Analysis and Research of Magnetorheological Elastomers Piezoresistive Conductivity

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On the basis of particle structure analysis, this paper studies the conductive mechanism of magnetorheological elastomers (MRE), and verifies the MRE conduction mechanism model based on the combination of tunnel current and conduction current. The piezoelectric characteristic of MRE is analyzed theoretically and the basic theoretical model of piezoelectric is established. A set of MRE piezoelectric characteristic test device has been designed independently. The test device is used to test the resistance values of MRE samples prepared in this experiment. Finally, the test results of each group are analyzed and compared, and the results show that this element can realize the stability test of MRE piezoelectric conductivity. Under different orders of magnitude, the low particle volume content is more sensitive to the conductivity of MRE samples, and the current flowing through the MRE sample shows a significantly non-linear relationship with the voltage applied to the sample.

Keywords : MRE, conduction mechanism, piezoresistive characteristics, test experiment, nonlinear

1. Introduction

MRE, like MRF, is a new type of material that has a continuous, rapid and reversible change in rheological properties under a magnetic field [1]. MRE also has controlled mechanical, electrical, magnetic and thermal properties, showing significant piezoelectric and temperature resistance characteristics [2]. These properties make MRE an important field in the study of polymer functional materials. In recent research, Wang Guangshuo and Ma Yingying et al. prepared the MR fluid of composite materials. This MR fluid was prepared from the Fe₃O₄/ MoS₂ nanocomposites and its corresponding MR performances were examined using a rotational rheometer [3]. Some research prepared calcium ferrite ($CaFe_2O_4$) nanocrystal dusters by a solvothermal method and used them to make MRF [4]. There are also studies using MnFe₂O₄/GO to prepare MRF and the corresponding MR properties were investigated using a Physica MCR301 rheometer fitted with a magneto-rheological module [5].

MRE is an intelligent material with development potential [6]. The application of MRE in its conductive properties is still very little, and its conductive mechanism

©The Korean Magnetics Society. All rights reserved. *Corresponding author: Tel: 0532-860-57111 Fax: 0532-860-57111, e-mail: skdjdxy17@163.com has similarities with the common conducting polymeric materials [7]. Many researchers have carried out theoretical analysis and experimental tests on the specific conditions of conductive polymer materials. However, there is no systematic theory on the conductive mechanism, so the research on the electrical conductivity of MRE should be further explored. The research on the conductivity of MRE can enrich the application field of conductive composite polymer materials, and also provide technical support for the research and optimization control of new sensors. It is the basis of the MRE sensing characteristics research.

With the discovery of later studies, when there is no magnetic field, no conductive chain is formed inside the MRE, but the polymer composite still has a certain conductivity and also very sensitive to external excitation response [8]. Therefore, the research on the influence factors of MRE electrical conductivity can be used to provide certain technical support for the optimization control and research of flexible sensor parts in life [9].

2. Experimental Conditions

2.1. Experimental Basis

MRE is mainly composed of carbonyl iron powder, silicone rubber and curing agent [10]. The type, size and content of ferromagnetic particles have a great influence

Table 1. Iron powder content in different MRE (Volume ratio v/v).

The sample number	Curing additive	Iron powder	Silicone rubber
MRE-1	10 %	1 %	89 %
MRE-2	10 %	0.1 %	89.9 %

on the performance of MRE [11]. Granular materials require high permeability and high saturation magnetization to enhance the magnetic force of particles [12]. Meanwhile, granular materials must also have very low remanence to ensure the stability of MRE performance [13]. The properties of the matrix material require good stability, aging resistance and small permanent deformation [14]. It is generally required that the viscosity of the substrate before curing is not more than 10 Pa·s, and the modulus of elasticity after curing is smaller. The dosage of additive plays an important role in reducing the viscosity of the matrix before curing and the modulus of elasticity after curing and the modulus of elasticity after curing the preparation of the MRE.

According to the above requirements, the ferromagnetic materials used in this experiment are produced by Jiangsu Tianyi superfine metal powder Co.. Ltd. Its model is MRS-MRF-35 carbonyl iron powder and the average particle size is 3.14 mm. In this experiment, MRE matrix material selection of Dow Corning 184 silicone rubber, including polymer raw materials and solidified additives, the use of mass ratio is 10 : 1, its basic performance meets the above requirements. The MRE of single particle size was prepared according to Table 1.

