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Progress on improvement and application of magnetostrictive properties of the Fe-Ga alloy

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Abstract. Fe-Ga alloy is a novel material with high magnetostriction, good mechanical properties, and wide application prospects. At present, few reviews systematically summarize the improvement and application of magnetostrictive properties of Fe-Ga alloys. Combined with the latest research progress, this paper reviews Fe-Ga alloys and their magnetostrictive properties and expounds on the methods to improve the magnetostrictive properties of Fe-Ga alloys from three aspects: component design, preparation methods, and heat treatment. For example, doping Dy, Ce, Pr, and C elements, improving the heat treatment process such as introducing an H2S atmosphere and strong magnetic field, and improving the preparation methods such as directional solidification and rapid laser solidification technology. The magnetostrictive properties of Fe-Ga alloys have a wide application in the field sensors, transducers, flexible electronic devices, vibrators, actuators, and so on. This paper provides new ideas for improving the magnetostrictive properties of Fe-Ga alloy and promoting its wide application.

Keywords: magnetostriction; Fe-Ga alloy; research progress; doping

1. Introduction

The magnetostrictive effect is the phenomenon that the size of ferromagnetic materials or ferromagnetic materials changes due to the variation of the magnetization state under the effect of an external magnetic field. The magnetostrictive effect in ferromagnets is excited by the spin-orbit coupling of atoms or ions of the material itself. From the perspective of the minimum energy principle, when the magnetization state of magnetic materials changes, to maintain the minimum energy, its shape or volume would inevitably change. The magnetostriction effect exists in all natural substances, but only the materials with an absolute magnetostriction coefficient λs larger than 50ppm are regarded as magnetostrictive materials by researchers.

Magnetostrictive materials possess many excellent characteristics, such as large magnetic elastic coupling coefficient, large output stress, fast mechanical response, strong stability, and so on. The following mechanical magnetic energy conversion effects are the basis for its application: magnetostriction effect, inverse magnetostriction effect (Village effect), △E or △G effect, Wiedemann effect and Jump effect. It can be used in the field of sensors, sonar systems, actuators, flexible electronic devices, and so on [1].

Although traditional magnetostrictive materials, such as Fe and Ni, provide mechanically robust intelligent materials, they have poor magnetostrictive properties and strong temperature dependence. Giant magnetostrictive materials, such as Terfenol-D(Tb-Dy-Fe), have a high magnetostrictive coefficient, large output power, and easy processing procedure, but the required magnetic field is high, the eddy current loss at high frequency is large, and the price is expensive and brittle [2]. Piezoelectric ceramics, such as PZT and PMN, provide large strain values but limit mechanical robustness and exhibit temperature-sensitive characteristics. To fill the gap between traditional magnetostrictive alloys and rare earth giant magnetostrictive alloys, it is urgent to develop new magnetostrictive materials with large magnetostriction, low price, high mechanical strength, and narrow hysteresis.

Until the beginning of this century, Clark [3] found that the magnetostrictive coefficient of iron is only ~20ppm, and the saturated magnetostrictive coefficient of its single crystal along the (100) crystal direction could be as high as 400ppm if added with Ga. This magnetostrictive material that

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combines the properties of high magnetostriction, low saturated magnetic field, good mechanical properties, and low manufacturing cost is rare for more mature intelligent materials, filling the gap between traditional magnetostrictive materials and giant magnetostrictive materials. Fe-Ga alloy can be applied in aviation, navigation, robotics, energy, biomedicine, and many other fields, and have a great scientific research value [4].

In this paper, the improvement and applications of magnetostrictive properties of Fe-Ga alloy are reviewed. Firstly, the properties and preparation methods of Fe-Ga alloy are explored. Secondly, the factors that influence the magnetostriction effect are described. Further, the methods to improve the magnetostriction properties of Fe-Ga alloy are explored from the aspect of component design and process technology. Finally, the existing applications are summarized, and future research directions are prospected.

