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ARTICLE

The Tiny Stress Impedance Effect of $Fe_{73.5}Cu_lNb_{1.5}Si_{13.5}B_9Mo_{1.5}$ Powder/Silicon Rubber Composite Film and $Fe_{73.5}Cu_lNb_3Si_{13.5}B_9$ Amorphous Ribbon with Lamellar Structure

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Abstract: The composite piezomagnetic film was prepared by using $Fe_{73.5}Cu_lNb_{1.5}Si_{13.5}B_9Mo_{1.5}$ amorphous powder with particles size of 40 μm as dispersing agent and silicon rubber as the matrix. Meanwhile, the $Fe_{73.5}Cu_lNb_3Si_{13.5}B_9$ amorphous ribbon was selected as piezoelectric layer. The testing frequency was ranged from 10 kHz to 1 MHz, and the pressure stress was from 0 to 0.14 MPa. The tiny stress impedance (TST) effects of the two types of layer structures were compared in the experiment, the two types included piezomagnetic/piezoelectric/piezomagnetic (pm/pe/pm) layer and piezoelectric/piezomagnetic/piezoelectric (pe/pm/pe) layer. It is found that both of the layer structures have magnetoelectricity coupling effects. The magnetoelectricity coupling effect enhances with the increase of pressure stress before 0.02 MPa, and afterward the magnetoelectricity coupling effect comes to a maximum value, which has little contribution to TSI. Since two types of layer structure are both of varying magnetoelectricity coupling effect, pm/pe/pm is piezomagnetic based and pe/pm/pe is piezoelectric based.

Key words: stress-impedance; layer structure; piezomagnetic; piezoelectric; magnetoelectricity coupling effect

Since Joule found magnetostriction at 1982, it has become a profound and significant issue to magnetoelasticity area. While whichever theory has a mutual one in correspondence. The so called SI is the mutual theory of the magnetostriction. In 1992, Professor Murillo et al of Nagoya Univ Japan have firstly reported that Fe base FeCoSiB amorphous alloy in high frequency alternating magnetic field can present a remarkable SI effect. After that, it has become a hot issue to the experts and scholars from electromagnetism area at home and abroad.

In our work, two types of layer structures of these two films have been made. We discussed pm/pe/pm and pe/pm/pe layer structures about the influence of TSI and the repeatability and stability of SI effect at tiny stress. In addition, we used magnetoelectricity coupling mechanism and its constitutive relation to explain the effect of TSI.

1 Experiment

$Fe_{73.5}Cu_lNb_{1.5}Si_{13.5}B_9Mo_{1.5}$ amorphous powder (mole ratio) with the grain size of 40 μm was used in the experiment and 107# silicon rubber was used as the basic resin which was mixed with silane ethyl with the mass ratio of 9:1[1]. 20% in mass ratio of acetone was used as the dilute material. After that, the resin basic material and amorphous powder were mixed with mass ratio at 1:3. The mixture was stirred in a vacuum agitated reactor (below 0.1 Pa) and then poured into molds, pressed into the size of 30 mm×10 mm×0.3 mm composite film which was consolidated at NPT for 24 h. The piezoelectric layer of $Fe_{73.5}Cu_lNb_3Si_{13.5}B_9$ ribbon was made by magnetic control spitting system and the size of which was also 30 mm×10 mm×0.3 mm. The interface of two layers was bonded in a vacuum environment and a stress of 20 MPa was perpendicularly exerted to junction surface until piezomagnetic was fully polymerized.

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To ensure the stability and accuracy of stress loading, a device and a mold of testing TSI were designed, shown in Fig.1.

The testing mold can be divided into upper piece and downpiece as shown in Fig.1a. Pressure head area is 9.8 mm^2. Firstly the layer film was put in the direction of paralleling to down-mold axle wire, and then a copper coil was inlaid into the two sides of the film. Finally the upper piece of the mold was covered and the squeeze head was put into die hole.

