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Rheological properties of carbon nanotubes-reinforced magnetorheological elastomer

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Abstract. Magnetorheological elastomer (MRE) based on the natural rubber with different types of multiwall carbon nanotubes (MWCNT) as additives were synthesized. MRE with pristine MWCNTs was prepared as a control and the carboxylated (MWCNT-COOH), as well as hydroxylated (MWCNT-OH) were introduced as new additives in MRE. Their rheological properties under different magnetic field were evaluated by using the rheometer (MCR 302, AntonPaar, Austria) equipped with the electromagnetic device. The dependency of MREs towards excitation frequencies under different magnetic field was investigated. It is shown that the storage modulus and loss factor of MRE with functionalized MWCNTs exhibited noticeable increment in MR performance compared to control parallel with the frequencies increment.

1. Introduction

Magnetorheological (MR) materials have attracted interest in academic and industrial areas due to their mechanical and rheological properties can be changed and altered rapidly and reversibly by applying magnetic field [1,2]. Among MR materials family, MR elastomer (MRE) have been extensively studied due to their obvious advantage; MREs has a stable performance since the magnetic particle cannot settle with time, and also their unique mechanical performance in which their field dependent modulus compared to the other MR materials.

MREs mainly consists of non-magnetic solid-phase material such as rubber, micrometer size soft magnetic particles, and additives. There are two kinds of dispersion of soft magnetic particles in MREs namely: isotropic and anisotropic. In the stage of designing the MRE, material selection plays important roles in better performances. Few types of rubber have been used by other researchers like Silicon rubber [3,4], natural rubber [5,6] and other types of rubber [7,8]. Carbon black is frequently used as filler on fabricating the MRE. However, recently many other ingredients and additives have been explored by another researcher to get the optimum performance of MR effect for variable applications. Nanomagnetic materials have attracted the remarkable attention of researchers due to their strong interconnected three-dimensional networks at relatively lower filler loading [9].

Nayak et al., fabricated silicon rubber with dispersed 7% nano-sized carbon black and tested in the oscillation mode. It has been found that Young modulus and rheological properties of MRE with nano-sized carbon black increased with 56% and 22% compare to MRE without carbon black [10]. Li et al.

[11,12], investigated dynamic mechanical behavior silicone rubber based MRE using 1 and 3.5 wt% of multiwall carbon nanotube as additives. By conducting both shear and compression investigations, at all dynamic, MRE with MWCNT exhibit higher zero-field stiffness and damping and the dynamic mechanical properties also increased. However, more study has to be continued further to investigate the contribution of MWCNT in MRE properties. Also, functionalized MWCNTs, carboxylated (MWCNT-COOH) and hydroxylated (MWCNT-OH) also have been introduced, and their rheological properties such as storage modulus, and loss factor are investigated in details.

2. Materials and Fabrication

2.1 Raw Material

Natural rubber (NR) manufactured by Malaysian Rubber Board was selected as the matrix. Soft magnetic carbonyl iron particles (type OM) with an average diameter of 6μm purchased from BASF, Germany were used. Three types MWCNT i.e. Pristine MWCNT, carboxylated (COOH-MWCNT) and hydroxylated (OH-MWCNT) were used as an additive which obtained from Shenzhen Nanotech Port Co. Ltd, China. The outer diameter of 10-20nm and particle length in between 10-30 μm with a purity level of MWCNT used is 95%. The vulcanizing agent and the plasticizer were sulfur and epoxidized palm oil (EPO), respectively. Meanwhile, the zinc oxide (ZnO) and stearic acid were used as activators. The N Cyclohexylbenzothiazole-2-sulphenamide (CBS) used as an accelerator. Santoflex 13 used as antiozonant and as a synthetic polymer stabilizer.