2.2. Preparation tools of MRE

(1) Electronic balance

The HZT-A+200 type electronic balance produced by Fuzhou Hua Zhi Scientific Instruments Company Ltd. is selected in this experiment, which is mainly used for weighing raw material and beaker to prepare MRE with different particle volume ratio. The main parameters of the HZT-A+200 type electronic balance are as follows: the stabilization time of weighing is no more than 3 seconds, the weighing range is 0.01 g~200 g, and the minimum displayed value is 0.01 g.

(2) Ultrasonic cleaning machine

The JP-010T type ultrasonic cleaning machine produced by Shenzhen Jie Meng Cleaning Equipment Company Ltd. is selected in this experiment, which is mainly used to clean the experimental devices such as mould, beaker, and a stirring rod, to ensure the effectiveness of the experimental data. The main parameters of the JP-010T type ultrasonic cleaning machine are as follows: the maximum capacity is 2.0 L, the ultrasonic frequency is 40 KHz, and the ultrasonic power is 80W.

(3) Vacuum drying oven

The DZF-6022 type vacuum drying oven produced by Shanghai Heng Science Company Ltd. is selected in this experiment. It is mainly used to remove the mixed air when mixing the magnetorheological mixture and to solidify the silicone rubber. The main parameters of the DZF-6022 type vacuum drying oven are as follows: the vacuum degree is 133pa and the temperature range is $+10\sim200$ °C of room temperature.

(4) Other tools

Beaker, used for holding and weighing of silicone rubber; mortar, container for mixed silicone rubber and carbonyl iron powder. The magnetic source of permanent magnet is used as the solidified magnetic field of MRE, and the HT20 Tesla meter is used to measure the magnetic induction intensity of the strong static magnetic field directly because the MRE solidification is basically a one-dimensional uniform magnetic field.

2.3. Mould preparation of conductive element

The material used in this experiment is pure copper, as shown in Fig. 1 below. It should be cleaned up in the ultrasonic cleaning machine and dried before use.

The mould structure designed for the preparation of MRE, as shown in Fig. 2(a), adopts the preparation method of immobilization of mould and MRE sample. The main components are divided into three parts: retaining ring, fastened bolt and nut, upper and lower end covers with circular convex platforms. The main material is non-magnetic material (pure copper), which can prevent mag-



Fig. 1. (Color online) Conducting element material.

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1. Retaining ring 2. End cover 3. MRE samples enclosed in baffle and end cover. **Fig. 2.** (Color online) Mold structure of conductive element.

netic agglomeration. The upper and lower end covers with circular convex platforms are connected by the retaining ring, and the distance between the upper and lower convex platforms is 1 mm (that is the thickness of the MRE sample), the diameter of the MRE sample is 16 mm. Bolt and nut are fastened on the end covers. When the solidification of silicone rubber is finished, the rings will be removed, and the end covers are always kept in contact with the sample, which can ensure the combination of the mold and the sample in the maximum extent, improve the reliability and accuracy of the experiment, and prepare for the later experimental measurement. After the removal of the ring, the solidified MRE sample and the mold constitute a conductive element together, as shown in Fig. 2(b).

2.4. Preparation of MRE

(1) Clean

For the ensurence of the accuracy and effectiveness of the experimental data, beakers and other experimental supplies should be cleaned in the ultrasonic cleaner and be dried prior to use.

(2) Configuration of materials

In this experiment, the content of carbonyl iron powder = Carbonyl iron powder / (Carbonyl iron powder + silicone rubber + curing additive). As for the experiment on the different volume ratio of carbonyl iron powder, the amount of carbonyl iron powder, silicone rubber and curing additive for two different volume ratios should be calculated in advance according to the requirements of the experiment.

(3) Weighing

The volume of carbonyl iron powder, silicone rubber and curing agent is calculated according to the different volume ratio obtained by step (2), and the amount of experimental material is weighed by electronic balance.

(4) Mixed and remove bubbles



Fig. 3. (Color online) Mixing and grinding, add curing agent and vacuum.