The stress continuous loading system Fig.1b is controlled by a PC which can adjust stress steps. Loading system can be divided into 7 parts, including stress sensor, buffering layer, testing mold, pressure head, drift sensor, lever arm and motor-driven nut. It will inflict continuously the stress from 0~1 MPa with the pressure head. PC is combined with stress sensor and motor-driven nut. The feedback data from stress sensor is input to PC and the control instruct is output to motor-driven nut so as to control the stress. In our experiment, the stress ranges was from 0 to 0.14 MPa, and step size was 0.01 MPa.

The laminating pattern of the film could be divided into two types, one was pm/pe/pm and another was pe/pm/pe, as shown in Fig.2. The silicon rubber & $Fe_{73.5}Cu_1Nb_{1.5}Si_{13.5}B_9Mo_{1.5}$ amorphous powder was the piezomagnetic layer and $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ amorphous ribbon was the piezoelectric layer. Both of them had two identical magnetoelectricity coupling interfaces, and each interface has a magnetoelectricity coupling effect. The influences of TSI induced by magnetoelectricity coupling effect will be discussed in details below.

This lamellar magneto-electricity composite structure consists of a piezomagnetic layer and a piezoelectric layer. The constitutive equation[2] of piezomagnetic layer can be written as:

$$\begin{aligned} S_3 &= sT_1 + qH_2 \\ B_1 &= qT_2 + \mu H_\mathrm{n} \end{aligned} \tag{1}$$

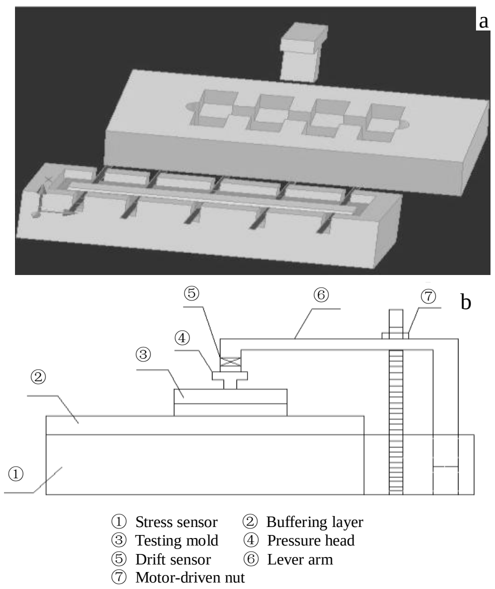

Fig.1 Testing mold and device of layer film

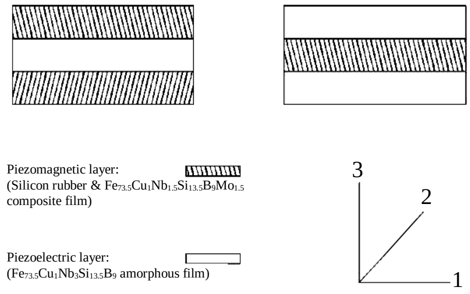

Fig.2 Two types of laminating patterns

And constitutive equation of piezoelectric layer can be written as:

$$\begin{aligned} S_3 &= sT_1 + \mathrm{d}E_2 \\ D_1 &= \mathrm{d}T_2 + \zeta E_n \end{aligned}$$

The constitutive equation of composite structure merged by piezomagnetic layer and piezoelectric layer can be written as:

$$\begin{aligned} S_3 &= sT_1 + \mathrm{d}E_2 + qH_2 \\ D_1 &= \mathrm{d}T_2 + \zeta E_n + \alpha H_n \\ B_1 &= qT_2 + \mu H_n + \alpha E_n \end{aligned} \tag{2}$$

where, d, q, α are piezoelectric modulus, piezomagnetic coefficient, magnetoelectricity coefficient, respectively; s, ζ, μ are elasticity soft degree coefficient, permittivity, permeability, respectively. While T, D, B, S, E, H represent stress, electric displacement, magnetic flow, strain, electric intensity and magnetic field intensity, respectively. The index n represents homologous coupled field. Magnetoelectricity coupling coefficient can be calculated by equation[4]:

$$\alpha = \mu_0 \left(\frac{\partial M}{\partial E} \right)_H \tag{3}$$

here, μ_0 is vacuum permeability, M is magnetic torque, E is electric field excited by piezoelectric layer, and H is magnetic field excited by piezomagnetic layer.

