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## Frequency and Amplitude Dependence of Magnetorheological Elastomers Composites

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# Frequency and Amplitude Dependence of Magnetorheological Elastomers Composites

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**Abstract.** In this study, anisotropic and isotropic magnetorheological elastomers (MREs) based on nickel zinc ferrite and natural rubber were prepared. The amount of nickel zinc ferrite was varied at five different levels (20, 40, 60, 80, and 100 phr) to assess its optimum content for enhancing dynamic mechanical performance of MREs.  $\tan \delta$  was measured through parallel plate rheometer over frequency range of 0.1-100 Hz and strain amplitude range of 0.1-6%. The results revealed that  $\tan \delta$  increased with increasing waste nickel zinc ferrite content and reached maximum value at 100 phr for both isotropic and anisotropic MREs. SEM micrograph showed that anisotropic MREs had chain-like columnar structures of magnetic particles in the matrix as a consequence of an applied magnetic field during curing. For frequency sweep test,  $\tan \delta$  for anisotropic MRE improved 244% and for strain amplitude test 159% improvement compared to isotropic MRE.

## 1. Introduction

Magnetorheological elastomers (MREs) are a new group of elastomeric composites which consist of a non-magnetic elastomeric matrix with a suspension of magnetic particles. MREs offer several distinct advantages when compared with basic rubbers. The performance of MREs mainly promote by the viscoelastic properties of the rubber matrix as do basic rubber, but inclusion of magnetic particles in rubber enables additional reinforcement through magnetic particles interaction and interfacial friction between the particles and rubber matrix. Furthermore, the stiffness and dynamic mechanical properties of the MREs can be varied by application of an applied magnetic field during fabrication or in service. MREs are being used in various applications such as adaptive tuned vibration absorbers [1], auto-motive engine mounts [2] and semi active seismic dampers [3].

MREs can be fabricated into isotropic MREs and anisotropic MREs [4]. Isotropic MREs can be characterized by having uniform dispersion of magnetic particles embedded in the rubber matrix. Anisotropic MREs have a formation of chain-like structure within the rubber matrix resulting from subjecting the materials to an external magnetic field during curing. Anisotropic MREs are found to produce materials with larger stiffness and better dynamic properties [5].

Natural rubber is generally chosen as matrix material because of its associated ease of processing, and natural rubber has the highest failure strain compared with any rubber and unbeatable in term of the dynamic properties [6-8]. The secondary raw materials from the manufacturing are abandoned regardless of the large amount of ferrite (as high as 70% in nickel zinc ferrite waste) and cannot be recycled because its composition is complicated and has many kinds of impurities. Furthermore, the waste contains heavy metal elements which could cause dangerous effect to human health and environment if it is not treated properly prior to disposal [9, 10]. Nickel zinc ferrite are soft ferromagnetic materials commonly used for manufacture of



electronic inductors, core material for power transformers in electronics, antennas and devices that are applied in the communication, lighting, alternative energy and automotive [10,11].

The incorporation of waste nickel zinc ferrite in MREs is an attractive approach from the view point of profit earning, recycling and sustainable environment. This work aims to assess the potential of nickel zinc ferrite as the magnetic filler particles for MREs. The MREs were prepared with different nickel zinc ferrite (0, 20, 40, 60, 80, 100 phr) in a natural rubber matrix. The dynamic properties of isotropic and anisotropic MREs were compared with unfilled rubber compound. The dynamic properties were investigated using lost tangent, commonly called  $\tan \delta$  under the influence of frequency and strain amplitude mode.

## 2. Materials and Methods

### 2.1 Materials

Natural rubber (grade SMR L) and other chemicals including zinc oxide, stearic acid, N-cyclohexyl-2-benzothiazole sulfonamide (CBS), tetramethylthiuram disulphide (TMTD), paraffin oil, sulphur and N-Isopropyl-N'-phenyl-P-phenylenediamine (IPPD) were purchased from Zarm & Chemical Supplier Sdn Bhd. Nickel zinc ferrite with average size of 6  $\mu\text{m}$  was obtained from ACME Ferrite Products (M) Sdn. Bhd.