2. Properties and Preparation of Fe GA Alloy

2.1 Properties

Fe-Ga alloy possesses the characteristics of strong mechanical properties (500MPa), small brittleness, good ductility, and corrosion resistance [4]. Owing to their excellent mechanical property and the ability to establish anisotropy through stress annealing, Fe-Ga alloy can maintain its magnetic properties under tensile conditions and can withstand large loads, shocks, vibrations, and long-term harsh conditions [5]. In addition, Fe-Ga alloy has a high Curie temperature (TC ~ =700 ° C), which is weakly dependent on temperature. It can operate in a wide temperature range without significant changes in properties [6].

Fe GA alloy has excellent magnetostrictive properties. Its saturation magnetization field is low, and it can exhibit a good magnetostriction effect (~ 350ppm) with an ultralow magnetic field of about 100oe (8000Am − 1) [4]. In addition, Fe-Ga material has high relative permeability, low coercivity, high coupling coefficient low hysteresis, and excellent soft magnetic properties.

2.2 Preparations

2.2.1Preparation of Fe GA single crystals (and oriented polycrystals)

The preparation of Fe-Ga alloy single crystal material mainly adopts the directional solidification technology. This technology removes heat from molten metal in one direction, causing the liquid-solid interface to move from one end of the mold to the other. The growth direction of the grain is perpendicular to the liquid-solid interface. Through the elimination of the grain, the material with a certain preferred orientation is obtained. The commonly used directional solidification methods include the Bridgman method, flat zone melting, and Czochralski method. The advantages and disadvantages of these methods are shown in Table 1. Directional solidification has the problems of high cost and complex processing, so it is mainly applied in laboratories or fields with extreme demand for performance.

Table 1. Comparison of directional solidification method [7].

Category	Merit	Defect
Bridgman	Few parameters, materials preferred orientation usable controlled	Long melting time, much material loss; Hard maintain steep temperature gradient
Zone melting	Short melting time, less material loss	Much parameters, materials' preferred orientation hard controlled; Limited bar size
Czochralski	Maintain steep temperature gradient,stable single crystal growth condition, high properties and few defects of preparative crystal	Complicate technology process, long crystal growth period, much material loss, poor technology stability, hard be practical

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2.2.2Preparation of Fe-Ga polycrystalline block

Although the Fe-Ga alloy single crystal oriented along (100) possesses the best magnetostrictive properties, its size is limited and its mechanical properties are not as good as those of polycrystalline alloys. The polycrystalline block of Fe-Ga alloy is mainly prepared by vacuum melting (Fig. 1). Based on the difference in heating sources, vacuum smelting can be divided into vacuum induction furnace smelting, vacuum arc furnace smelting, and electron beam smelting. Among these techniques, vacuum arc melting is the most widely used one, which utilizes arc discharge to heat materials in vacuum condition. During smelting, the melted material can be connected to the negative pole and the water-cooled copper crystallizer to the positive pole. After electrified, arc discharge is generated between the two poles, which converts electric energy into heat energy and generates a high temperature to melt the material. In the smelting process, the liquid metal droplets fall into the metal pool after passing through the high-temperature arc area and solidify into ingots in the water-cooled copper crystallizer. The melted polycrystalline alloy is usually placed in a vacuum and sealed quartz tube, and repeated overturning smelting and long-term high-temperature annealing are carried out to eliminate the heterogeneity of microstructure and composition.

In addition, the rapid laser solidification method, powder metallurgy method, electric explosion method, and rolling method can also be used to prepare Fe-Ga magnetostrictive polycrystalline materials.

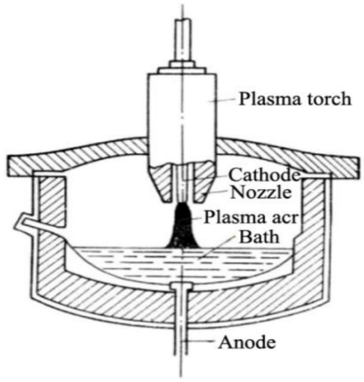

Fig 1. Schematic diagram of vacuum melting [7].