The silicone rubber and carbonyl iron powder after weighing are mixed in a mortar and stirred mechanically for 10 minutes, Then add the curing agent and continue stirring for 10 minutes, vacuuming for 30 minutes to remove bubbles. The purpose of removing bubbles again is to remove the air mixed with the MRF mixture during stirring the greatest extent, and avoid the air gap in the MRE sample after curing which may cause much error in the experiment. as shown in Fig. 3.

(5) Structuration of particles

After the bubbles were removed, the mixture was injected into the MRE mold. Then the mold is fixed in the strong static magnetic field device which is consisted by two high temperatures resistant permanent magnets. The magnetic induction intensity is 115 mT, as shown in Fig. 4.



Fig. 4. (Color online) Static high-intensity magnetic field device.

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(6) Curing of matrix material

The magnetic field is curing device and the conductive element mold are placed in the vacuum drying box, as shown in Fig. 5. It is gradually heated to 120 °C then wait for 60 minutes after the temperature reaches 120 °C. The mold is took out for cooling after the completion of the silicone rubber curing. When the cooling is finished, remove the ring and carry out the experiment while the end cover always keeps the original state of contact with the sample.



Fig. 5. (Color online) Base material curing device.





(a) Volume ratio 1%(b) Volume ratio 0.1%Fig. 6. (Color online) MREs samples.



Fig. 7. Microstructures of the MREs samples.



Fig. 8. The chain-like structure picture of the MREs samples.

The MRE samples with single particle size of different proportions prepared by this mold are shown in Fig. 6. The microstructure of longitudinal section of the MRE observed under the vacuum scanning tunneling microscope is shown in Fig. 7. The chain-like structure of the MREs samples is shown in Fig. 8. It can be seen that ferromagnetic particles in the longitudinal section of the MRE prepared by this method are basically columnar arranged along the direction of the magnetic field, and the internal structure is compact, which accords with later experimental research and experimental requirements.

3. Theoretical Analysis of Piezoresistive Conduction

The conduction principle of MRE is the migration of internal electrons in the polymer system to form a current. The conductivity of MRE is anisotropic. When the volume of ferromagnetic particles is relatively small, MRE is conductive in the direction of particle chain, while MRE is insulated in the direction perpendicular to the particle chain [15]. The conductive mechanism of MRE is attributed to two basic theories: one is the theory of conduction channel and the two is the theory of the tunnel effect [16]. When the current flows through MRE, the conduction current and the tunnel current work together in the particle contact area inside the matrix material. Tunneling effect theory plays a leading role in low filler concentration and low voltage of MRE, while conduction mechanism of conductive channel theory is significant mainly at high concentration of conductive filler.

According to the microstructure of MRE samples, it is known that when the content of conductive fillers is low, the ordered structure of MRE particles is reduced to particle chain, and the single chain structure is used to analyze the conductive mechanism between MRE particles. Two adjacent particles are selected in the direction of the



Fig. 9. A longitudinal section of MRE.

particle chain to analyze the conductivity of the body element of MRE. The schematic diagram of the longitudinal section of the body element is shown in Fig. 9.

The total height (H) of the representational element:

$$H = 2R + h \approx 2R \tag{1}$$

Where, R is the radius of the ferromagnetic particles, h is the distance between particles and particles;

MRE cross-sectional area of the body element:

$$S_r \approx \frac{2\pi R^2}{3\varphi} \tag{2}$$

Where S_r is the cross-sectional area of the body element; φ is the particle volume ratio. MRE cross section area of the body element can be regarded as the average cross section area of the MRE particle chain. MRE is composed of a single representational element. So, the average electric field intensity and the average current density of MRE are the same as the average electric field intensity and the average current density of the body element [17]. According to the relationship between electric field intensity,current density and conductivity.

$$j = \sigma E \tag{3}$$

Where, *j* is the current density; σ is the conductivity; *E* is the electric field intensity. It can be obtained that the conductivity of MRE is equal to that of the body element.

In the case of only the matrix material conductance, the conduction current density formula is as follows.

$$j_c = \sigma_f E \tag{4}$$

Where, j_c is the conduction current density

The tunnel current density is generally expressed by Fowler-Nordheim [18, 19]:

$$j_t = AE^2 \exp\left(-\frac{B}{E}\right) \tag{5}$$

Where, j_t is the tunnel current density:

A and B are the characteristic constants of composites c material, the expression is as follows.

$$A = \frac{q^3 m_e}{16\pi^2 m_{em} \varphi_0} \tag{6}$$

$$B = \frac{4(2m_{em})1/2\varphi_0^{3/2}}{3qh}$$
(7)

Where, q is the electron charge, m_e is the mass of the electron in a vacuum, m_{em} is the mass of electrons in the matrix material, h is the Planck's constant, φ_0 is the heat barrier height.

