated $\alpha\text{-Fe}(\text{Si})$, but increases the volume fraction of remaining amorphous phase in the as-quenched ribbons. It has been found that by adding small amount of Cu and Nb in Fe-Si-B alloy, the crystallized microstructure of Fe-Cu-Nb-Si-B after annealing was drastically refined to a

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Fig. 1 The XRD patterns of $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons.

nanometer-scale dimension.¹⁾ The microstructural evolution in the nanocrystallization of $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$ was investigated by atom probe field ion microscopy (APFIM) and high resolution transmission electron microscopy (HRTEM),^{5,6)} and results indicated that Cu atoms form clusters prior to the onset of the crystallization reaction, which act as nucleation sites of $\alpha\text{-Fe(Si)}$, and stimulate the nucleation of the Fe-Si primary particle. With increasing Cu content, the amount of Cu clusters as nucleation sites of $\alpha\text{-Fe(Si)}$ increases,²⁻⁴⁾ improving the nanocrystallization of $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons. The phenomenon that the excess Cu addition reduces the volume fraction of precipitated $\alpha\text{-Fe(Si)}$ in the as-quenched ribbons is connected with the reduction of effective number density of nucleation sites for $\alpha\text{-Fe(Si)}$. Since Cu is virtually insoluble in Fe, Cu prefers to precipitate. With increasing Cu addition in alloys, there are two competitive tendencies in as-quenched ribbons: (1) the change of Cu cluster number; (2) the growth of Cu cluster. If the coarsening kinetics is very fast, the second tendency dominates, and the number of Cu cluster decreases correspondingly. That is to say, an optimal Cu content exists for improving the nucleation of $\alpha\text{-Fe(Si)}$. Similar case has been found in Fe-Cu-Mo-Si-B as-quenched ribbons.¹⁰⁾ In order to explain the optimal Cu contents for obtaining the highest permeability in annealed $\text{Fe}_{74.5-x}\text{Si}_{13.5}\text{B}_9\text{Nb}_3\text{Cu}_x$ ($x = 0, 0.5, 1$ and 1.5) nanocrystalline ribbons, the kinetics of copper clustering and primary crystallization have been studied by small-angle neutron scattering (SANS) and high-sensitivity differential scanning calorimetry (DSC).⁷⁾ Results showed that an optimal Cu content for obtaining the largest permeability was $x = 1$, corresponding to the largest effective number density of Cu clusters. The coarsening of the clusters occurs prior to the heterogeneous nucleation of Fe(Si) primary crystals. The excess Cu addition results in the coarsening of Cu clusters and the decrease of the effective number density of Cu clusters.⁷⁾

Fig. 2 The Cu content dependence of the initial permeability μ' for $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons.

Table 1 Values of initial permeability μ' and saturation magnetostriction λ_s for $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons.

X	1	1.5	2	2.5	3	3.5	4
μ'	4000	7000	11000	15000	35000	10000	6000
$\lambda_s (\times 10^{-6})$	4	2	1	<1	<1	1	2

In addition, the precipitation of Fe_{23}B_6 and Fe_3B phases can be observed in the as-quenched ribbons with high Cu contents (see Fig. 1). Fe-B phases usually occur for $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$ ribbons annealed at high temperatures.⁸⁾

Figure 2 shows the initial permeability μ' dependence on the Cu content for $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons. The value of initial permeability for amorphous $\text{Fe}_{73.5}\text{Cu}_1\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ ($x = 1$) is only 4000. With increasing Cu content, μ' of as-quenched ribbons increases evidently, due to the increase of volume fraction of $\alpha\text{-Fe(Si)}$. The initial permeability μ' experiences a peak value of about 35000 at $x = 3$ ($\text{Fe}_{71.5}\text{Cu}_3\text{Ta}_3\text{Si}_{13.5}\text{B}_9$). With further increasing Cu content, the value of μ' decreases. For example, the value of initial permeability μ' is only 10000 for $x = 3.5$. There may be two mechanisms responsible for the low permeability of the as-quenched ribbons with high Cu contents: (1) the reduction of nucleation of $\alpha\text{-Fe(Si)}$; (2) the precipitation of Fe-B phases which result in the hardening of ribbons. Values of initial permeability μ' and saturation magnetostriction λ_s for $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons are also listed in Table 1. The smallest value (less than 1×10^{-6}) of λ_s also occurs at about 2.5–3, corresponding to the optimal Cu content with the largest value of permeability.