2.2.23Preparation of Fe-Ga alloy strip

The preparation methods of Fe-Ga alloy thin strip mainly include a rolling method and rapid quenching method. The rolling method mainly uses the friction between the rotating roll and Fe-Ga alloy to drawing the alloy into the two rolls. The alloy will produce a certain degree of plastic deformation and form a certain texture when it is extruded through the roll. This method is cheap and practical. The prepared samples have good magnetostrictive properties and are suitable for large-scale production.

The rapid quenching method (Fig. 2) is a non-equilibrium solidification (quenching) method. To use the strip sample uniformly, the polycrystalline with the same composition after arc melting is generally used as the raw material. The emotional gas is introduced from the upper end of the quartz tube by melting the master alloy through induction heating. The pressure produced by the gas overcomes the surface tension and sprays the alloy liquid from the lower nozzle of the quartz tube onto the high-speed rotating roller below. The alloy liquid cools and solidifies rapidly at the moment of contact with the roller. Under the action of the centrifugal force generated by the rotation of the roller, it is thrown forward rapidly in the form of a thin strip [7]. Compared with directional solidification, rapid quenching has the advantage of short processing time, but there are too many variables in the processing process, and the performance of the finished product is not easy to control.

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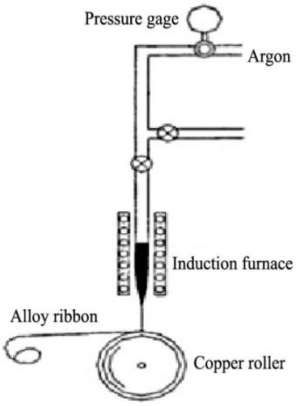

Fig 2. Schematic diagram of (b)single roller quenched [7].

3. Methods of Improving Magnetostrictive Properties of Fe-Ga Alloy

3.1 Magnetostrictive Properties

As shown in Figure 3, the magnetostriction of Fe-Ga alloy increases with the increase of Ga content, and two magnetostriction peaks appear near the Ga content of 19% and 27% respectively [8]. With the addition of Ga atoms, the alloy maintains a disordered body-centered cubic A2 phase (bbc structure) at low Ga composition. When the content of Ga is more than 20%, the ordered phases of body-centered cubic B2 and D03 (bcc structure), face-centered cubic L12 and closely packed hexagonal D019 appear.

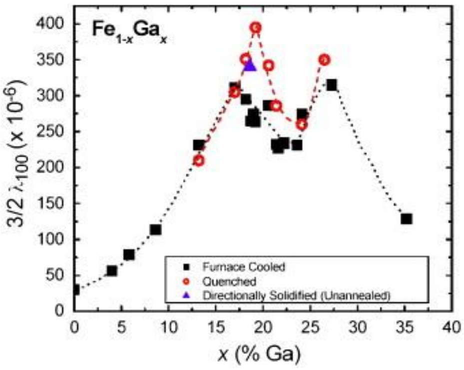

Fig 3. Saturation magnetostriction along(100)vs. atomic % Ga in Fe–Ga alloys[8]