2 Results and Discussion

Two microstructure pictures of layer structures were done by the high depth of field optical microscope. Fig.3 shows 1000 times-enlarged photographs of two types of the layer. It can be observed from Fig.3 that the surface of piezomagnetic layer is full of micro pores and scratch. The amorphous powder is dispersed in rubber equably. These micro pores were formed with the polymerization process of the silicon rubber. With polymerization progress, the volume of the silicon rubber will reduce in a certain dimensions, which has been reflected by these micro pores. Meanwhile, the surface of piezoelectric layer is smooth as glass, which has been prepared by magnetron sputtering method.

After that, the TSI of the two types of the layer structures with the stress ranges of 0~0.14 MPa was tested by the Agilent 4284A AC impedance analyzer. The results are shown in Fig.4.

The values of the impedances with the two type structures were tested at the frequency from 10 kHz to 1 MHz. Variation

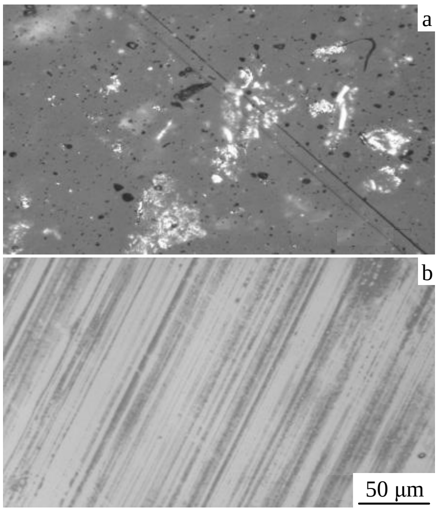

Fig.3 Microstructure pictures of two types of layer: (a) piezomagnetic layer and (b) piezoelectric layer

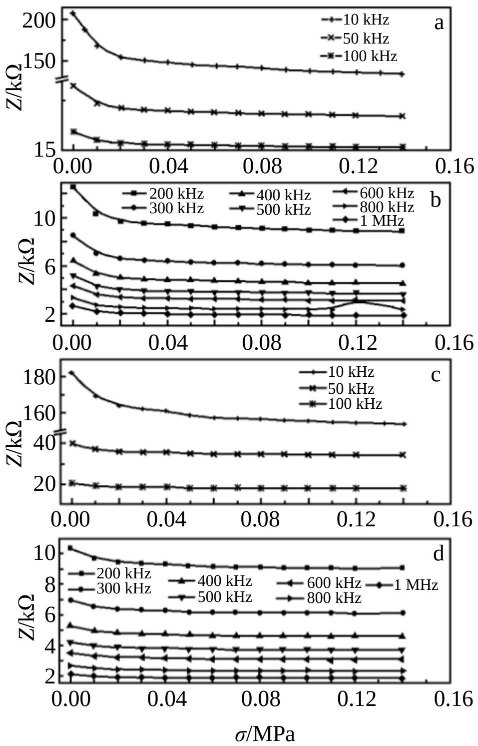

Fig.4 Changes of impedance with stress at different frequencies in two structures: (a, b) piezomagnetic/piezoelectric/piezomagnetic and (b, d) piezoelectric/piezomagnetic/piezoelectric

trends of the impedances with the stresses are approximately similar at each testing frequency. Fig.4 displays whatever pm/pe/pm or pe/pm/pe film impedance exhibits an exponential form decline before 0.02 MPa stress, and afterward which shows a slow linear decline. Since the magnetoelectricity coupling effect enhances before 0.02 MPa stress and afterward the magnetoelectricity coupling effect reaches a maximum, the impedance changes not so obviously after 0.02 MPa stress. This statement magnetoelectricity coupling effect can be eliminated and only have TSI effect. In addition, the impedance also declines with enhancing of testing frequency which can regard these films as capacitive reactance force sensor. About the stress sensibility, pm/pe/pm is more sensitive than pe/pm/pe film and the value of the impedance is larger at same testing frequency and stress.

