Vol 18 No 3, March 2009
1674-1056/2009/18(03)/1261-05
Chinese Physics B
© 2009 Chin. Phys. Soc.
and IOP Publishing Ltd

Annealing effects on the microwave permittivity and permeability properties of $Fe_{79}Si_{16}B_5$ microwires and their microwave absorption performances*

Han Man-Gui(韩满贵)†, Ou Yu (欧 雨), Liang Di-Fei (梁迪飞), and Deng Long-Jiang(邓龙江)

State Key Laboratory of Electronic Thin Films and Integrated Devices,
University of Electronic Science and Technology of China, Chengdu 610054, *China*

(Received 6 July 2008; revised manuscript received 19 August 2008)

This paper reports that amorphous magnetic microwires ($Fe_{79}Si_{16}B_5$) have been fabricated by a melt-extraction technique and have been annealed at 600°C and 750°C respectively. Differential scanning calorimeter measurements show that nanocrystalline magnetic phase (α-Fe) has been formed in the amorphous matrix when it was annealed at 600°C. Hard magnetic phase (Fe_2B) was formed in the microwires annealed at 750°C, which increases the magnetic coercivity. Microwave permittivity and permeability are found to be dependent on the microstructures. The permittivity fitting results show that multi Lorentzian dispersion processes exist. For microwires annealed at 750°C , their resonance peaks due to the domain wall movements and natural resonance are found higher than those of microwires annealed at 600°C. The microwave absorption performance of microwires annealed at 600°C is found better than microwires annealed at 750°C.

Keywords: amorphous magnetic microwires, magnetic permeability, permittivity, microwave absorber

PACC: 7540G, 7550K, 7720, 3240

1. Introduction

Fe-based amorphous and nanocrystalline wires have been widely reported for their giant magnetoimpedance (GMI) effect, and have been mainly exploited for sensor developments, such as magnetic field sensors, position sensors etc.[1,2] Recently, there was an ever increasing interest in studying their electromagnetic (EM) wave absorption properties due to their potential applications in anti-EM interference coatings, microwave darkrooms.[3,4] It is well known that Fe-based nanocrystalline materials can be fabricated by properly annealing amorphous Fe–Si–B alloy: nano scale α-Fe grains are embedded in an amorphous matrix.[5] However, to our best of knowledge, the possible application of Fe–Si–B nanocomposite in EM wave absorber has not been well exploited yet, especially in the forms of microwires. In this paper, we report the annealing effect on the microwave permittivity and permeability properties of microwires, and then compare their microwave absorption performances.

2. Experimental details

$Fe_{79}Si_{16}B_5$ microwires were fabricated by the melt extract method. Detailed manufacture technique has been reported elsewhere.[4] Annealing treatments on magnetic wires under argon atmosphere for 1 hour were carried out at 600°C and 750°C respectively. x-ray diffraction (XRD) measurements with Cu Kα radiation were carried out to study the effect of annealing on the phase transformations. The differential scanning calorimeter (DSC) measurements with a heating rate of 10°C/min were conducted to find the crystallization temperatures for microwires. The M–H hysteresis loops were measured for the heat treated microwires on a vibrating sample magnetometer (VSM). It was found that the as-prepared microwires were very flexible and can not be easily cut into short pieces. After annealing treatments, microwires became brit-

*Project supported by the National Natural Science Foundation of China (Grant No 60701016) and the Science Foundation for Young Faculties of UESTC (Grant Nos L08010301JX0618 and L08010301JX05013).
†Corresponding author. E-mail: mangui@gmail.com

tle, and the heat treated microwires were cut into pieces with the average length of 1 mm, and were homogenously mixed with wax (the microwires / wax weight ratio is about 2:1), then the complex relative permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) and the complex relative permeability ($\mu_r = \mu' - j\mu''$) were measured on an Agilent 8720 ET Network Analyser within a frequency range of 1 GHz–18 GHz. The permittivity and permeability dispersion spectra have been fitted by using a nonlinear least mean square technique to uncover the underlying physics.

