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Effect of annealing treatments on the microwave electromagnetic properties of amorphous FeCuNbSiB microwires*

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The amorphous FeCuNbSiB microwires are fabricated by using the melt extraction method and annealed separately at temperatures T = 573, 673, 723 and 773 K for 1 h. The effect of annealing treatment on the microwave electromagnetic properties of FeCuNbSiB wires/wax composites has been investigated for the first time. It is found that in a frequency range of 0.5–4.0 GHz, the complex permittivity, permeability, magnetic and electric loss tangents of FeCuNbSiB wires/wax composites are strongly dependent on the annealing temperature and frequency. For T = 573, 723 and 773 K, two resonance peaks are found at frequency f = 1.2 and 3.3 GHz. However, for T = 673 K, only one resonance peak occurs at f = 3.3 GHz. The resonance peak at f = 1.2 GHz is believed to be due to the stress-induced anisotropy, while the resonance peak at f = 3.3 GHz is attributed to the random anisotropy.

Keywords: amorphous alloys, permeability, permittivity, microwires
PACC: 7540G, 7550K, 7720, 3240

1. Introduction

Fe-based amorphous and nanocrystalline wires have been widely reported due to their giant magnetoimpedance (GMI) effect,[1] and employed mainly in sensor applications, such as magnetic field sensor,[2] position sensor *etc*.[3] Usually the reported methods of fabricating magnetic microwires include melt-extraction method,[4] Taylor–Ulitovsky method,[5] Taylor method of producing glass-coated wire,[6] the in-rotating-water quenching technique.[7] Different fabrication methods result in different domain configurations in wires,[8] and the rapid cooling rate during wire fabrication also can result in a complex stress distribution in microwires.[6] Recently, there has been an ever increasing interest in studying the electromagnetic (EM) wave absorbers due to their applications in anti-EM interference coating, microwave darkroom.[9−12] According to EM energy loss mechanisms, EM wave absorbers can be divided into three categories: dielectric loss, electronic loss and magnetic loss. For the magnetic loss, EM energy can be dissipated by magnetic hysteresis loss, domain wall resonance absorption, and spin resonance absorption. The usually adopted magnetic materials for EM wave absorber are ferrite (spinnel ferrites and hexaferrites), magnetic particles, polycrystalline Fe fibres, Co fibre/epoxy resin composite, etc. Ferromagnetic nanocomposites are often used in high frequency signal and high power electronic devices. FeCuNbSiB nanocomposite is an excellent soft magnetic material with the Snoek's limit in GHz range.[13] To our knowledge, the possible application of FeCuNbSiB in EM wave absorber has not been explored yet, especially in the form of amorphous microwires.

In this paper, we investigate the microwave properties of amorphous FeCuNbSiB magnetic microwires annealed at different temperatures.

2. Experimental details

The FINEMET® (with standard alloy composition $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$) microwires are fabricated by using the melt extraction method. A differential

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scanning calorimeter (DSC) curve is obtained on a differential scanning calorimeter at a heating rate of 10 K/min to find the crystallization temperatures of amorphous wires. The x-ray diffraction (XRD) is used to check the amorphous state for the prepared microwires. Annealing treatments of magnetic wires under argon atmosphere for 1 h are carried out at different temperatures: 573, 673, 723 and 773 K separately. The heat treated microwires are cut into 1-mm long, and homogenously mixed with wax (microwires / wax volume ratio is about 1:2), then the mixture is made into a toroidal shape sample for measuring the complex relative permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) and the complex relative permeability ($\mu_r = \mu' - j\mu''$) by using an Agilent 8720 ET network analyser within the frequency range of 0.5–4.0 GHz.

3. Results and discussion

The fabrication of amorphous FeCuNbSiB microwires is schematically shown in Fig.1(a). The tip edge of rotating copper wheel cuts the melted alloy and draws it into a microwire in an argon atmosphere. The rotation speed of wheel can be used to control the cooling rate to obtain amorphous microwires. The cooling rate is about 10^6 K/s. The as-prepared microwires are shown in Fig.1(b). The average diameter of wires is about 50μm.