The magnetoimpedance $\Delta Z/Z_0 = (Z(H) - Z(0))/Z(0)$, magnetoresistance $\Delta R/R_0 = (R(H) - R(0))/R(0)$ and magnetoreactance $\Delta X/X_0 = (X(H) - X(0))/X(0)$ under $H = 7162 \text{ A/m}$ for $\text{Fe}_{71.5}\text{Cu}_3\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ ($x = 3$) as-quenched ribbons are shown in Fig. 3, here $Z(0)$, $R(0)$, $X(0)$ and $Z(H)$, $R(H)$ and $X(H)$ are values of impedance, resistance and reactance with fields of zero and H , respectively. There is a large magnetoreactance ($\Delta X/X_0 = -56.67\%$) at low frequency $f = 50 \text{ kHz}$. The magnetoresistance $\Delta R/R_0$ has a peak value of -18.16% at $f = 5 \text{ MHz}$, whereas magnetoimpedance $\Delta Z/Z_0$ experiences a maximum value of -12.88% at $f = 2 \text{ MHz}$. Similar case has been found in

Fig. 3 The magnetoimpedance $\Delta Z/Z_0$, magnetoresistance $\Delta R/R_0$ and magnetoreactance $\Delta X/X_0$ A/m for as-quenched $\text{Fe}_{71.5}\text{Cu}_3\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ ($x = 3$) under $H = 7162$ A/m.

Fig. 5 The Cu content dependence of maximum magnetoimpedance $(\Delta Z/Z_0)_{\max}$ for $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons under $H = 7162$ A/m.

Fig. 4 The frequency dependence of magnetoimpedance $\Delta Z/Z_0$ for $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons under $H = 7162$ A/m.

annealed Fe-Zr-B and Fe-Zr-B-Al nanocrystalline ribbons.¹²⁾ The frequency dependence of magnetoimpedance $\Delta Z/Z_0$ under $H = 7162$ A/m for $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons is shown in Fig. 4. The maximum magnetoimpedance $(\Delta Z/Z_0)_{\max}$ can be derived from Fig. 4 and its Cu content dependence is plotted in Fig. 5. An optimal Cu addition for obtaining largest magnetoimpedance occurs at $x = 3$, which is consistent with the results of permeability and magnetostriction.

4. Conclusions

Fe-Cu-Ta-Si-B nanocrystalline ribbons with appropriate Cu contents can be prepared without annealing processes. For $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons, the nucleation of $\alpha\text{-Fe}(\text{Si})$ enhances with the increase of Cu content. An optimal Cu content for obtaining the largest permeability, smallest saturation magnetostriction and largest magneto-

impedance is about $x = 3$. The initial permeability $\mu' = 35000$ and the saturation magnetostriction $\lambda_s < 1 \times 10^{-6}$ can be obtained for as-quenched $\text{Fe}_{71.5}\text{Cu}_3\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ ($x = 3$) nanocrystalline ribbon prepared with wheel speed of 40 ms^{-1} , whose average grain-size of $\alpha\text{-Fe}(\text{Si})$ is about 10.5 nm . With further increasing Cu content ($x \geq 3.5$) in $\text{Fe}_{74.5-x}\text{Cu}_x\text{Ta}_3\text{Si}_{13.5}\text{B}_9$ as-quenched ribbons, soft magnetic properties deteriorate due to the drop of nucleation of $\alpha\text{-Fe}(\text{Si})$ and the precipitation of Fe-B phases.

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