Effects of Multi-walled Carbon Nanotubes on the Microwave Absorbing Properties of Magnetorheological Elastomers

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Abstract. The microwave absorbing properties of carbonyl iron particles (CIPs) based magnetoheological elastomer (MRE) filled with multi-walled carbon nanotubes (MWCNTs) were examined. Absorbents including the CIPs and MWCNTs were mixed and added to the silicone rubber (SR) matrix for further compression molding and vulcanization. The complex permittivity and complex permeability were measured over a frequency range of 2-18GHz. As the MWCNT weight percentage increased, the reflection loss (RL) of the MRE composites changed significantly, which is attributed to the impedance mismatch. The MRE composites containing 28 wt% CIPs and 0.1 wt% MWCNTs with a thickness of 3 mm display dual resonance peaks with a maximum RL of -53 dB and the effective frequency bandwidth (RL<-10dB) reaches 7.7 GHz. This suggests that the MRE composites could serve as a potential candidate for absorbing incident microwave with excellent flexibility.

Keyword. Microwave absorbing, magnetoheological elastomer, multi-walled carbon nanotube, carbonyl iron particle.

1. Introduction

Rapid development of modern electronic information technology benefits the human beings, while also brings electromagnetic pollution that has gradually attracted people’s attention. As a continuation of noise, air, and water pollution, electromagnetic (EM) interference and radiation problem has become an increasingly striking concern [1-2]. Fabricating flexible absorbing composite material provides an effective way to overcome these issues due to the dielectric or magnetic loss [3].

Carbonyl iron particles (CIPs) are widely used as an effective absorbent owning to the large values of saturation magnetization and high Snoke’s limit at gigahertz [4]. Magnetorheological materials belong to a class of magnetically functional materials that undergo continuous, rapid, and reversible changes in rheological properties under an applied magnetic field. As a branch of magnetorheological material, magnetorheological elastomer (MRE) is prepared by dispersing ferromagnetic particles such as CIPs, iron oxide, etc., in a polymer. At present, the commonly used substrates are polyurethane, silicone rubber, nitrile rubber, natural rubber, etc., which have characteristics such as magnetic modulus, magneto-resistance, magnetostriction, and magnetic damping [5]. In recent years, the excellent properties of new carbon materials such as carbon nanotubes having low density, corrosion resistance, good mechanical properties, and good dielectric properties, have gained widespread attention in the field of wave absorption. Carbon nanotubes are one-dimensional carbon nanomaterials made by curling graphite sheets, processing excellent mechanical properties, thermal properties and electrical conductivity [6]. Pure carbon materials exhibit excellent dielectric loss, but no magnetic loss, resulting in poor microwave absorbing performance. Thus, combining carbon materials with magnetic metals or magnetic metal oxides, such as CIP, Fe$_3$O$_4$, etc., provides an opportunity to improve the...
impedance matching performance by adjusting the electromagnetic parameters of composite materials. The purpose of this work is to fabricate a new type of microwave absorbing material by mixing carbonyl iron powder, carbon nanotubes into curing silicone rubber and investigates the effects of MWCNTs on the microwave absorbing properties of as-prepared MRE composites.

2. Experimental Section

2.1. Raw Materials

The matrix used in this work was Silicone Rubber-704RTV was supplied by Guangdong Evergrande New Material Technology Co., Ltd Raw commercial spherical CIPs were supplied by China Metallurgical Research Institute. The MWCNTs were purchased from BeiJing DeKe DaoJin Science And Technology Co., Ltd, the Dimethicone was supplied by Shanghai Myrel Chemical Technology Co., Ltd.

2.2. Material Preparation

Weigh the required medicines according to the formula (see table 1), add the weighed CIP and CNT to the mixture of silicone rubber and silicone oil, stir the mixture at a speed of 2000 r/min, and mix after stirring for 30 minutes. The material is poured into a circular mold with a diameter of 20mm and a thickness of 1 mm. Then it is quickly placed in a vacuum drying box to extract the air from the mixture. Then the mold is placed under a tablet press and the mixture is compacted at a pressure of 2000 psi. The mixture was cured at room temperature for 12 hours. The cured absorbing material was made into a ring sample with a thickness of 1 mm, an inner diameter of 3 mm, and an outer diameter of 7 mm for testing.

| Group  | Silicone rubber: silicone oil | CIP weight percentage (wt%) | CNT weight percentage (wt%) |
|--------|-----------------------------|-----------------------------|-----------------------------|
| MRE-1  | 3:1                         | 28                          | 0                           |
| MRE-2  | 3:1                         | 28                          | 0.1                         |
| MRE-3  | 3:1                         | 28                          | 0.2                         |
| MRE-4  | 3:1                         | 28                          | 0.3                         |
| MRE-5  | 3:1                         | 28                          | 0.4                         |
| MRE-6  | 3:1                         | 28                          | 0.5                         |

2.3. Testing and Characterization

Scanning Electron Microscope-EM-30AX: COXEM; Network vector analyzer-ZNB20: ROHDE&SCHWARZ; Automatic two-way electric mixer-ZSJ-1: Jintan Xinhang Instrument Factory; Digital display vacuum drying box: Shanghai Yiheng Scientific Instrument Co., Ltd.; Tableting machine-YT-LH20D: Dongguan Yitong Testing Equipment Technology Co., Ltd.; Electronic weighing balance-FA2204B: Shanghai Jingke Tianmei Scientific Instrument Co., Ltd.