In order to emerge a visual phenomenon of pm/pe/pm and pe/pm/pe[5-9] films SI tendency, the curves have been made at frequency of 10 kHz on two types of layer, as shown in Fig.5. Where, SI%=$|Z_\sigma - Z_0|/Z_0$ [3].

Fig.5 presents the stress-impedance of the thin composite films constituted by different laminating ways. According SI% function, the curves of SI% and Z are a mirror symmetry[10-13] and the value of SI% can reflect the degree and trend of Z. The curves are more smooth and the linear fitting degree[14] is better for pm/pe/pm thin films, which is opposite for pe/pm/pe thin films. The variation of pm/pe/pm thin films, coupled with SI% data, is far larger than that of pe/pm/pe thin films, which can reflect that its sensitiveness is superior to that of pe/pm/pe thin films.

Magnetoelastic effect mechanism is an important characteristic of force sensor, which contains electricity magnetoelastic effect and magnetic magnetoelastic effect. In order to test the magnetoelastic effects of these two kinds of the thin films, the experiment of stress upload and offload were done four times. The degree of magnetoelastic effect is represented by the average area of the curves upon loading and unloading. And the larger mean value means the more serious magnetoelastic effect[15], as shown in Fig.6.

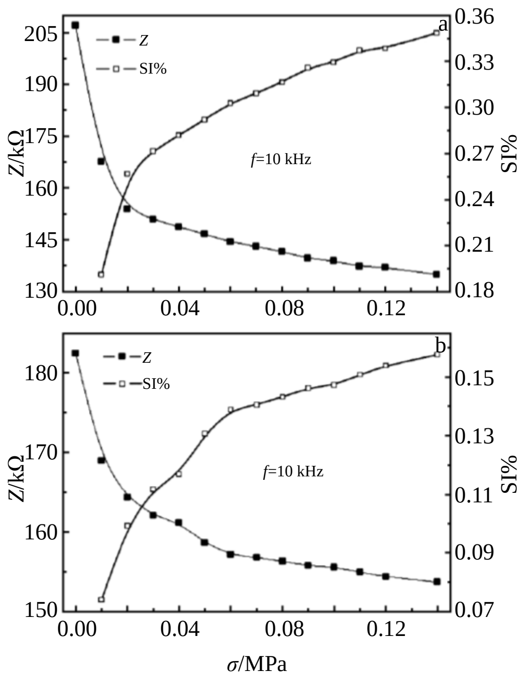

Fig.5 Curves of Z and SI% with stress at 10 kHz frequency in two structures: (a) piezomagnetic/piezoelectric/piezomagnetic and (b) piezoelectric/piezomagnetic/piezoelectric