2.2 MRE Fabrication

In this study, three groups of MRE samples were prepared with different types of MWCNT; MWCNT, MWCNT-COOH, and MWCNT-OH (0.1 wt%). For comparison, the reference samples consist of NR and CIP was also prepared. The fabrication process of MREs requires three steps: sonication of MWCNTs using horn type ultrasonicator, mixing the rubber and other additives via double roll mill, which ensures all components were dispersed homogeneously in the NR matrix and then vulcanized to ensure the structures were locked into place in the matrix. First, 0.1 wt% of MWCNTs were sonicated in the epoxidized palm oil (Rovpro 5300, Rovski Industries Sdn. Bhd) for 15 minutes with 10 seconds off pulse by using the horn type ultrasonicator (FB 705, Fisher Scientific (M) Sdn Bhd, Malaysia). Secondly, 100 phr rubber, 10 phr epoxidized palm oil, 30 wt% of carbonyl iron particles (CIP), 0.1 wt% of MWCNT and additives (5 phr zinc oxide, 2 phr stearic acid, 1.5 phr santoflex, 2.5 phr sulfur, and 1 phr cyclohexyl-2-benzothiazolesulfenamide were mixed via double roll mill. Finally, the MRE samples were vulcanized at 150˚C for 60 minutes under high pressure (MDR 2000).

2.3 Rheological Properties

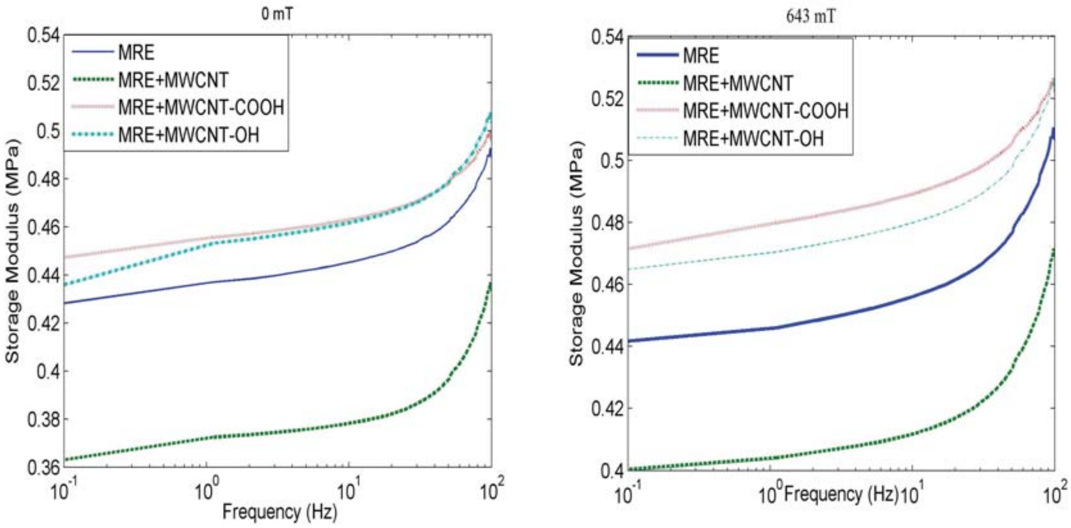
The rheological properties of MRE samples were measured by using an oscillation magnetorheological device (MRD), Rheometer (Physica MCR 302, Anton Paar, Germany) under the variable magnetic fields. The circular sample with the 1mm thickness was used and placed in the cavity base plate. Each sample has been tested in the oscillatory mode to investigate the dynamic rheological properties. The electric currents were varied at 0 and 4 A. The magnetic flux density calculated which corresponding to the 0, and 4 A were 0, and 643 mT, respectively. MR effect formula can be calculated as below;

$$\frac{G'_{\max} - G_0}{G_0} \times 100\% \qquad (1)$$

The samples were tested under the frequency sweep test. The shear strain was fixed at 0.6%, and the frequency was varied from 0.1 to 100 Hz.