In the diversion area between particles, tunnel current and conduction current work together. So the current density of the conduction area is the sum of the two. Its formula is as follows.

$$j_i = j_t + j_c = \left[\sigma_f + AE \exp\left(-\frac{B}{E}\right)\right]E$$
(8)

Where, j_i is the Current density, E is the applied electric field, A is the number of times an electron tries to cross the "forbidden" zone per second, -B/E is the probability of a specified electron crossing the energy band.

The formula for the average current density of the entire MRE is as follows.

$$j_m = 3\phi \left[\frac{2A}{h_0^2} E_0 \exp\left(-\frac{h_0 B}{2RE_0}\right) + \frac{\sigma_f}{Rh} \right] r^2 E_0$$
(9)

Where, j_m is the average current density.

According to formula (3), the theoretical resistivity of MRE can be obtained, and its formula is as follows.

$$\sigma_m = 3\phi \left[K_1 E_0 \exp\left(-\frac{K_2}{E_0}\right) + K_3 \right] r^2$$
(10)

It is shown that the resistivity of MRE and electric field intensity are nonlinear. When the cross-sectional area of MRE is S_0 , the voltage u is applied, and the thickness is H_0 . The current formula of MRE is as follows.

$$I_{0} = 3S_{0}\phi \left[K_{1}\frac{u}{H_{0}}\exp\left(-\frac{K_{2}H_{0}}{u}\right) + K_{3}\right]\frac{r^{2}u}{H_{0}}$$
(11)

The above formula shows that the current flowing through the MRE consists of two parts: the tunnel current and the conduction current. Its formula is as follows.

$$I_{0t} = 3S_0 \phi K_1 \frac{r^2 u^2}{H_0^2} \exp\left(-\frac{K_2 H_0}{u}\right)$$
(12)

$$I_{0c} = 3S_0 \phi K_3 \frac{r^2 u}{H_0}$$
(13)

The expressions of K_1 , K_2 and K_3 defined in the above formula are as follows.

$$K_1 = \frac{2A}{h^2} \tag{14}$$



Fig. 10. Microscopic model of two adjacent soft magnetic particles for MREs.

$$K_2 = \frac{hB}{2R} \tag{15}$$

$$K_3 = \frac{\sigma_f}{Rh} \tag{16}$$

Based on the Hertz contact theory, the particles will deform under pressure. The microscopic structure model of adjacent soft magnetic particles interacting with each other is shown in Fig. 10.

$$F = \pi R^2 \sigma_{\rm t} \tag{17}$$

Where, *r* is the effective radius of intergranular contact points, *R* is the particle radius. The particles in MRE are surrounded by matrix materials. *F* is the pressure between adjacent particles. σ_t is the applied stress of MRE.

When two particles are squeezed under pressure, the center is turning from an arc to a small round surface. The radius expression is as follows.

$$r = \sqrt[3]{\frac{3FR(1-v^2)}{2E}}$$
(18)

Where, v is the Poisson's ratio of soft magnetic particles. *E* is the Modulus of elasticity. Substituting (17) into (18), the formula of contact radius and applied stress is obtained. Its formula is as follows.

$$r = \sqrt[3]{\frac{3\pi\sigma_{\rm t}(1-\nu^2)}{2E}R}$$
(19)

Suppose the radius of the initial contact surface is approximately r_0 . According to the equation (19), Get the initial stress σ_0 and its relationship. Its formula is as follows.

$$r_0 = \sqrt[3]{\frac{3\pi\sigma_0(1-\nu^2)}{2E}}R$$
 (20)

Considering the influence of initial contact surface, the deformation formula of particle under external force is as follows.

$$a = \left(\sqrt[3]{\sigma_0 + \sigma_t} - \sqrt[3]{\sigma_0}\right)\sqrt[3]{\frac{3\pi(1 - \nu^2)}{2E}}R$$
(21)

The formula for increasing the radius of the guide surface is as follows.