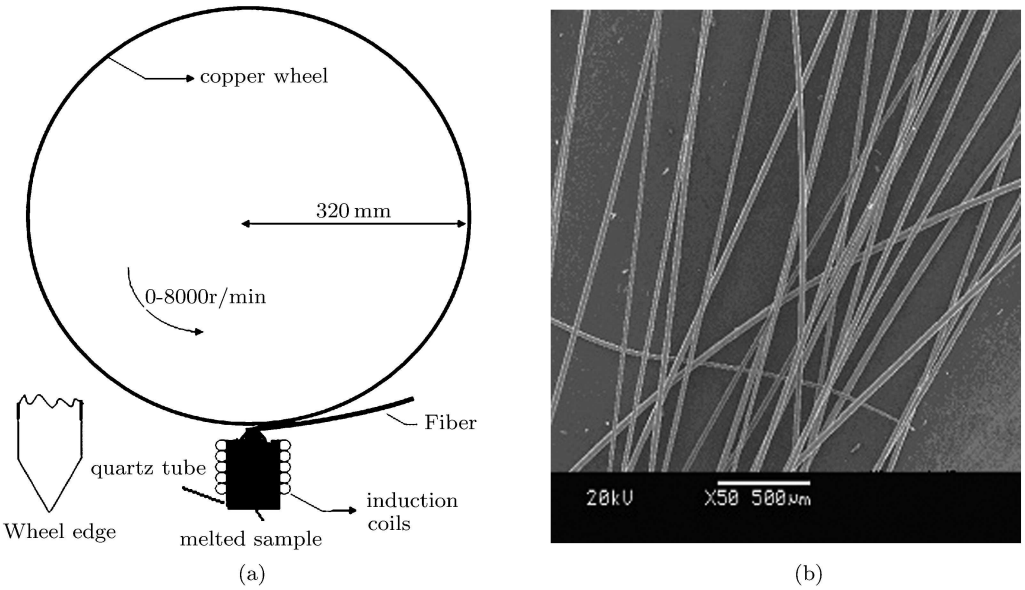


Fig.1. (a) Preparation of amorphous FeCuNbSiB microwires. (b) as-prepared amorphous FeCuNbSiB magnetic wires.

The XRD pattern of as-prepared microwires is shown in Fig.2. The figure shows a typical pattern of amorphous alloys, indicating that the as-prepared microwires are in an amorphous state. The broad peak centred at $2\theta = 45°$ confirms the characteristics for the short-range order of the Fe-based amorphous sample.

In order to find the crystallization temperatures for amorphous microwires, the DSC measurement is carried out as shown in Fig.3. As pointed out in Ref.[14], for FeCuNbSiB alloys, there exist two crystallization temperatures, which is also confirmed in Fig.3. Our DSC measurement reveals that the primary crystallization peak temperature (T_1) is 812 K and the secondary crystallization peak temperature (T_2) is 952 K. These two crystallization temperatures are close to those found in a similar alloy composition $Fe_{72}Cu_1Nb_{4.5}Si_{13.5}B_9$ ($T_1 = 815$ K and $T_2 = 961$ K) in Ref.[14]. According to Ref.[14], at the primary crystallization temperature, nanocrystalline α-Fe (Si) grains are formed in the amorphous matrix. At the secondary crystallization temperature, nanocrystalline Fe- and/or Nb-borides are formed. Similar results have also been reported in Ref.[15]. Therefore, our annealing treatments below the primary crystallization temperature will have an effect on the test samples, resulting in reduction of internal stress, which has formed by the ultra-fast cooling during the preparation of amorphous microwires.