3. Results and Discussion

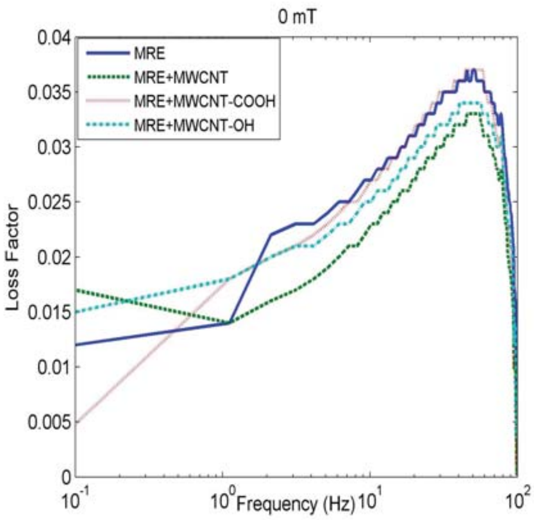
The variation of storage modulus and loss factor of the MRE samples are shown in Figure 1 (a)-(b) as a function of frequency. The linear viscoelastic response of MRE samples has been studied using constant strain amplitude of 0.6% and frequency sweep from 0.1 to 100 Hz. Figure 1(a-b) hows the storage modulus increase with the increasing of the frequency with and without magnetic field. In the case of MRE samples without MWCNT, the initial and maximum storage modulus is found to be considerably lower than the MRE samples with carboxylated and hydroxylated MWCNTs. Upon addition of 0.1 wt% of carboxylated and hydroxylated MWCNT, both storage modulus showed noticeable increment, nevertheless the pristine MWCNT stay lower trend in the region of lower and highest frequencies. The distribution of CIP and MWCNTs in the matrix was random [13] , which lead to larger moveable of CIP and MWCNTs in the matrix, resulting in higher storage modulus. In addition, the high specific areas and essential modulus of MWCNT enhance the stiffness of rubber. This phenomena also will lead to increment in the storage modulus [14].



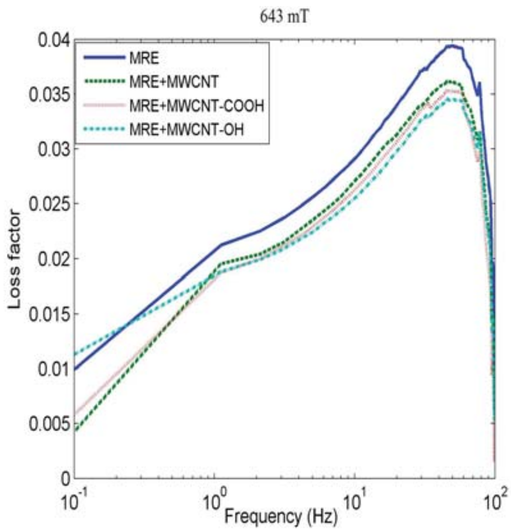
(a) **(b)**

Figure 1 Storage modulus vs frequency (a) 0 mT (b) 643 mT

Loss factor can be used to describe the ratio of the loss modulus to storage modulus. It is also related to the damping property of MREs caused by friction in materials. Figure 2 shows the relationship between the loss factor and different frequencies at room temperature. As shown in Figure 2, the loss factor of all MRE samples increased remarkably parallel with the increased of frequencies. As shown in Figure 2(a)-(b), the loss factor of MRE without MWCNT at 50 Hz slightly increased from 0.037 to 0.039 at zero field and 643 mT. The loss factor of MRE with carboxylated MWCNT showed slightly increased compared to MRE with pristine and hydroxylated MWCNTs. However at frequencies above 60 Hz, all the MRE samples with and without MWCNTs, decreased rapidly with and without magnetic field. Moreover, MRE with MWCNTs show lower loss factor as compared to MRE without MWCNT. It is due to the decreasing in the slipping displacement as a results from the strong interaction force between matrix and particles which lead to decreament in energy dissipation [15]. In addition, this result is consistent with the results of Jiang et. al [16] which natural rubber has better filler compatibility compared to other rubbers which lead to better filler matrix interaction [17] . Besides, this finding is in agreement with Ramana et al. whereby the functional groups of MWCNTs attached to the surface of MWCNTs will react with the MRE during curing process. Subsequently the covalent linkage would built which contribute to better interaction of the MRE [18]. At higher magnetic field, all the MRE samples with the MWCNTs showed almost the same trend at higher magnetic field of 643 mT.



(a)



(b)

Figure 2 Loss factor vs frequency (a) 0 mT (b) 643 mT

4. Conclusion

The rheological properties of the MRE with and without the different types of the MWCNTs have been analyzed in the present work. It has been shown that the incorporation of the different types of the MWCNTs affected towards the MR performance. The change brought by the study is tremendously important to interpret the function of the different types of the MWCNT as new additives in the MRE materials. The addition of the MWCNTs into the matrix leads to lower loss factor ratio and improved storage modulus. The MRE without MWCNT has shown the highest loss factor in both zero field modulus and at higher current induced. Rheological studies indicate that the interaction and compatibility between MRE with CIP and MWCNTs play an important role for the improvement of modulus and changes in loss factor of MRE properties.

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