### 2.2 Preparation of nickel zinc ferrite with natural rubber MREs

Ferrite was first washed with water and gently stirred in a container. This step was repeated until no foreign particles left on the surface of the water. Then, the nickel zinc ferrite was filtered and dried at 150 °C in an oven until a constant weight was achieved. The compound formulation is given in Table 1. Formulations were compounded using a conventional laboratory two roll mill (model XK-150) according to ASTM D3184-80. The compounding began with mastication of the rubber for 2-3 minutes. Additives (other than accelerators and sulphur) were then added followed by waste nickel zinc ferrite; addition of accelerators and sulphur were delayed to the last part of the process to prevent premature vulcanization during compounding. Compounded rubber samples weighing 4 g were used to determine the crosslinking time ( $t_{90}$ ) by using Mosanto Rheometer (MDR 2000) under temperature of 150 °C.

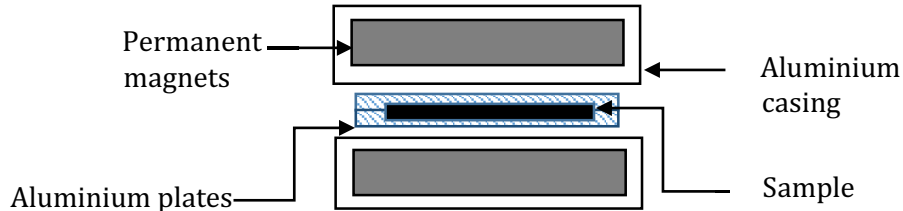
**Table 1.** Formulation of rubber compounds

Material	Function	Content (phr*)
Natural rubber	Matrix	100
ZnO	Activator	5
Stearic acid	Activator	1
Nickel Zinc Ferrite	Filler	0, 20, 40, 60, 80, 100
Paraffin oil	Plasticizer	4
CBS	Accelerator	2
TMTD	Accelerator	1
IPPD	Antioxidant	2
Sulphur	Crosslinking agent	1.5

\*parts per hundred rubber

Compounded rubber samples weighing 45 g were placed in a mould 124 × 70 × 3 mm. The anisotropic MREs were pre-cured in a specially developed magnetic mould (as shown in Figure 1) at 80 °C for 30 minutes to align in the magnetic particles along the direction of magnetic mould

and subsequently were locked into place upon curing in a compression moulder at 150 °C under a pressure of approximately 12 MPa.



**Fig.1.** Sketch of specially developed magnetic mould for MRE curing

### 2.3 Morphology test

The microstructure of isotropic MRE and anisotropic MRE were observed using a field emission scanning electron microscopy (SUPRA<sup>™</sup> 35, V, ZEISS USA)). The samples were immersed in liquid nitrogen before being fractured using two pliers in order to bare their interior. The surfaces were coated with a thin layer of gold prior to observation at an accelerating voltage of 20 kV.

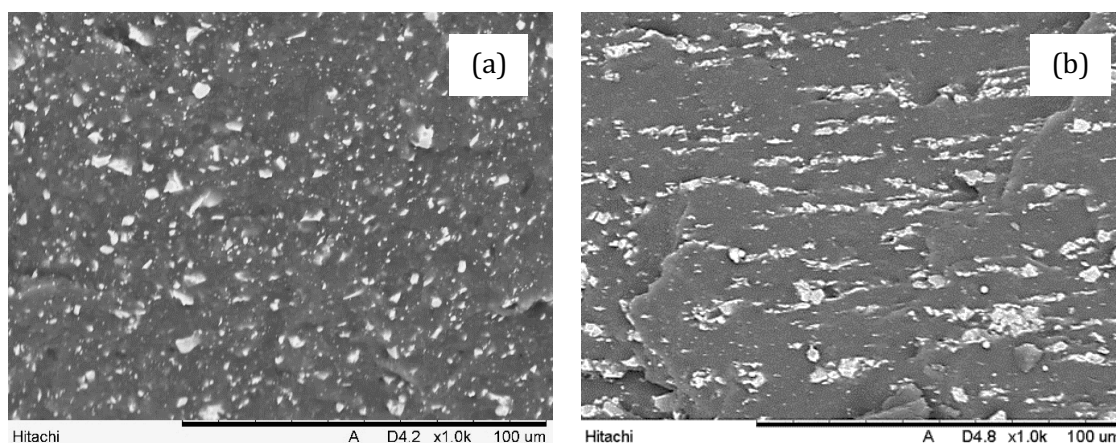
### 2.4 Dynamic mechanical properties test