3. Results and discussions

The morphology of as-prepared microwires with an average diameter of 33 μm is shown in Fig.1(a). The DSC curve is shown in Fig.1(b), and two exothermic peaks are found at 532.1°C and 665.1°C. An alloy with a similar composition ($Fe_{77.5}Si_{13.5}B_9$) was reported to have two exothermic peaks, one was at 535°C and the other one was at 550°C.[4] Their first nanocrystalline temperature is close to ours, but the

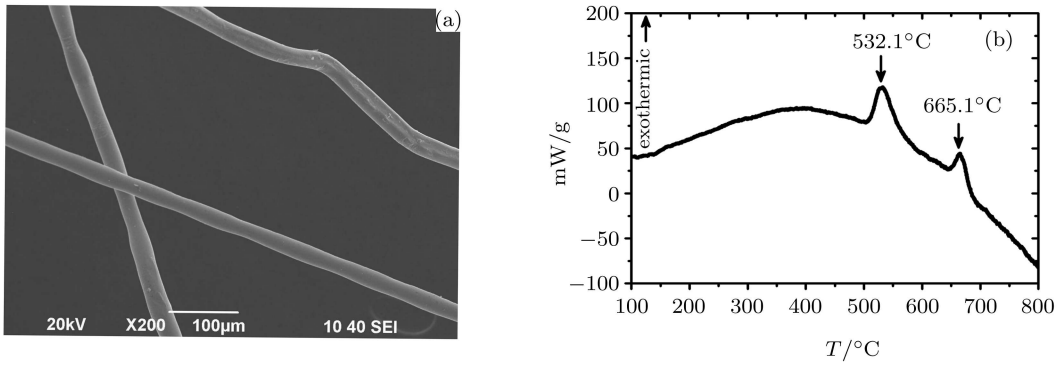


Fig.1. (a) SEM images of as-prepared microwires, (b) DSC curve.

second one is lower than ours. We think it might be due to the different compositions. The first peak in our DSC curve is believed due to the formation of α-Fe nanocrystalline grains. The second peak is due to the formation of a hard magnetic boride (Fe_2B). To verify this, we have carried out the XRD measurements on the annealed microwires. As shown in Fig.2, for the as-prepared microwires, a broad diffraction peak is found around 45°, which is a typical characteristic of amorphous Fe-based alloys (such as $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy),[5,6] and indicates that the as-prepared microwires are in the amorphous state. The broad peak centred at $2\theta = 45°$ confirms the characteristics of short-range order of Fe atoms in the Fe-based amorphous alloys. After the microwires have been heat treated at 600°C, nanocrystalline α-Fe grains have been formed, as the diffraction peaks show. The average grain size can be estimated to be about 14 nm using the Scherrer's formula: $d = 0.9\lambda/(B_\theta \cos\theta_B)$, where λ is the wavelength of x-ray, B_θ is the full width at half maximum; θ_B is the centre of a diffraction peak.[5] When the microwires were heat treated at 750°C, it is shown in Fig.2 that the Fe_2B borides have been precipitated. The average size of nanocrystalline grains increase to 25.5 nm.

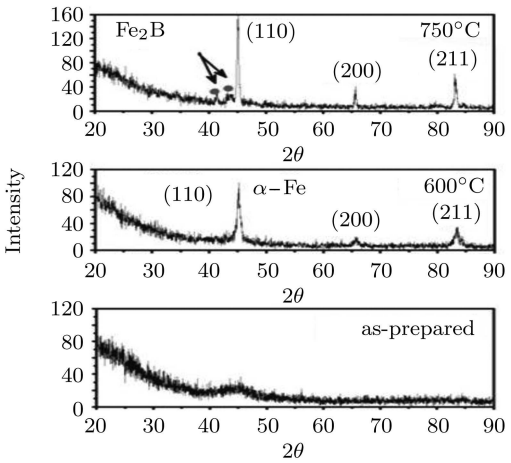


Fig.2. XRD patterns of microwires.

The magnetic hysteresis ($M - H$) loops of randomly oriented microwires are shown in Fig.3. Both the as-prepared microwires and the microwires annealed at 600°C show good soft magnetic properties. Their coercivity values are about 5 Oe (1 Oe = 79.5775 A/m). Their saturation magnetization values (emu/g) are 109 and 104, respectively. However, the coercivity of microwires annealed at 750°C is about 110 Oe, its larger coercivity is believed due to the forma-

tion of hard magnetic phase Fe_2B. For the microwires annealed at 750°C, the saturation magnetization is 104 emu/g.