$$r_Z = r_{z0} + a \tag{22}$$

Substituting (21) and (22) into (10), The resistivity of the MRE under pressure is obtained. Its formula is as follows

$$\sigma_m = 3\phi \left[K_1 E_0 \exp\left(-\frac{K_2}{E_0}\right) + K_3 \right] \left(K_4 + K_5 \sqrt[3]{\sigma_0 + \sigma_1} \right)^2 \quad (23)$$

The current - voltage formula of MRE under pressure is as follows.

$$I_{0} = 3S_{0}\phi \left[\frac{K_{1}}{H_{0}^{2}}u^{2}\exp\left(-\frac{K_{2}H_{0}}{u}\right) + \frac{K_{3}}{H_{0}}u\right] \left(K_{4} + K_{5}\sqrt[3]{\sigma_{0} + \sigma}\right)^{2}$$
(24)

The expression of K_4 and K_5 defined in the above formula is as follows.

$$K_{4} = r_{z} - \sqrt[3]{\frac{3\pi\sigma_{0}(1-\nu^{2})}{2E}}R$$
(25)

$$K_{5} = \sqrt[3]{\frac{3\pi(1-\nu^{2})}{2E}}R$$
(26)

4. Experimental Study of Piezoresistive Conductivity

4.1. Experimental test

Based on the characteristics of MRE, research and analysis on the conduction mechanism based on the conduction current and tunnel current, A set of testing device based on MRE piezoelectric conductivity is established,then test the piezoelectric characteristics of MRE. First it needs to put pressure on the material, for ease of measurement,apply certain pressure to the vertical direction of the sample. The experimental test system of MRE piezoelectric characteristics is shown in Fig. 11.

The power supply model used in the experiment is GPC-3060D. It can achieve three sets of regulated power supply output (Two sets of adjustable, one set of fixed voltage). Adjustable voltage can be adjusted manually from 0 to 32 V, the display accuracy is 0.25 V. The voltage meter is a multimeter for the Agilent digital. The



(a) MRE sample conductance test physical picture



(b) Piezoresistive characteristics of the experimental test device diagram

Fig. 11. (Color online) Piezoresistive conductance test system of the MREs.

measurement accuracy is 0.001 mV.

The upper and lower copper ends of the conductive element mould are used as electrodes, Weld the copper wire separately, A closed loop is connected with the DC stabilized current supply and resistance through the wire, Series resistors play a role in the partial pressure, in case of measurement errors due to excessive internal resistance of the power source. The current direction is parallel to the pressure direction, therefore, the experiment was designed and manufactured by the screw and designed the piezoelectric characteristic experimental test device of MRE. As shown in Fig. 12.

Specific testing process: Place MRE sample on the pressure machine's workbench; The working table of the press is controlled up and down by stepping motor, which can adjust the pressure applied to the sample; The DC voltage is applied at both ends of the MRE sample while maintaining the pressure, the current that flows through the MRE is calculated.

Adjust the output voltage U of DC stabilized voltage, the resistance in the series circuit is R_0 . The voltage U_1 at both ends of R_0 is measured by the desktop multimeter.



Fig. 12. (Color online) Experimental device for the piezoresistive characteristic of MRE.

Calculate the current flowing through the circuit according to the test results. Its formula is as follows.

$$I = \frac{U_1}{R_0} \tag{27}$$

The conductance formula of MRE sample in the conducting element is as follows.

$$G = \frac{I}{U - U_1} \tag{28}$$

The conductivity formula of the MRE sample in the conducting element is as follows.

$$K = \frac{GL}{A} \tag{29}$$

Where, G is the conductance of MRE sample, conductance can be measured by current and voltage, the resistivity value can be obtained according to the relation. A is an effective contact surface for MRE samples, L is the height of the sample(the distance between the upper and lower cover plate).

4.2. Analysis of experimental results

Based on the established MRE piezoelectric characteristic test system and test method, the piezoelectric characteristic of MRE is tested. In order to further study the pressure-sensitive conductivity characteristics of MRE samples, two groups of MRE samples with different particle volume ratios were experimentally measured. One is the MRE sample with a volume ratio of 1 %, and the other is 0.1 %.

