

A theoretical model for the field-dependent conductivity of magneto-rheological gels and experimental verification



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ARTICLE INFO

Article history:

Received 14 December 2015

Received in revised form 11 April 2016

Accepted 3 May 2016

Available online 4 May 2016

Keywords:

Magneto-rheological gel (MRG)

Magneto-resistance mechanism

Theoretical model

Carbonyl iron powder (CIP)

Verification

ABSTRACT

A magneto-rheological gel (MRG) is a kind of smart material fabricated by dispersing ferromagnetic particles in high viscosity polymer gel. The internal particles of MRG are uniformly dispersed under off-state. While with the precondition of an external applied magnetic field, the particles will form a reversible chain-like structure, in which process the electrical resistivity of MRG has an immense change. In order to deeply understand the changing mechanism of the magneto-resistance of MRG, a theoretical model based on both the magnetic-dipole behavior and the percolation theory was proposed in this work. This model incorporates the parameters of the magnetic field intensity and carbonyl iron powder (CIP) volume fraction. As a further effort to explore the magneto-resistance properties of MRG, a series of experimental testing were conducted. The magneto-resistance properties of MRG with different CIP volume fractions and under different magnetic fields were investigated. The experimental results indicated that the conductivity of MRG was significantly increased with the increasing magnetic field and CIP volume fraction. In addition, in order to verify the effectiveness of the proposed theoretical model, the experimental measured results and theoretically obtained results were compared. The validation results demonstrated that the theoretical model proposed in this work is capable of reproducing the field-dependent magneto-resistance properties of MRG.

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1. Introduction

Conductive polymer composites (CPCs) which are mainly prepared by dispersing the conductive particles in an insulating matrix have many remarkable advantages as compared to alternative materials, for instance, light weight, wide range of electrical conductivities, ease of shaping, low density as well as chemical resistance [1,2]. As a consequence of their conductive properties, CPCs are valuable materials for implementation in an industrial field. For instance, they are widely used as adhesives and circuit elements in electronic applications [3,4], as them protection devices applied to electromagnetic interference (EMI) shielding which can protect against radiation and electrostatic dissipation (ESD) to prevent harmful arcing discharges [5]; Also, they have potential applied value as sensitive pressure [6], temperature [7] and chemical [8] sensors. The common used fillers of CPCs are carbon black, graphite, ceramic, carbon fiber, metal, etc. Under

the external stimuli such as pressure, temperature and magnetic field, the distribution and contact area between each filler will be changed. Therefore it lead to huge changes in conductance which corresponding to the piezoelectric-resistance [9], thermo-resistance [10] and magneto-resistance [11]. Generally, the CPCs are sensitive to external stimuli only when the fillers' concentration reaches to a percolation threshold. While, as the percolation threshold of CPCs is usually relatively high [12,13], to get such a high volume fraction of filler particles and reproducibly in CPCs becomes a tough challenge [14].

Magneto-rheological (MR) materials [15], which are normally prepared by dispersing micron-sized magnetic particles into soft matter matrix (fluid, elastomer, gel, foam, etc.), can overcome the aforementioned problems. Due to magnetic interactions, the conducting magnetic particles within MR material can self-assemble align into columnar or chainlike structures with a uniform magnetic field [16]. The local volume fraction of particles around the chain is very high, thus the percolation threshold value of MR materials is very low [1,6]. That is to say, MR materials can form into a conductor at low particle concentration and exhibit unidirectional conduction. Consequently, a lot of research have

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been carried out to study the electrical conductivity of MR materials. Martin et al. [13] reported that by filling nickel particles with a volume fraction of 7.5%, magneto-rheological elastomers (MRE) exhibited giant thermos-resistance, piezo-resistance, and chemical-resistance. Zhu et al. [6] and Mietta et al. [17] analyzed the influences of magnetic field and compression on the conductance of MRE samples which were prepared at low particle fraction and low magnetic field, they concluded that the resistance of the MRE decreased with the increases of the both ones. Kchit et al. [18] developed a new MRE whose resistance was strongly sensitive to temperature and pressure, and they drew a conclusion that by replacing the matrix with a softer elastomer, a magneto-resistance with a decrease by three orders of magnitude was obtained under a field of only 100 kA m^{-1} (1250 Oe). By using magnetorheological fluid (MRF), Kim et al. [19] fabricated a magnetic sensor and through which they could measure the external magnetic field. Mietta et al. [20] investigated the impacts of various external loading on the resistance of anisotropic MRE, and a flexible, anisotropic and portable stress sensor (logarithmic reversible response between 40 and 350 kPa) was fabricated by them.