Researchers have conducted intensive studies on the mechanism of the large magnetostrictive coefficient of Fe-Ga alloy. But so far, a unified understanding has not been formed. In current research works, the large magnetostrictive effect of Fe-Ga alloy is mainly understood from three aspects. In terms of electronic structure, calculations of the first principle show that Ga atoms can enhance the spin-orbit coupling of the A2 phase, so solid solution Ga will improve the magnetostrictive properties [8,9]. In terms of microstructure, from the perspective of crystal texture, the (100) direction has the largest magnetostrictive coefficient, and the (110) and (111) crystal systems deviated from the (100) direction have lower magnetostrictive coefficients [7]. From the perspective of phase structure, Fe-Ga alloy is a heterogeneous material, and high-temperature A2 contributes the most to the relative magnetostrictive properties. In addition, the tetragonal nanophase (modified-D03) is dispersed on the A2 phase matrix, forming Ga-Ga short-range order along the (001) direction, enhancing local magnetocrystalline anisotropy and inducing tetragonal distortion of the A2 matrix phase, thereby improving magnetostrictive properties [10]. In terms of macroscopic properties, the large magnetostrictive effect is closely related to modulus softening [11]. Fe-Ga alloy with strong magnetostrictive properties will have the minimum value of shear elastic coefficient $c'=[(c11-c12)/2]$. The reduction of c' will improve the strain output capacity of the material.

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3.2 Component Design

In recent years, researchers have also tried to add many trace alloying elements as the third element to Fe-Ga alloy to improve the magnetostrictive properties of the alloy. It can be roughly divided into the following three categories: transition elements, main group elements, and rare earth elements.

3.2.1Rare earth element

Rare earth elements include Tb, Dy, Sm, Ce, Pr, etc. The 4f electron orbitals of Tb and Dy are anisotropic ellipsoidal. When an outer magnetic field is applied, the spin magnetic moment and orbital magnetic moment rotate, affecting the grain orientation of polycrystalline alloys. Using the low elastic coefficient of Fe-Ga alloy, the matrix can be softened and even tetragonal distortion can be induced by trace solid solution rare earth elements and a modified-d03 phase can be induced, which can further improve the magnetostrictive properties of Fe-Ga alloy [12].

The (Fe0.83Ga0.17)100 - xTbx(x=0~0.5) alloy was prepared by directional solidification method, with the achievement of high magnetostrictive properties of ~255ppm, ~6.6% large strain, and excellent mechanical properties [13]. Fitcharov et al. [12] added Tb to Fe81Ga19 alloy, which increased the Curie temperature by 150K (a Curie temperature rise of 150K,) and improved the stability of magnetic moment below room temperature. (Fe0.83Ga0.17)100–xDyx(0<x<0.42) strips were prepared by melt spinning. When x=0.25, the maximum vertical magnetostriction coefficient of the strip rectangle upward was ~620ppm, and the Curie temperature first increased and then decreased with the increase of x [14]. However, when Fe87Ga13Dyx is prepared by induction melting and the concentration is high (x=2.0), dλ///dH, magneto-mechanical coupling, and △E effect would all decrease [15]. In addition, Wu et al. [16] prepared (Fe0.83Ga0.17)100-xSmx (0<x<0.42) strips by melt spinning, and the vertical magnetostriction value reached 500ppm at x=0.25. He et al. [10] pointed out that the best trace dopants are light rare earth Ce and Pr, with the transverse magnetostriction up to ~800ppm, and even the longitudinal magnetostriction as high as 2000ppm is expected to be achieved.

3.2.2 3d, 4d transition elements

3d and 4d transition elements include Ni, V, Co, Cr, Co, Zn, etc. V, Cr, Mn, and Mo atoms have a stabilizing effect on the D03 phase, which is not conducive to improving the magnetostrictive properties of Fe-Ga alloy. Summers et al. [17] found that the addition of Ni and V to change the electronic structure of the alloy did not significantly change the magnetostrictive properties of the alloy. Co atom substitution can improve magnetostrictive properties, Curie temperature, magneto-mechanical coupling performance, elastic modulus E0, and zero field shear modulus G0, but a large CO substitution could weaken the role of Ga atoms [18]. Gao et al. [19] found that adding Cr to polycrystalline Fe-Ga alloy can significantly improve the mechanical properties at room temperature, while the magnetostrictive coefficient is only 70ppm. If Fe73Ga18Zn9 is prepared by adding 9% Zn into fe73ga27 alloy, after quenching and aging at 700 ℃ for 24 hours, it can improve the magnetostrictive property and maintain stability, and improve the ferromagnetic shape memory effect, which provides the possibility for the application of the alloy in high-temperature environment [20].