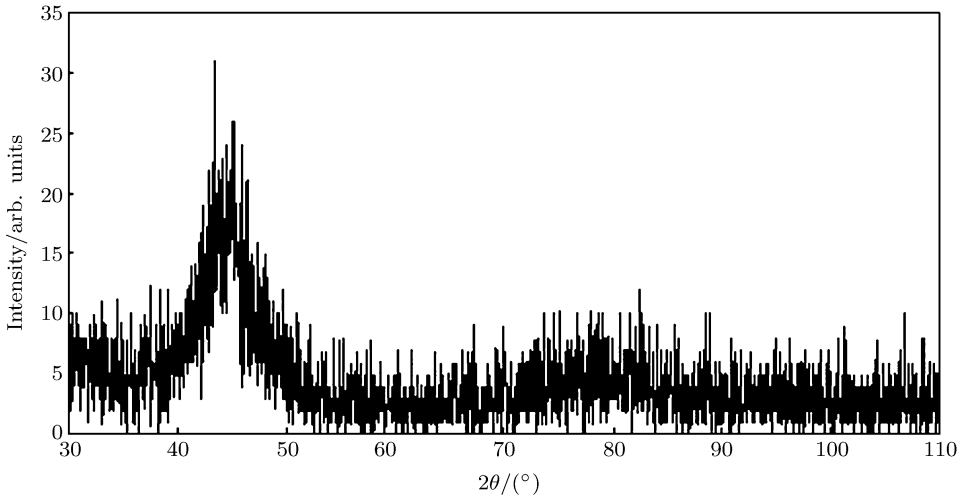


Fig.2. XRD pattern showing amorphous state for the as-prepared microwires

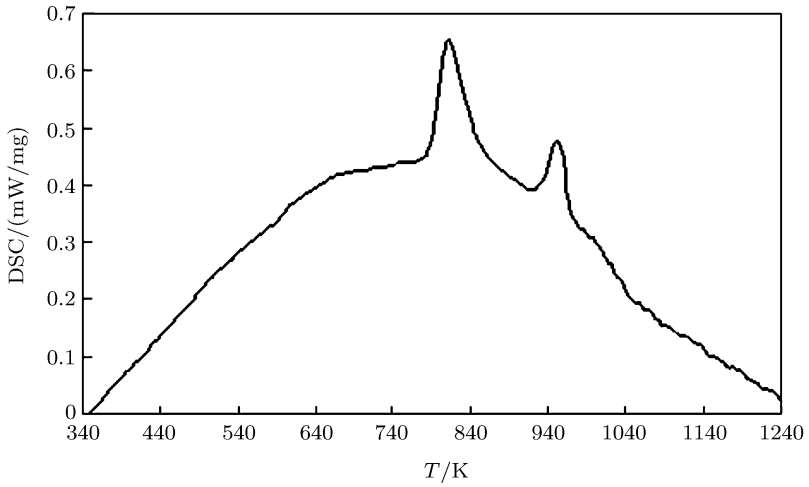


Fig.3. DSC curves of as-prepared FeCuNbSiB microwires.

Figure 4(a) and (b) show the complex relative permittivity of FeCuNbSiB/wax composites in the frequency range of 0.5–4.0 GHz. Figure 4(a) shows the real parts (ε'), and Fig.4(b) shows the imaginary parts (ε'') of the relative permittivity. For ε' values, at T = 573 and 723 K, ε' values do not vary significantly within the measurement frequency range. At T = 773 K, it has a maximum value at f = 2 GHz; while for T = 673 K, ε' has a minimum value at f = 2 GHz and have two peaks at f = 0.8 and 3.6 GHz. At T = 723 and 773 K, ε'' decreases with increasing the frequency, showing a relaxation feature. However, for T = 573 and 673 K, ε'' increases with increasing frequency. Also, the close examination of Fig.4(b) can find that in the frequency range 0.5–3.0 GHz, ε'' is much larger for T = 723 and 773 K than for T = 573 and T = 673 K. The dependence of the electric loss tangent ($\tan\delta = \varepsilon''/\varepsilon'$) on frequency and annealing temperatures is shown in Fig.4(c). As shown in the figure, the electric loss tangent is strongly dependent on the frequency and annealing temperature. With increasing annealing temperature, the electric loss tangent value increases too. Especially, in the lower frequency region, the effect of annealing temperature on the electric loss is significant.