The dynamic mechanical properties of rubber composites were measured using parallel and plate rheometer (MCR 301, Anton Paar Company, Germany) on a circular specimen of 20 mm diameter and 3 mm thickness in shear mode at room temperature. The dynamic mechanical properties were investigated using loss tangent, commonly called  $\tan \delta$ .  $\tan \delta$  is the comparison of the energy loss to that stored and is obtained by dividing loss modulus ( $G''$ ) with the storage modulus ( $G'$ ).  $\tan \delta$  was measured over the frequency range of 0.1-100 Hz at a fixed strain amplitude of 2%. For the influence of strain amplitude on  $\tan \delta$ , the samples were measured at strain amplitude range of 0.1-6% at a fixed frequency of 10 Hz.

## 3. Results and discussion

### 3.1 Morphology

Figure 2 shows SEM images of isotropic and anisotropic MREs. It can be observed that isotropic MRE have a homogenous dispersion of nickel zinc ferrite particles embedded in a natural rubber matrix (Fig. 2a). Figure 2b shows anisotropic MRE cured under an applied magnetic field. It can be seen that the alignment of magnetic particles has occurred in the rubber matrix due to the influence of magnetic force to the filler particles in which the north pole of one particle attracts the south pole of its neighbor, thus resulting the formation of chain-like structures inside the rubber matrix.