3.2.3Main family elements

The main group elements include Si, Ge, be, Sn, Al, C, B, etc. The substitution of Si or Ge for Ga atoms hinders the formation of Ga-Ga atom pairs, resulting in the decline of the magnetostrictive properties of the alloy. A small amount of Sn or Al atom substitution may produce lattice distortion and affect spin-orbit coupling, and slightly improve the magnetostrictive properties of the alloy. Al substitution can reduce the cost of the alloy. The magnetostriction of annealed sheet added with 1.0% B is ~160ppm[21]. The single crystal sample of slow cooling Fe81.3Ga18.6C0.08 has magnetostriction of ~400ppm, but it shows high enough permeability to form eddy currents proportional to the thickness of the sample, which limits its work at high frequencies [21]. In

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addition, when trying to roll Fe81Ga19 alloy added with C, cracks formed along the grain boundary, and the sample broke in the subsequent hot rolling process, resulting in the alloy being too brittle to be made into thin plates.

3.3 Processing Method

As verified in previous research, the solidification transformation of Fe-Ga alloy is very sensitive to the cooling rate. At a high cooling rate, the alloy is rapidly cooled from a high temperature melting state to room temperature in a very short time, which can retain the disordered A2 phase and inhibit the formation of the ordered phase. At the same time, saturated Ga atoms cannot precipitate due to rapid cooling, forming a large number of Modified-D03 phases containing Ga-Ga atom pairs, thereby improving magnetostrictive properties.

Magnetostrictive properties are very sensitive to the thermal history. The conventional heat treatment process can effectively control the internal microstructure of Fe-Ga alloy, and reduce material defects and internal stress. The heat treatment procedure is to put the material into the furnace first, raise the temperature to a certain temperature under the protection of Ar gas, then keep it for some time, and then cool it by furnace cooling, air cooling, or water quenching. In addition, the heating rate, heat treatment temperature, holding time, cooling mode, as well as the introduction of an H2S atmosphere and strong magnetic field can effectively improve the internal structure and magnetostrictive performance [22]. Srisukhumbowornchai et al. [23] studied the as cast polycrystalline Fe72.5Ga27.5 alloy and found that the magnetostriction of the A2 phase obtained by quenching the sample at 875 ℃ was higher than that obtained by holding at 730 ℃ for 220h. Clark et al. [24] showed that furnace-cooled fega single crystals λ100 reaches the peak when the Ga content is 17%, while the quenched alloy λ100 reached the peak when the Ga content was 19%.

Using magnetic field annealing to treat Fe-Ga alloy, the magnetic field plays an important role in the precipitation and development of ferromagnetic nanoparticles, and significantly promotes the rapid phase transition from ordered bcc to ordered fcc, which enhances the magnetostrictive properties of Fe GA alloy. DS (001) Fe73Ga27 alloy was annealed in the magnetic field at 1200Oe and 720 ℃ in a short time of 2min. Under a low magnetic field (800Oe), the saturation magnetostriction of Fe73Ga27 polycrystalline alloy is significantly increased to 370ppm [25].

Fe-Ga alloy was prepared by rapid quenching method, and the disordered A2 phase under high temperature was retained to room temperature to improve the concentration of Ga atom pairs. During the solidification of the thin strip, due to the large temperature gradient and stress gradient on the free surface and the roller surface, columnar crystals with (100) orientation are formed, and large magnetostriction is obtained. However, its later processing is difficult, with high stress and brittleness.

The Fe-Ga alloy sheet prepared by the rolling method can not only obtain the alloy with texture but also meet the requirements of high-frequency use and reduce the eddy current loss during high-frequency use. The rolling process is complex, suitable for large-scale production, and low price.