Figure 1 shows a scanning electron microscope image of the sample. From the figure, it can be clearly seen that the CIP is relatively uniformly dispersed in the matrix.

Figure 2 shows a partial energy spectrum analysis of CIP/CNT-MRE. It can be seen that the CIP content is relatively high, indicating that CIP is effectively and uniformly dispersed inside the matrix.

The absorption performance of an absorbing material can be evaluated by reflection loss (RL) (dB) [7]:

\[
RL(dB) = 20 \log \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right| 
\]  

\[
Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\varepsilon_r}} \tan \left[ j \frac{2\pi f d}{c} \sqrt{\mu_r \varepsilon_r} \right] 
\]
In the formula: $Z_{in}$ is the input impedance of the absorbing material; $f$ is the frequency of the incident electromagnetic wave; $d$ is the thickness of the absorbing material; $c$ is the propagation rate of the electromagnetic wave in vacuum; $h$ is the Planck constant; $Z_0$ is the impedance of free space.

When the value of $RL$ is less than $-10$ dB, it is equivalent to 90% of the electromagnetic waves are absorbed; while when the value of $RL$ is less than $-20$ dB, it is equivalent to 99% of the electromagnetic waves are absorbed. The requirements for wave absorbing materials can be summarized as being light in weight, thin in thickness, and having a frequency range as wide as possible, and can serve as both functional materials and structural materials.

3. Results and Discussion
As can be seen from the figure, the $\varepsilon'$ of CIP / CNT - MRE shows a downward trend as the frequency increases, but there will be rising fluctuations of different amplitudes in some frequency bands, 0, 0.1%, 0.2%, 0.3% The $\varepsilon'$ corresponding to the samples with CNT content of 0.4% and 0.5% were maintained at 3.37, 4.43, 4.76, 4.90, 5.21, and 5.40, respectively. It can be seen that as the CNT content increases, the $\varepsilon'$ of the elastomer increases. On the other hand, $\varepsilon''$ showed irregular changes with the increase of CNT content, but both showed large fluctuations in 3-5 GHz and 10.5-16.5 GHz, that is, the sample had strong absorption in the corresponding frequency band.
The $\mu'$ of CIP / CNT - MRE generally shows a trend of increasing with the increase of CNT content, and generally decreases with the increase of frequency. 0, 0.1%, 0.2%, 0.3%, 0.4% and 0.5% CNT content the maximum $\mu'$ corresponding to MRE is maintained at 0.83, 0.86, 0.87, 1.07, 1.09, and 1.10, respectively. $\mu''$ also showed irregular fluctuations with increasing frequency, and the fluctuation range remained between -0.8-0.

Figure 5 is a simulation diagram of the RL effect of a 3mm CIP / CNT - MRE. It can be seen from the figure that the RL value of the MRE with 0 CNT content is very small and there are almost no absorption peaks. After adding CNT, the RL value of the MRE increases significantly, but the RL value of the MRE does not increase gradually with the increase of the CNT content the samples with CNT content of 0.1% have the best absorbing performance. The absorption peaks of samples with different CNT content are -8.9 dB, -53 dB, -25 dB, -27 dB, -22 dB, and -24 dB, respectively. The effective absorption band width of reflection loss (<-10 dB) is 7.7 GHz. The results also confirmed that the material's absorbing performance is related to the impedance matching and the absorption performance of the material itself, that is, the maximum absorption peak will appear when the material has good absorption performance and meets the impedance matching.

4. Conclusion
In this study, we have prepared a new CIPs-based MRE composite filled with MWCNTs. The addition of MWCNTs can significantly increase the complex dielectric values of MRE composites, but shows
less influence on the complex permeability. The microwave absorption property of the MRE composite with larger content spherical CIPs and less content MWCNTs changed significantly. As the MWCNT weight percentage increased, the reflection loss (RL) of the MRE composites changed significantly, which is attributed to the impedance mismatch. The MRE composites containing 28 wt% CIPs and 0.1 wt% MWCNTs with a thickness of 3 mm display dual resonance peaks with a maximum RL of -53 dB and the effective frequency bandwidth (RL<-10 dB) reaches 7.7 GHz.

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