**Fig. 2.** SEM images of surface of; (a) isotropic MRE, (b) anisotropic MRE

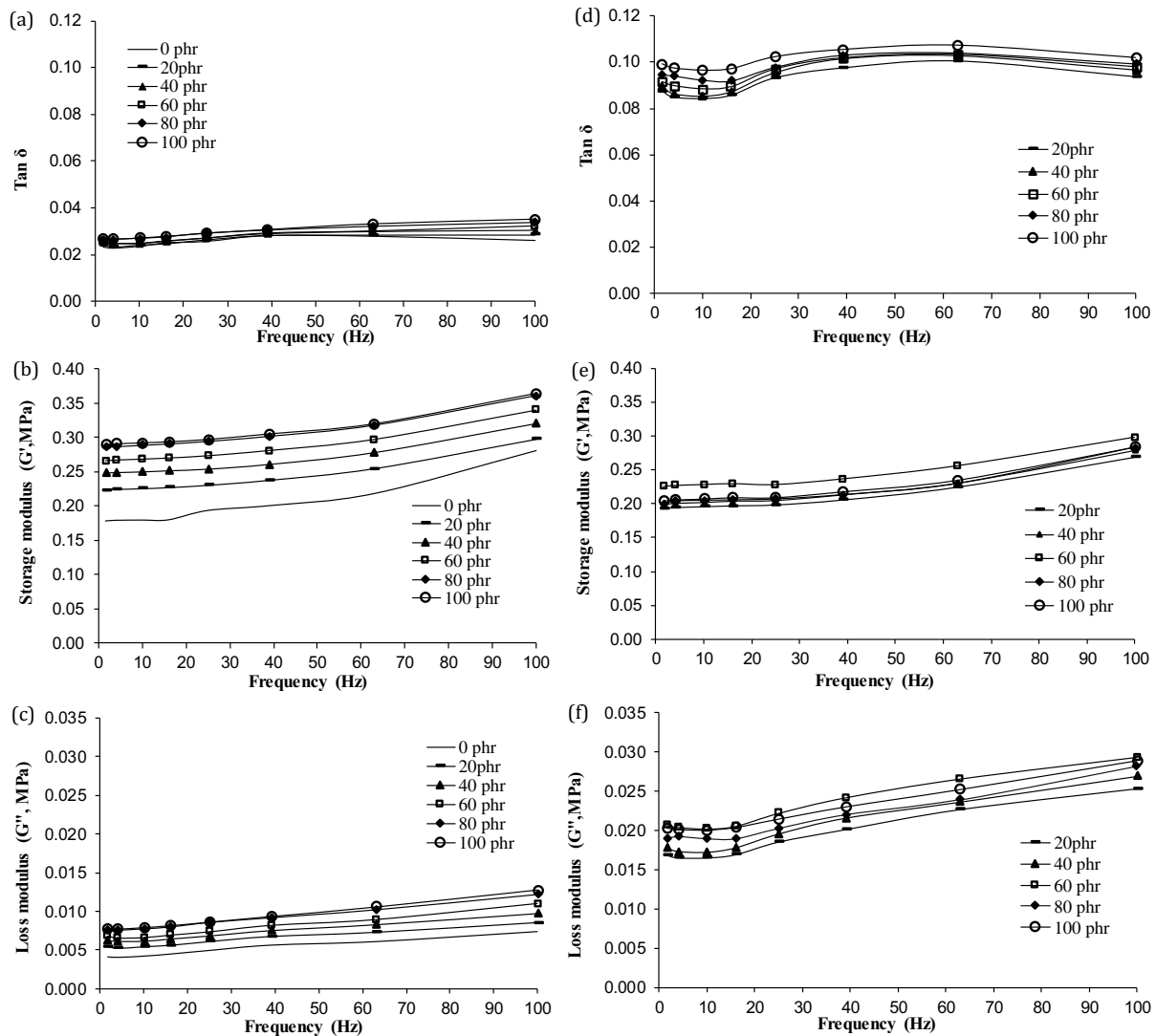
### 3.2 Dynamic mechanical properties

#### 3.2.1 Frequency sweep measurement

The variation of  $\tan \delta$  with frequency for isotropic and anisotropic MREs at different waste nickel zinc ferrite contents is shown in Figure 3. Generally,  $\tan \delta$  in Figures 3a and 3d gradually increased with increasing frequency for both isotropic and anisotropic MREs and slightly decreased at high frequency for anisotropic MREs. However,  $\tan \delta$  of anisotropic MREs was higher compared with isotropic MREs with improvement of approximately 250%, 245%, 247%, 235%, and 243% for 20, 40, 60, 80, and 100 phr of filler, respectively. It is also apparent that the  $\tan \delta$  for anisotropic and isotropic MREs increased with increasing nickel zinc ferrite contents. The changes in  $\tan \delta$  can be further explained with storage modulus ( $G'$ ) and loss modulus ( $G''$ ).

Storage modulus ( $G'$ ) and loss modulus ( $G''$ ) are plotted in Figure 3b and 3c for isotropic MREs and Figure 3e and 3f for anisotropic MREs.  $G'$  and  $G''$ , similar to  $\tan \delta$ , were frequency dependent and the increase in  $\tan \delta$  as the frequency increased was mainly due to the increased in  $G''$  as opposed to  $G'$ . The increase of  $G''$  as the frequency increased for isotropic MREs could be due to increase of energy absorbed through stretching and recovery of rubber molecular chain during viscoelastic flow under dynamic loading and interfacial damping between the particles and rubber matrix [12]. For anisotropic MREs, similar mechanism would be involved with additional energy absorbed through magnetism induced damping as the formation of chain like columnar structure in anisotropic MRE improved damping [13]. This phenomenon could be attributed to separation of dipole-dipole interaction between neighbouring particles.