Figure 5(a) and (b) show the complex relative permeability in the selected GHz region. With increasing frequency, μ' decreases for T = 573, 673 and 723 K, while for T = 773 K, there is a peak at f = 0.85 GHz. As for μ'', at the lower end of the selected frequency band, the difference in μ'' value is significant, especially for T = 673 K and T = 773 K. It should be noted that there are two peaks at f = 1.2 GHz and 3.3 GHz in the μ''–f curve for the samples annealed at T = 573, 723, 773 K. However, for T = 673 K, only one peak occurs at f = 3.3 GHz. The dependence of the

magnetic loss tangent ($\tan\delta = \mu''/\mu'$) on frequency and annealing temperature is shown in Fig.5(c). For annealing treatments at $T = 573$, 673, 723 and 773 K, there is a magnetic loss peak at $f = 3.3$ GHz, while for samples annealed at $T = 573$, 723 and 773 K, another magnetic loss tangent peak is found at $f = 1.2$ GHz.

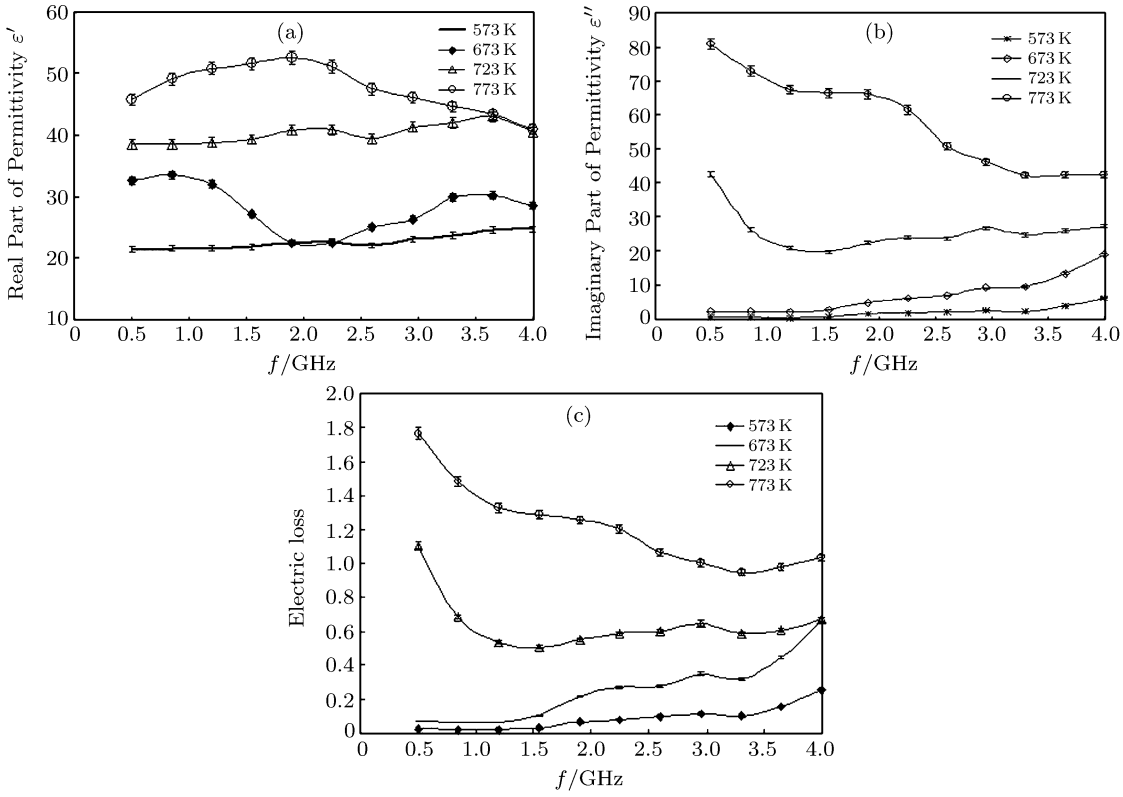


Fig.4. Frequency dependences of real part of permittivity ε' (a), imaginary part of permittivity ε'' (b), and electric loss tangent ($\tan\theta = \varepsilon''\varepsilon'$) (c) for amorphous microwires annealed at different temperatures.