Ga rich phase precipitates in vacuum melting, and the microstructure of Fe-Ga alloy obtained is an equiaxed crystal with coarser grains, and the magnetostriction value is small. Compared with the vacuum melting method, the directional solidification method can obtain (001), (110) and (100) preferred orientation crystal structures. If the cooling rate and temperature gradient are properly controlled, the magnetostrictive properties can be effectively improved. For columnar crystals grown in one direction, Clark et al. [26] obtained (100) oriented single crystals with a growth rate of 2mm/h by using the modified brilliant method, and their magnetostrictive properties can reach 300ppm. These alloys with large magnetostrictive properties are obtained from the fact that the Fe-Ga alloy has a bcc structure (100) direction is its easy magnetization direction.

It is worth noting that a high solidification rate is a necessary condition to obtain good magnetostrictive properties, and the most prominent characteristics of laser rapid solidification technology are rapid solidification and non-equilibrium structure formation. The extremely short

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action time and extremely small action area (micron level) of the laser can achieve an extremely fast cooling rate, which provides a strong guarantee for retaining more A2 phase and Modified-D03 phase and inhibiting the formation of ordered phase during solidification. Laser forming can produce an extremely high-temperature gradient and very fast solid-liquid interface advancing speed, which is conducive to obtaining polycrystalline alloys with a certain orientation texture. In addition, based on the discrete stacking principle, laser rapid prototyping can expand the above unique solidification structure to three-dimensional entities, which provides a technical way for the preparation of Fe-Ga alloys with complex structures. Additive manufacturing technology represented by laser rapid solidification is a hot spot in the future research of Fe-Ga alloy preparation technology, and it is also the latest development direction of advanced manufacturing technology.

4. Application of Magnetostrictive Properties of Fe-Ga Alloy

4.1 Sensor

The magnetostrictive inverse effect and Weidman inverse effect of magnetostrictive materials can be applied for sensor, which requires fast response, good stability, good mechanical properties, and high strain. Fe-Ga alloy exactly has the above advantages, which makes it more suitable for some execution and sensing applications than piezoelectric or terfenol-D, and is preferred in harsh and vulnerable environments.

At present, the relatively advanced sensors include magnetic sensors, acoustic sensors, micro displacement sensors, force sensors, and so on. Figure 4 is a schematic diagram of a non-contact torque sensor based on a magnetostrictive circuit. The torque on the shaft generates tension in one belt and compression in the other belt, causing changes in the magnetization in the two belts and changes in the magnetoresistance in the belt. Moreover, it also results in differences in their longitudinal fields. Both effects can be utilized to detect torque when the shaft rotates relative to the fixed magnetic sensor. The displacement sensor accurately measures the change of position through the pulse signal. Because the pulse signal propagates at the speed of sound, the displacement distance can be accurately determined by measuring the propagation time of the pulse signal.

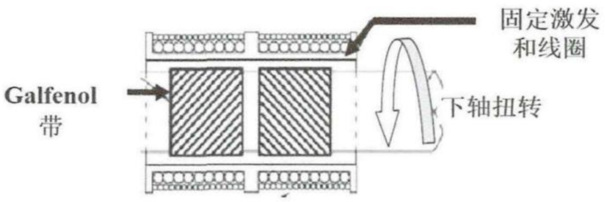

Fig 4. schematic diagram of a non-contact torque sensor based on a magnetostrictive circuit[27]

Combined with thin film preparation technology and epitaxy, magnetoelastic materials, and microelectromechanical systems (MEMS) technology, the size of magnetostrictive sensors is greatly reduced, and the cost-effectiveness and sensitivity are improved. On the micro-scale, Fe-Ga alloy has good toughness and is easy to be epitaxially deposited on a silicon substrate, which makes it very suitable for the application of microsensors [28]. On the nanometer scale, Fe-Ga alloy nanowires are fabricated on silicon substrates. Each nanowire is sensitive to the difference of small frequency bands. By controlling the aspect ratio of nanowires, the aspect ratio of sound waves can be increased to epitaxial decompose the spectrum of sound signals. It can distinguish large spectrum of acoustic signals in space and is expected to be used in micro acoustic, tactile, and magnetic sensors in weapons and equipment, ocean exploration, and other fields.