The increase of  $G''$  with increasing waste nickel zinc ferrite content could be explained by the increased in energy absorbed due to interfacial friction caused by the increased in the interfacial area and the volume with the increase in waste nickel zinc ferrite content. It can also be seen that  $G'$  increase with increasing waste nickel zinc ferrite content for both isotropic and anisotropic MREs. The increment of  $G'$  for isotropic MREs could be related to efficient stress transfer between homogeneously distributed particles and rubber matrix which improved stiffness and strength of the material; and for anisotropic MREs, the increase of  $G'$  could be attributed to efficient reinforcing effect provided by more bulkier chains and columnar structures as the volume of the waste nickel zinc ferrite increased in the rubber matrix.



**Fig. 3.** (a)  $\tan \delta$ , (b)  $G'$ , and (c)  $G''$  of isotropic MREs and (d)  $\tan \delta$ , (e)  $G'$ , and (f)  $G''$  of anisotropic MREs

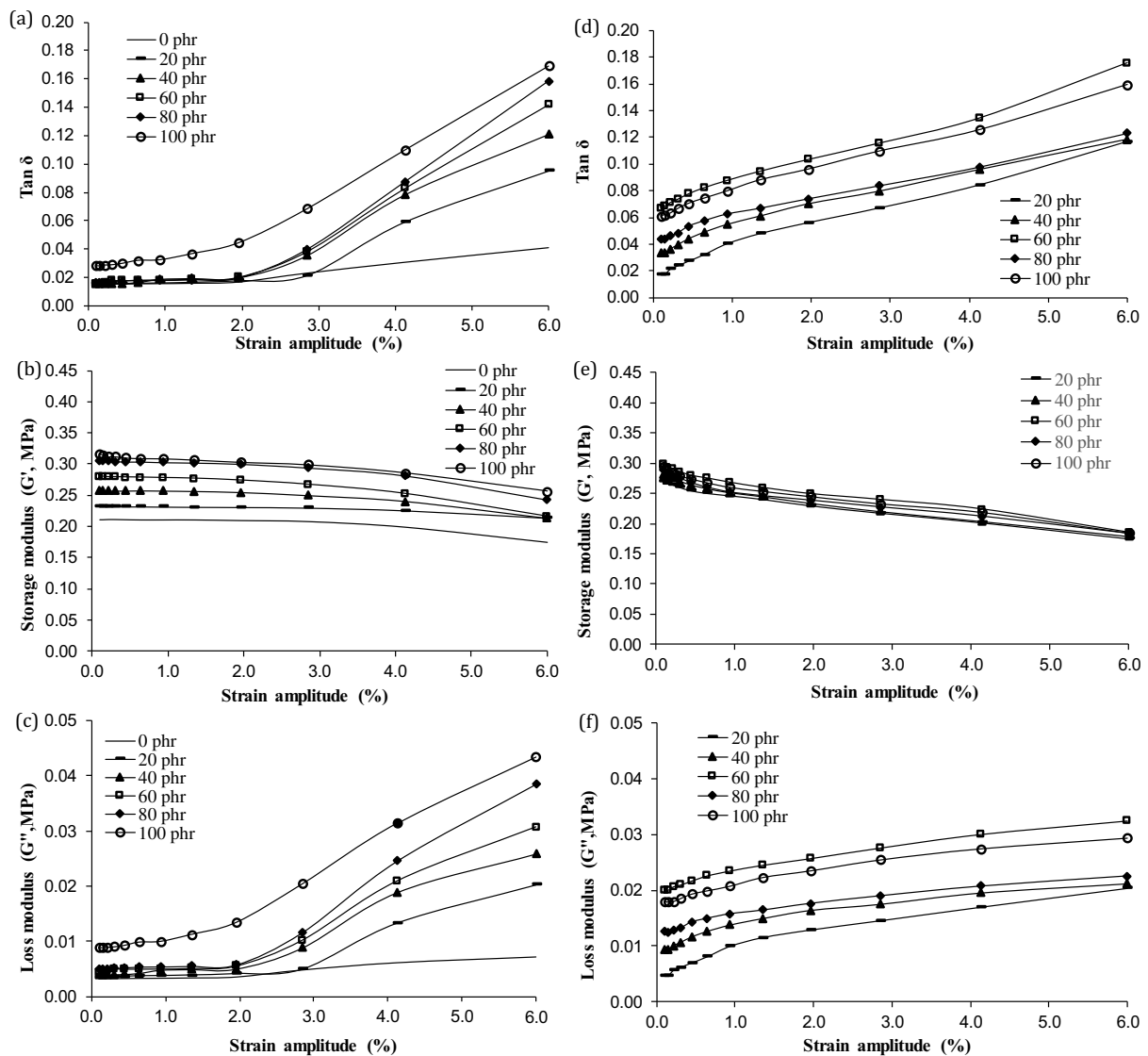
### 3.2.2 Strain amplitude measurement

The variation of  $\tan \delta$ ,  $G'$ ,  $G''$  with strain amplitude for isotropic and anisotropic MREs at different waste nickel zinc ferrite contents are shown in Figure 4. The relationship between  $\tan \delta$  and strain amplitude for isotropic (Fig. 4a) and anisotropic (Fig. 4d) MREs is approximately linear, with  $\tan \delta$  increased with increasing strain amplitude up to 6%. Again, it is apparent that the changes in  $\tan \delta$  with increased strain amplitude is mainly due to the increase in  $G''$ . Besides, it can be seen that;  $\tan \delta$  and  $G''$  for anisotropic MREs were higher compared to isotropic MREs at low strain amplitude up to 2.5%, and the values of  $\tan \delta$  for isotropic and anisotropic MREs were comparable at high strain amplitude.

The amplitude dependence for isotropic MREs is related to Payne effect [14,15]. This effect can be explained by the breakdown of filler aggregates to release trapped and occluded rubber which allows more rubber to take part in energy dissipation, filler-rubber detachment, and reformation to increase at higher strain amplitude. For anisotropic MREs similar mechanisms would be