Considered as a new branch of MR materials, MR gel (MRG) consisting of a high viscosity polymer gel impregnated with ferromagnetic particles. It improved the stability of the system as it overcame the challenge of particle sedimentation which existed in MRF [21]. In our previous work, the magneto-resistance characteristics of MRG under magnetic field were studied [11], the results showed that the resistance of MRG was sensitive to the changes of magnetic field, and it performed a decreasing trend as the magnetic flux density varied from 0.1 T to 1 T. To date, a lot of researches on the conductive mechanism of MRE have been studied [4,6,22]. While, MRE is mainly made by dispersing micrometer-sized magnetically permeable particles in a solid or rubber-like non-magnetic matrix material [23]. Due to the particles have been fixed in the matrix, the electrical resistivity of MREs were insensitive to the changes of magnetic field replaced by sensitive to pressure [18]. Therefore, the theoretical model of MRE based primarily on compressive stress which the MREs were assumed anisotropic and the mathematical models were supposed to be an ideal chain structure [22]. On the contrary, the MRG possess an off-state viscosity which other than MRE and the ferromagnetic particles in MRG can move freely under the magnetic field [24]. When a magnetic field is applied, the particles driven by the magnetic field force rearrange to form chains or columnar structures along the direction of magnetic field [25], and the MRG transition from an insulator into a conductor in this process [11]. It can be seen that the conductive mechanism of MRG is different from MRE and this difference between MRG and MRE are the major motivations of this research.

As a consequence, in an effort to explore the conductivity of MRG, the main contributions of this work include: (1) a new theoretical model of MRG was developed to precisely predict the magnetic field-dependent conductivity of MRG, and the effect of magnetic field and particle concentration on the model output were discussed. (2) The effectiveness of the proposed model was validated through a comparative work between measured results and theoretically obtained results, among which several samples of MRG were fabricated and their magneto-resistance properties were explored at different magnetic fields. And it is shown that the proposed model can reproduce well experimental results without causing significant difference.

2. Theoretical model

A series of conductive polymer composites (CPCs) which the resistivity between insulating materials ($10^{15} \Omega \text{ m}$) and conductive material ($10^{-7} \Omega \text{ m}$) have appeared since the 1950s [26]. Up to now,

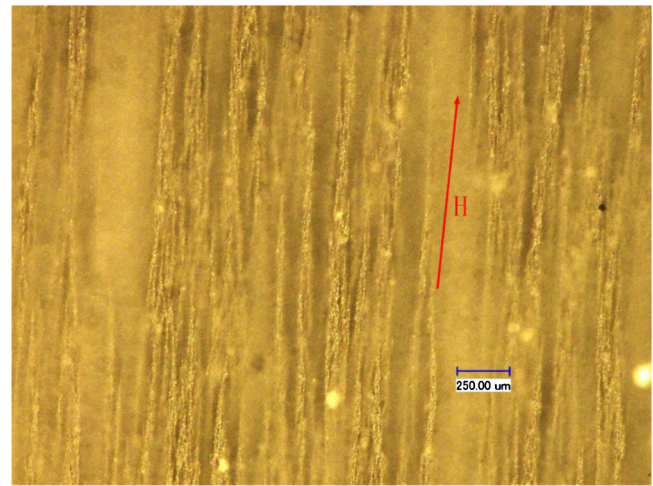


Fig. 1. A typical microstructure of MRG under a magnetic field.

different types of conductive ingredients such as metal particles, carbon blacks, carbon fibers etc. or conducting polymers such as polyaniline has been used as conductive fillers in CPCs [27]. Regardless of the type of filler, percolation theory has occupied most of the literature about the phenomenological description of the conductive binary mixtures transitive from insulator to conductor [28–31]. According to the research of Kirkpatrick [32], when the conductive contents above the percolation threshold, the conductivity (σ) of CPCs can be expressed as

$$\sigma = \sigma_0(P - P_c)^t \quad (1)$$

where σ is the conductivity of the mixture, σ_0 is the pre-factor, P is the probability of finding the conductive phase in the mixture and can be equivalent to the volume fraction of conductive phase greater than the critical concentration, P_c is the critical or percolation threshold and t is the conductivity exponent.

MRG is a kind of CPCs and consisting of a high viscosity polymer gel impregnated with ferromagnetic particles such as carbonyl iron powders (CIPs). In the absence of an applied magnetic field, the conductive ferromagnetic particles in MRG are homogeneously distributed in the matrix with an off-state viscosity, which exhibit a typical spherical structure and perform a stable property. The particles dispersed in the matrix of MRG are considered: separate, adjacent and touching [26]. When the volume fraction of particles is low, the distance between particles shall no less than $1 \mu\text{m}$ [33]. Therefore, the particles can be considered to be separated from each other as the space between each particle is filled with polymer matrix and which leads to the non-contact of particles. With the increase of particle volume fraction, the particles in MRG will coagulate to form a network which builds up the conductive paths and the MRG exhibits an insulator to conductor transition. Obviously, the percolation theory that is Eq. (1) can be used to interpreting the electrical conductivity of MRG in the process. However, the particles in MRG are soft magnetic particles and can be magnetically modelled as identical induced dipole moments. When the magnetic field is applied, the particles are magnetized instantly and can be seen as a magnetic dipole with two opposite magnetic charge [34]. Driven by the attractive force between positive and negative poles, the particles will self-assembly to form columnar or chain-like structures along with the direction of the applied magnetic field [11]. In here, a typical microstructure of polyurethane-based MRG under a magnetic field have been given to confirm this view as show in Fig. 1. It should be noted that the microstructure photograph was obtained by a digital microscope (model VHX-600, Keyence; Resolution: 1600×1200 PPI) and the

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