(1) The two groups of MRE samples with different volume ratios were tested under different voltage for the corresponding current value and resistivity. Experimental tests were carried out under no pressure, and the different voltages corresponded to the current value and the

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(a)Volume ratio 1% MRE-1 resistivity calculated (b)Volume 1% MRE-1 current experimental value



(c) Volume ratio 0.1% MRE-2 resistivity calculated (d) Volume 0.1% MRE-2 current experimental value **Fig. 13.** Current and electrical resistivity depend on voltage variation without pressure.



(a) Volume ratio 1% MRE-1,30 minute resistivity values (b) Volume ratio 1% MRE-1,60 minute resistivity values



(c) Volume ratio 1% MRE-1,90 minute resistivity values (d) Volume ratio 1% MRE-1,120 minute resistivity values **Fig. 14.** Volume ratio 1 % MRE-1, The same time interval resistivity varies with voltage.



Fig. 15. Volume ratio 1 % MRE-1, The same time interval current varies with the voltage.



Fig. 16. Volume ratio 0.1 % MRE-2, The same time interval resistivity varies with voltage.

resistivity, as shown in Fig. 13.

As can be seen from Fig. 13, the initial MRE resistivity reaches a maximum at a small initial voltage. The resistivity of the MRE sample is very sensitive with the changes in applied electric field and the resistivity decreases with increasing voltage. When the initial voltage increases from 0 to 24 V, the resistivity declines most rapidly. When the voltage is greater than 24 V, the resistivity tends to decrease with the increase of applied voltage. The current increases with the increase of voltage, the greater the voltage is, the more obvious the current increases, and the current and voltage show obvious nonlinear relationship.

(2) In this experiment, the same pressure of 108 MPa was applied to the two kinds of MRE samples with a volume ratio of 1% and a volume ratio of 0.1% to unload them suddenly. The experimental data was tested every 30 minutes and compared with that under no pressure to obtain Fig. 14, Fig. 15, Fig. 16, Fig. 17 shows the trend of the current and the resistivity of the MRE with voltage under the applied pressure.

As can be seen from Fig. 14, Fig. 15, Fig. 16, Fig. 17,

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as the load acting on the MRE changes, the conductance of the MRE also changes with time changes and the applied electric field. After pressure was applied to the sample and unloaded suddenly, the trend of current and resistivity was consistent with that of the non-applied pressure at the same time. Due to the viscoelasticity of matrix, the compressive stress decayed with time, the resistivity of MRE sample increased continuously and the current flowing through the sample decreased continuously. The resistivity of MRE sample with particle volume ratio of 0.1 % was obviously higher than that of MRE sample with 1 % The current is significantly less than 1 % of MRE samples. The MRE sample with a particle volume ratio of 1 % was suddenly unloaded after pressure application, After 120 minutes, the change of resistivity and current basically coincided with the reverse trend without pressure, and the original state was restored. It can be seen from the analysis that at different orders of magnitude, the lower particle volume content has a smaller effect on the viscosity of the mixture than the higher particle volume content, and is more sensitive to the conductance of the MRE sample.



Fig. 17. Volume ratio 0.1 % MRE-2, The same time interval current varies with the voltage.

5. Conclusion

In this paper, the piezoresistive conductance of MRE has been theoretically analyzed and experimentally studied. First of all, the conductive mechanism of MRE was analyzed to determine that it is a combination effect of conductive channel theory and tunneling theory.

Through the theoretical analysis of MRE's piezoresistive conductive characteristics, an MRE conduction model combining conduction current and tunneling current is proposed, and the formula of MRE resistivity and electric field strength is deduced. Based on the formation of grain structure in MRE and the determination of conductive paths, the relationship between the resistivity and the applied load of MRE was analyzed based on the increase of the area of conductive passage caused by the deformation of particles under the applied load.

A MRE conductivity testing device was designed and prepared to improve the bonding between the magnetorheological elastomers sample and the testing device. Under the action of static uniform magnetic field, the original recovery state of ferromagnetic particles is improved, magnetic agglomeration is prevented and the stability of MRE electrical performance test is improved. These are the preparation of high performance magnetorheological elastomers and provides an accurate reference for the accurate conductivity test. By testing the MRE's piezoelectric conductance and analyzing the experimental results, it is verified that the influence of the electric field and the compressive stress on the conductivity of the MRE is not coupled. The current flowing through the MRE sample and the voltage applied to the sample are obviously nonlinear relationship.

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