involved as well as magnetism induced damping. The higher  $\tan \delta$  and  $G''$  for anisotropic MRE at low strain amplitude supports that additional energy absorbed due to increase of energy required to breakdown interparticle magnetic interaction between neighboring particles.

At high strain amplitude, the magnetic interparticle interaction become weak and energy loss is largely reliant of the viscoelastic damping in the rubber matrix and the Payne effect. It can also be seen  $\tan \delta$ ,  $G'$  and  $G''$  for isotropic MREs increased with increasing waste nickel zinc ferrite, however, for anisotropic MRE,  $\tan \delta$ ,  $G'$  and  $G''$  increased with increasing waste nickel zinc ferrite up to 60 phr. The decrease in  $\tan \delta$ ,  $G'$  and  $G''$  at higher filler content could be attributed to poor filler-filler interaction in a bulkier chain like columnar structures and filler-rubber interactions, it is easily disrupted as the strain amplitude increased, and the magnetism induced damping also reduced [16].



**Fig. 4.** (a)  $\tan \delta$ , (b)  $G'$ , and (c)  $G''$  of isotropic MREs and (d)  $\tan \delta$ , (e)  $G'$ , and (f)  $G''$  of anisotropic MREs

#### 4. Conclusions

It was found that isotropic MREs had uniform waste nickel zinc ferrite particles distribution and alignment of magnetic particles occurred for anisotropic MREs as a consequence of curing the material under an applied magnetic field. The results revealed that  $\tan \delta$  was highest for anisotropic MREs with improvement of 244% compared with isotropic MREs over the whole frequency range explored and 159% improvement over the whole strain amplitude range explored, supporting that formation of chain like columnar structures contributing to higher mechanical performance which might be due to viscoelastic damping, interfacial damping as well as magnetism induced damping. The optimum filler content was found to be 100 phr for both isotropic and anisotropic MRE.

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