Anomalous behaviours of permeability are possibly due to the effect of annealing treatment on the magnetic properties of FeCuNbSiB wires. Here, the annealing treatment below the primary temperature ($T = 812$ K) can result in the following effects. (1) It reduces the residual stress formed by rapid cooling during the fabrication of wires; According to Ref.[16], the internal stress for the as-prepared amorphous FINEMET®($Fe_{73.5}Cu_1Nb_3Si_{16.5}B_6$) ribbon ($70 \times 2.5 \times 0.034 mm^3$) is about 15 MPa. While at $T = 853$ K, the internal stress is 0.2 MPa. Although in this paper, the FINEMET® alloy is prepared into the form of microwire, both the magnitude and distribution of internal stress might be different from those in ribbon. However, through the findings, we can learn that the internal stress is highly sensitive to annealing treatment for the FINEMET® type amorphous alloys, so are their magnetic properties. (2) The annealing treatments can change the saturation magnetostriction coefficient λ_s of the wires. The λ_s value of the amorphous FINEMET® alloy is about 8 ppm,[14] so the magnetoelastic energy can play an important role in the stress-induced anisotropy and the permeability of the samples studied. Therefore, the dependence of permeability on the annealing temperature in Fig.5 is due to the relief of internal stress and the variation of magnetostriction in amorphous microwire. The magnetic loss in GHz range is believed to be due to the resonance absorption. The resonance frequency of magnetization rotation under the action of anisotropy field, with no external static magnetic field applied, can be simply expressed as $2\pi f_r = vH_a$, where v is the gyromagnetic ratio, f_r is the resonance frequency, and H_a is the anisotropy field.[17] From Fig.5(c), the magnetic loss peaks at $f = 1.2$ GHz are sensitive to the annealing temperature, especially at $T = 673$ K, the resonance peak disappears, while for the peaks $f = 3.3$ GHz, the magnetic loss peaks are maintained for different annealing temperatures. It can be inferred that the magnetic loss peaks

at $f = 1.2\,\mathrm{GHz}$ might be due to the stress-induced anisotropy, the magnetic loss peaks at $f = 3.3\,\mathrm{GHz}$ might be attributed to the anisotropy that is insensitive to the annealing temperature, such as the local anisotropy (or random anisotropy), which is effective in amorphous alloys on a scale of a few nanometres.[18]