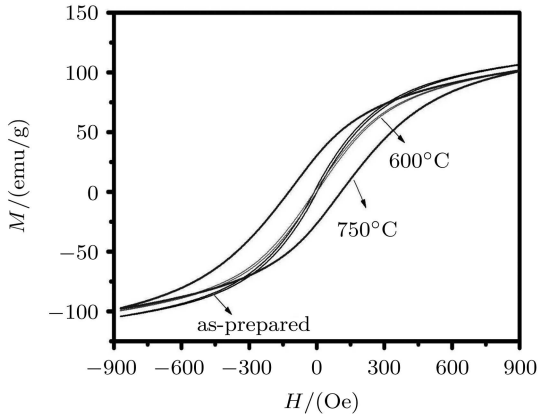


Fig.3. Magnetic hysteresis loop of randomly oriented microwires.

$$\varepsilon(f) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 - (f/f_\mathrm{r})^2 + \mathrm{i}(f/f_\mathrm{d})}. \qquad (1)$$

The microwave permittivity dispersion spectra are shown in Fig.4. As shown, the large permittivity values (for instance, for microwires annealed at 750°C, $\varepsilon' = 45$ at 1 GHz) might be due to the size increase of nanocrystalline grains. Because the grain boundaries are the polarization (P) sites under the external electric field (E), and the relationship of $\varepsilon = P/E$ holds, then increase of grain size will result in the increase of P because P is proportional to the separation distance of dipoles. The permittivity of microwires heat treated at 750°C is larger than those heat treated at 600°C at lower frequency (such as $f = 1 - 3$ GHz). Considering only one permittivity dispersion process (single Lorentzian dispersion law (LDL)), we can employ the LDL (see Eq.(1)) to fit the real parts of permittivity spectra.[7] In Eq.(1), ε_0, ε_∞ are the static permittivity and the permittivity at infinite frequency respectively. f_d is the Debye relaxation frequency. f_r is the natural resonance frequency. The fitting results in Fig.4 show that the fitting curves and the experimental curves agree well at low frequency and not well at high frequency range. In order to obtain better fittings at high frequency range, multi Lorentzian dispersion process should be considered. As shown in Fig.4, multi LDL fittings show good agreement with the experimental results, which indicate that multi Lorentzian dispersion behaviours play major roles in determining the permittivity dispersion. For microwires heat treated at 600°C, the fitted values of ε_0, ε_∞, f_d and f_r are 35.183, 22.065, 8.03 GHz and 3.77 GHz respectively. For microwires heat treated at 750°C, the fitted values of ε_0, ε_∞, f_d and f_r are 49.0, 23.326, 3.79 GHz and 4.62 GHz respectively.

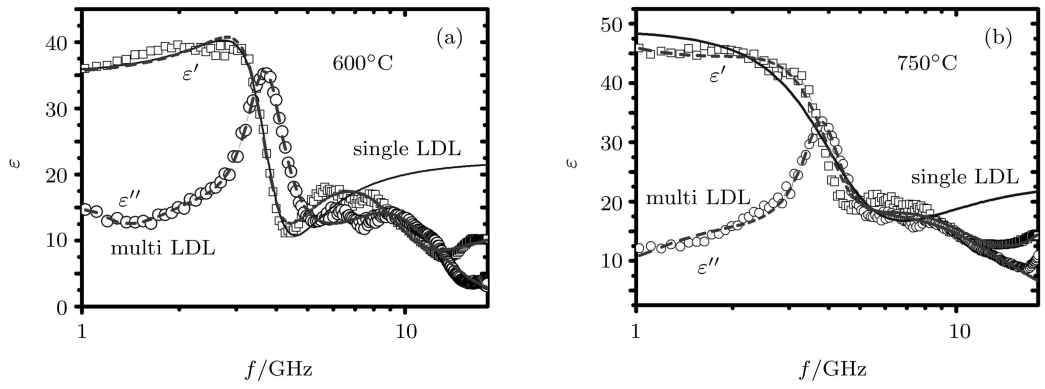


Fig.4. Microwave permittivity dispersion spectra of microwires annealed at different temperatures: (a) 600°C, (b) 750°C. The dotted and solid lines are fitting curves. The open symbols (squares and circles) are experimental data. Single LDL means that fitting is based on single Lorentzian dispersion process (solid lines). Multi LDL means that fitting is based on multiple Lorentzian dispersion processes (dotted lines).