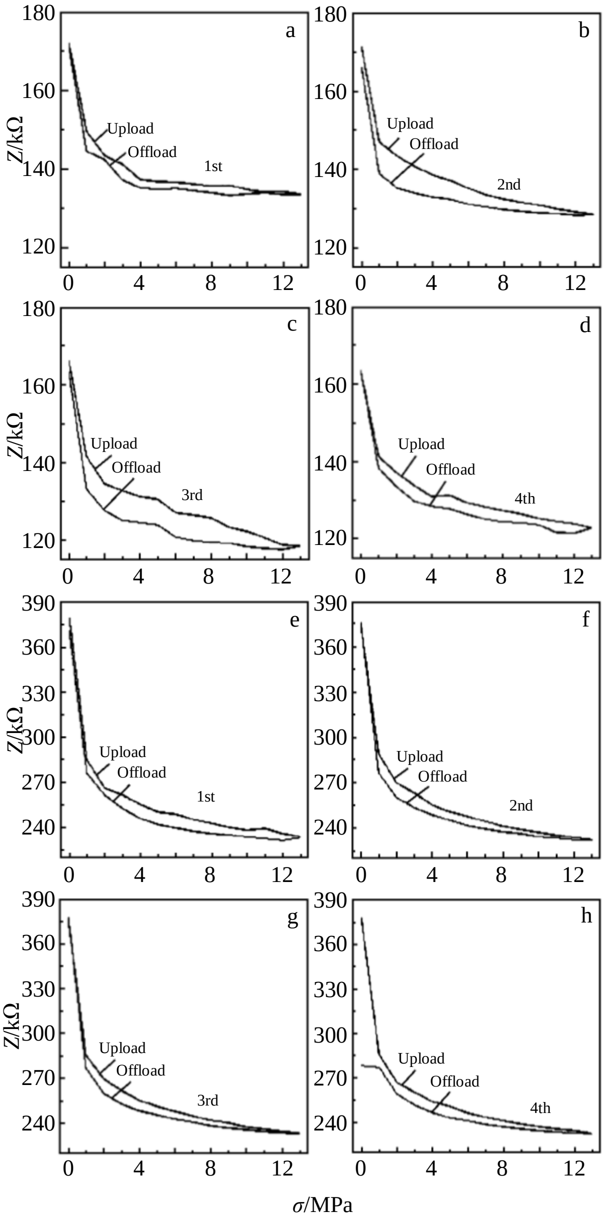

Fig.6 Four times repeated upload and offload stress curves in two structures: (a, b, c, d) piezomagnetic/piezoelectric/piezomagnetic and (e, f, g, h) piezoelectric/piezomagnetic/piezoelectric

From Fig.6 we can calculate the randomness of magnetoelastic after effect is intense. It will not present congruent relationship with times of upload and offload, but the TSI effect is more stable in pe/pm/pe layer. This because, both of the two structures have magnetoelectricity coupling layers, but pe/pm/pe layer is piezoelectrics dominants and pm/pe/pm layer is piezomagnetism dominants. So magnetoelectricity is more stable than piezomagnetism.

3 Conclusions

1) The two structures (pe/pm/pe and pm/pe/pm) consist of silicon rubber/$Fe_{73.5}Cu_lNb_{1.5}Si_{13.5}B_9Mo_{1.5}$ powder composite film and $Fe_{73.5}Cu_lNb_3Si_{13.5}B_9$ amorphous ribbon with two layer magnetoelectricity coupling interfaces.

2) The magnetoelectricity coupling effect increases with the increase of stress before 0.02 MPa, which makes the TSI effect obviously. But when the stress reaches to 0.02 MPa, the magnetoelectricity coupling effect gets to a maximal value.

3) The TSI has a functional relationship with stress and has no relationship with magnetoelectricity coupling effect.

4) Concerning the stability and the repeatability of the stress, pe/pm/pe is much better than pm/pe/pm due to its better stability of piezoelectric effect.

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$Fe_{73.5}Cu_{l}Nb_{1.5}Si_{13.5}B_{9}Mo_{1.5}$ 粉体/硅橡胶复合薄膜与 $Fe_{73.5}Cu_{l}Nb_{3}Si_{13.5}B_{9}$ 非晶薄膜层状结构的微应力阻抗效应研究

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摘　要：选用最大粒度为 40 μm 的 $Fe_{73.5}Cu_{l}Nb_{1.5}Si_{13.5}B_{9}Mo_{1.5}$ 粉体和硅橡胶混合制备的磁性薄膜作为压磁层。同时用 $Fe_{73.5}Cu_{l}Nb_{3}Si_{13.5}B_{9}$ 非晶薄膜作为压电层。测试频率范围从 10 kHZ 到 1 MHz，并且压应力从 0~0.14 MPa。比较了两组层合结构的微应力阻抗效应，其中包括：压磁/压电/压磁层合结构和压电/压磁/压电层合结构。结果证明两组层合结构都存在不同程度的磁电耦合效应。在压应力小于 0.02 MPa 时，磁电耦合效应随压应力增大而增大，大于 0.02 MPa 时，磁电耦合效应达到极大值，同时它对微应力阻抗效应贡献不明显。因为两组层合结构具有各自不同的磁电耦合体系，压磁/压电/压磁层合结构以压磁效应影响阻抗为主而压电/压磁/压电层合结构以压电效应影响阻抗为主。

关键词：应力阻抗；层状结构；压磁；压电；磁电耦合

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