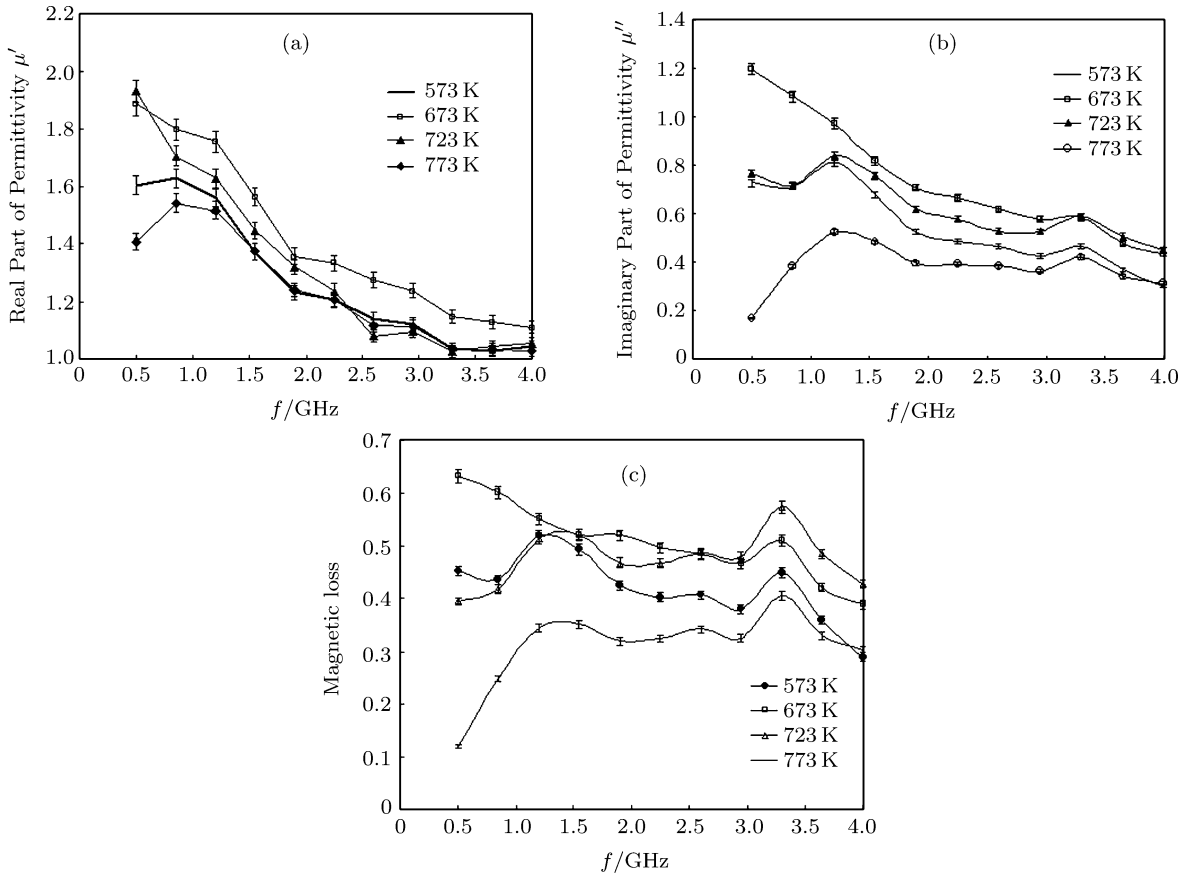


Fig.5. Frequency dependences of μ' (a), μ'' (b), and magnetic loss tangent ($\tan\theta = \mu''\mu'$) (c) for amorphous microwires annealed at different temperatures.

4. Conclusions

The amorphous FeCuNbSiB microwires are fabricated with an average diameter of $50\,\mu\mathrm{m}$. It is the first time the microwave properties (the complex relative permittivity and permeability) of the annealed FeCuNbSiB nanocomposite microwire/wax composites have been investigated. Our results reveal that ε_r, μ_r, electric loss tangent and magnetic loss tangent are strongly dependent on the annealing temperature and frequency in GHz range. The frequency spectra of permittivity and permeability show that FeCuNbSiB amorphous wires might be used for EM wave absorption if their properties are improved.

References

[1] Sanchez M L, Prida V M, Hernando B, Kurlyandskaya G V, Santos J D and Vazquez M 2002 *Chin. Phys. Lett.* **19** 1870

[2] Mahdi A, Panina L and Mapps D 2003 *Sens. Act.* A **105** 271

[3] Han M, Liang D F and Deng L J 2005 *J. Mater. Sci.* **40** 5573

[4] Rudkowski P, Rudkowka G and Stromolsen J 1991 *Mater. Sci. Eng.* A **133** 158

[5] *USSR patent,* No.128427

[6] Chiriac H and Ovari T 1996 *Prog. Mater. Sci.* **40** 333

[7] Ohnaka I 1980 *Japan Patent,* No. 64948

[8] Mohri K, Humphrey F, Kawashima K, Kimura K and Mizutani M 1990 *IEEE Trans. Magn.* **26** 1789

[9] Deng L W, Jiang J J, Fan S C, Xie W Y, Zhang X C and He H H 2003 *J. Magn. Magn. Mater.* **264** 50

[10] Jiu Rong Liu and Masahiro Itoh 2003 *Appl. Phys. Lett.* **83** 4017
[11] Sugimoto S, Maeda T, Book D, Kagotani T, Inomata K and Homma M 2002 *J. Alloys Compounds* **301** 330
[12] Yusoff A and Abdullah M 2004 *J. Magn. Magn. Mater.* **269** 271
[13] Mazaleyrat F and Varga L 2000 *J. Magn. Magn. Mater.* **215–216** 253
[14] McHenry M, Willard M and Laughlin D 1999 *Prog. Mater. Sci.* **44** 291
[15] Lin D M, Wang H S, Wu Y C and Lin M L 1999 *Chin. Phys.* **8** 455
[16] Carara M, Baibich M and Sommer R 2002 *J. Appl. Phys.* **91** 8441
[17] Chikazumi S 1997 in *Physics of Ferromagnetism* (2nd edition), Clarendon, Oxford
[18] O'Handley R 2000 *Modern Magnetic Materials: Principles and Applications,* Wiley, New York