The permeability spectra of the annealed microwires are shown in Figs.5(a) and 5(b). Interestingly, there are two distinct dispersion peaks found in each spectrum of $\mu' - f$. Moreover, the dispersion peak due to the domain wall movement (DWM) mechanism is usually lower than that due to the natural resonance (NR) mechanism. Therefore, we suppose that the peak at lower frequency is due to DWM, the other one is due to the NR. As shown, the resonance peaks have been found in the $\mu' - f$ spectra, the Debye dispersion law does not apply because of its relaxation characteristic. Therefore, we employed the resonance

- type permeability dispersion law with damping constant to fit the permeability dispersion spectra,[8] as shown in Eq.(2),

$$\mu_{\rm r} = 1 + \frac{\chi_{0j}(1 + {\rm i}\lambda_j f/f_{{\rm r}j})}{(1 + {\rm i}\lambda_j f/f_{{\rm r}j})^2 - (f/f_{{\rm r}j})^2}, \qquad (2)$$

where j =w, s denotes that the resonance peak due to DWM component or NR component. The λ_j denotes the damping constant for each component. The $f_{{\rm r}j}$ means the resonance frequency of each component. Employing Eq.(2), we found that separate fittings for each dispersion peak can show better agreements with the experimental data than the fittings considering both components simultaneously, as shown by the dotted lines in Figs.5(a) and 5(b). The fitting curves labelled as 'combined' mean that the fittings are carried out by considering the DWM and NR simultaneously. The parameters fitted are listed in Table 1. As indicated in Table 1, for each dispersion contribution component, all the parameters (χ_0, λ, $f_{\rm r}$) are larger for microwires heat treated at 750°C than those at 600°C, it shows that the annealing treatments have a significant effect on the microwave permeability properties. For instance, both the resonance frequencies due to the DWM mechanism and NR mechanism are larger for the microwires annealed at 750°C, we attribute this finding to the effect of annealing on the microstructures. Such as, the hard magnetic phase (Fe_2B) has been formed in the microwires annealed at 750°C, which will increase the anisotropy field for the NR mechanism. For this mechanism, its anisotropy field is proportional to the coercivity, and it is well known that the coercivity is a microstructure sensitive parameter.[9] For DWM mechanism, the resonance frequency can be described as $f_{\rm dw} = (9\pi\xi\rho)/(8a\mu_0\,{}^2M_{\rm s}^2)$, where '$\xi$' is the energy density of domain wall, 'ρ' is the resistivity and 'a' is the average distance of non magnetic impurities (or hard mantic phases) in a magnetic material, which is also microstructure-sensitive parameter.[10] The formation and distribution of hard magnetic Fe_2B are believed to be responsible for the "a" values and "$f_{\rm dw}$" values. Consequently, as shown in Table 1, the variation of damping constants and resonance frequencies are ascribed to the annealing which alters the microstructures of microwires.

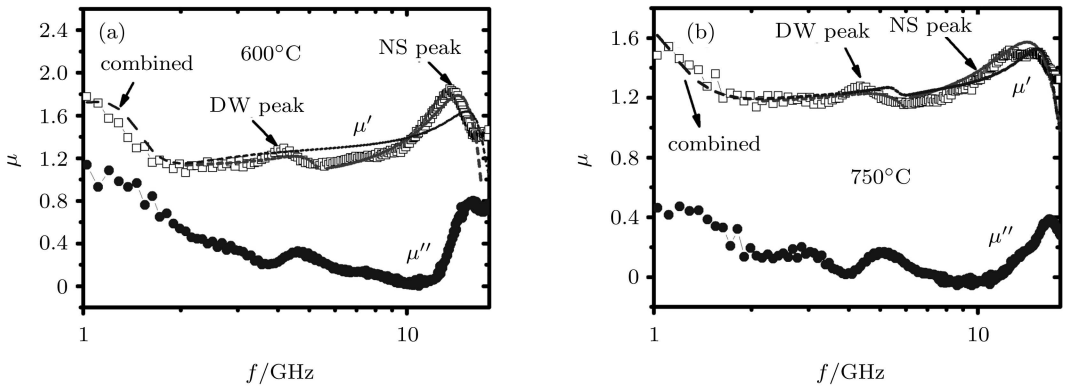


Fig.5. Microwave permeability spectra: (a) the real part and the imaginary part of microwires heat treated at 600°C; (b) the real part and the imaginary part of microwires heat treated at 750°C. Dotted lines are the fitting curves.

Table 1. Fitting results of domain wall movement components and natural resonance components for microwires heat treated at 600°C and 750°C.