4.2 Underwater Acoustic Transducer

The electromagnetic wave in the liquid decays quickly while the acoustic signal decays less. The lower the frequency of the emitted sound wave, the smaller the attenuation of the acoustic signal in the water. A Sonar system is the main way for ocean communication and detection. Its core

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component is an underwater acoustic transducer, which is used for signal transmission in water. The transducer uses the magnetostrictive effect to convert electromagnetic energy and acoustic energy. The performance of magnetostrictive material directly affects the performance of the transducer. Fe-Ga material can be miniaturized, with a fast low-frequency response, wide frequency band, small low-field drive, and can resist underwater explosion and shock waves. It is an ideal material for the preparation of underwater acoustic transducers.

4.3 Flexible Electronic Devices

Flexible electronic devices are devices that integrate electronic devices on non-planar, ultra-thin, flexible substrates. Among them, magnetostrictive films and devices are an important branch, and the requirements for their components are flexibility, including flexible power supply, flexible circuit, flexible display, flexible sensing, flexible storage, etc. Because magnetostrictive films grow on flexible substrates, it is easier to control magnetic anisotropy. Giant magnetostrictive material films (GMF) made of Fe-Ga materials have better properties than Terfenol-D films [29].

4.4 Miniature Vibrator

Fe-Ga alloy is easy to be processed into various shapes, which can also be easily deformed by a very low driving magnetic field. Therefore, a small permanent magnet can be used to produce a miniature vibrator, which also has the advantages of small driving coil voltage, low resistance, and loss.

4.5 Actuator

As the core component of the active control system, the magnetostrictive actuator made of giant magnetostrictive material is also playing a more and more important role in MEMS. The traditional actuator is made of piezoelectric ceramic material (PZT), but it has the problems of drift, aging, and overvoltage breakdown, so it cannot be used in the environment of low frequency and high stroke. Fe-Ga alloy materials have the advantages of large deformation, high Curie temperature, and high energy conversion efficiency, and are increasingly used in the design and manufacture of microactuators.

It is worth noting that the magnetostrictive actuator requires a compressive prestress to ensure maximum magnetostriction. This is because, under the action of ideal compressive stress, all magnetic moments are perpendicular to the rod axis in the demagnetized state, and the length of the sample is the shortest. When the magnetic field makes all magnetic moments parallel to the sample axis, the maximum total change of sample length can be obtained. Wu Fogle et al. [30] studied the stress annealing of magnetostrictive Fe-Ga alloy to produce embedded stress. In addition to having high yield strength, the built-in stress enables the produced Fe-Ga alloy to produce maximum magnetostriction under tensile stress. In addition, Yoo et al. [31] developed a magnetic field annealing program, which can also be used to generate an appropriate amount of embedded stress in Fe-Ga alloy.

5. Conclusion

As a new magnetostrictive smart material after Terfenol-D, Fe-Ga alloy has great development and application potential. It has excellent magnetostrictive properties, low saturated magnetic field, good temperature characteristics, good mechanical properties, and low manufacturing cost. There are many preparation methods such as directional solidification, vacuum melting, rolling, rapid quenching, and selective laser melting. The main influencing factors of magnetostrictive properties are electronic structure, microstructure, and shear elastic coefficient. At present, the magnetostrictive properties of Fe-Ga alloy can be improved from the aspects of component design, preparation method, and heat treatment. The magnetostrictive properties of Fe-Ga alloy are widely used in the fields of sensors, actuators, transducers, and flexible electronic devices. In the future, for

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Fe-Ga alloy magnetostrictive materials, researchers need to increase research on reducing eddy current losses at high frequencies and exploring new preparation methods and application fields. This paper guides people to better understand this material and the improvement of its magnetostrictive properties and inspires people to explore its potential applications.

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