(°C)	domain wall movement			natural resonance		
	$\chi_{0{\rm w}}$	$\lambda_{\rm w}$	$f_{\rm rw}$/GHz	$\chi_{0{\rm s}}$	$\lambda_{\rm s}$	$f_{\rm rs}$/GHz
600	0.119	0.1944	5.58	1.002	0.203	18.10
750	0.1623	0.3008	6.59	1.100	0.293	19.99

With the measured permittivity and permeability values, if the EM wave is normally incident on an absorber attached on a metal plate, then the EM wave absorbing performances can be evaluated by using the reflection loss (RL) in a dB unit. Here, we evaluate their EM wave absorption performances based on the following equations:[11]

$$Z_{\rm in} = Z_0\sqrt{(\mu_{\rm r}/\varepsilon_{\rm r})}\tanh\{{\rm j}(2\pi ft/c)\sqrt{\mu_{\rm r}\varepsilon_{\rm r}}\}, \qquad (3)$$

$$\mathrm{RL} = 20\log|(Z_{\rm in} - Z_0)/(Z_{\rm in} + Z_0)|\,, \qquad (4)$$

where Z_0 is the impedance of free space, t is the thickness of an absorber, c is the speed of light. As shown in Fig.6, if t is 1.25 mm, the microwires annealed at 600°C show better absorption performances, for microwires annealed at 600°C, the minimum RL is −15.8 dB at 15 GHz, while for microwires annealed at 750°C, the minimum RL is –9.4 dB at 14.33 GHz.

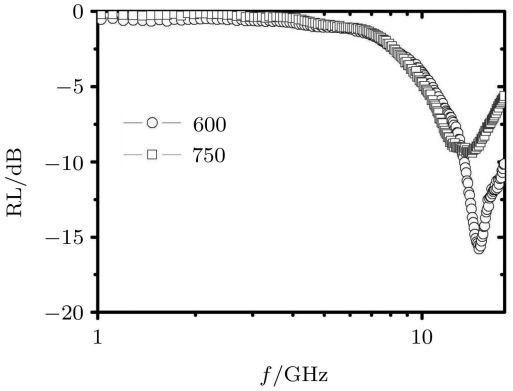


Fig.6. The microwave absorption properties of microwires.

4. Conclusions

In summary, amorphous FeSiB microwires have been fabricated by a melt–extraction method. The microwave measurements show that the permittivity and permeability are dependent on the annealing, which has altered the microstructures of microwires. For microwires annealed at 750°C, within the lower frequency range of 1–3 GHz, the permittivity values are found larger than those annealed at 600°C. Fitting results show that multi Lorentzian dispersion behaviours play major roles in determining permittivity dispersion. As for permeability, two dispersion peaks are found in the $\mu' - f$ for the microwires annealed either at 600°C or at 750°C. The peaks at lower frequency are believed due to the DWM mechanism, the other one at higher frequency is believed due to the NR mechanism. Besides, the microwires annealed at 600°C show better electromagnetic wave absorption ability in terms of reflection loss.

References

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

[2] Kane S N, Alves F, Gercsi Z, Mazaleyrat F, Gupte S, Chiriac H and Vazquez M 2006 *Sensor. Actuat.* A *Phys.* **129** 216

[3] Dudek C, Adenot-Engelvin A and Acher O 2006 *IEEE Trans. Magn.* **42** 2787

[4] Liang D F, Han M G, Yan B and Deng L J 2007 *Chin. Phys.* **16** 542

[5] McHenry M E, Willard M A and Laughlin D E 1999 *Prog. Mater. Sci.* **44** 291

[6] Lin D M, Wang H S, Wu Y C and Lin M L 1999 *Acta Phys. Sin.* (Oversea Editions) **08** 455

[7] Lagarkov A N, Matytsin S M, Rozanov K N and Sarychev A K 1998 *J. Appl. Phys.* **84** 3806

[8] Li Z W, Lin G, Chen L, Wu Y and Ong C K 2006 *J. Appl. Phys.* **99** 063905

[9] Jiles D C 2003 *Introduction to Magnetism and Magnetic Materials* (in Chinese, translated by Xiao C T), (Lanzhou, China: Lanzhou University Press) p285

[10] Liao S B 1988 *Ferromagnetism* (in Chinese) (Beijing: Science Publishing House) **3** p58

[11] Naito Y and Suetake K 1971 *IEEE Trans. Microw. Theory Techn.* **19** 65