

Investigation on magnetic field dependent modulus of epoxidized natural rubber based magnetorheological elastomer

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Abstract. This paper presents an investigation on the use of epoxidized natural rubber (ENR) as a matrix of magnetorheological elastomers (MREs). Isotropic ENR-based MRE samples were synthesized by homogeneously mixed the ENR compound with carbonyl iron particles (CIPs). The microstructure of the sample was observed, and the magnetic field-dependent moduli were analyzed using rheometer. The influences of excitation frequency, CIPs content and magnetic field on the field-dependent moduli of ENR-based MREs were evaluated through dynamic shear test. The microstructure of MRE samples demonstrated the dispersed CIPs in the ENR matrix. The remarkable increment of storage and loss moduli of the ENR-based MREs has exhibited the magnetically controllable storage and loss moduli of the samples when exposed to the magnetic field. Consequently, the CIPs content, frequency and magnetic field were significantly influenced the dynamic moduli of the ENR-based MREs.

1. Introduction

Magnetorheological elastomers (MREs) have been attracting research interests since rheological properties of MREs corresponding to field-dependent modulus can be controlled by adjustment of magnetic field. The MREs comprise of rubber elastomer and magnetizable particles which contribute to no leakage and settlement of particles as occur in magnetorheological fluids (MRFs) [1]. These benefits provide a wide range of potential applications for MREs, particularly in any applications that use rubber to control vibration and noise such as engine mount, building isolator, prosthetic device, tunable vibration absorber and etc. The magnetizable particles are frequently discussed which refer to the content, shapes and sizes of the magnetic particles. Results showed that all of these parameters have greatly influenced the performance of MREs. Soft spherical magnetizable particles with micron-sized are the most common particles that studied by researchers as these particles have low magnetic remnant, high magnetic permeability and magnetic saturation [2-3]. These characteristics contribute to high magneto-induced storage modulus. In terms of magnetic particle dispersion, the MREs can be classified into two categories which are known as anisotropic and isotropic MREs. The anisotropic



MRE is denoted as the one that contains aligned chain-like structure of magnetic particles as the magnetic field is applied during pre-configuration. In case of isotropic MRE, the magnetic particles are uniformly distributed in the matrix. The fabrication of the anisotropic MREs requires modification of the conventional rubber molding equipment's for pre-configuration stage and the chain direction must be taken into consideration for utilizing in any devices [4]. Due to this matter, the isotropic MRE was chosen in this study.

Despite of magnetic particles, previous studies also revealed that the selection of materials for MRE matrix is crucial. Many efforts on synthesizing the MREs using different types of rubber materials such as silicone rubber (SiR), natural rubber (NR), polyurethane (PU), cis-polybutadiene rubber (BR), nitrile rubber (NiR) and thermoplastic elastomer have been performed so as to implement the MREs in those applications [2]. However, fabrication of the MREs that encompasses desirable stiffness and damping properties are the most challenging part in this field. Compared to other types of matrices, more studies have been carried out on using the SiR as MRE matrix due to unnecessary of vulcanization. It is known as a soft elastomer matrix that can reach a high relative MR effect, but the magneto-induced storage modulus is not large enough which confines their practical applications. Other than SiR, the NR-based MREs were also showed interesting outcomes corresponding to higher magneto-induced storage modulus and degradation stability than the SiR [5]. Nevertheless, poor temperature, weather and oil resistance of NR lead to synthesis of the MREs containing dissatisfactory properties [2, 6].

Therefore, the researchers have chemically amended the NR through a process recognized as epoxidation in order to enhance the properties of the NR. The epoxidation process occurs in such a way that NR latex are chemically reacted with hydrogen peroxide or peracetic acid, and change the carbon-carbon double bonds into epoxy groups [7]. Epoxidized natural rubber (ENR) is the product of this process, and composed of a combination of natural and synthetic rubber properties. For instance, the ENR has high strength due to their ability to undergo strain crystallization and high degree of damping which are anticipated for synthesizing the MREs [8]. In fact, the epoxidation process improves the compatibility of NR with polar polymers and wears resistance. Similar to the NR, the ENR is also considered as eco-friendly material that derived from renewable resources and lead to less carbon footprint. With growing concerns of global climate change and because of its unique properties, the ENR becomes more prominent recently. Nonetheless, the ENR is not widely explored and there has been a few works focusing on utilizing the ENR as MREs matrix [9]. Therefore, the objective of this study is to investigate and analyze the field-dependent storage and loss moduli of the ENR-based MREs under shear deformation. A total of three isotropic ENR-based MREs were fabricated without application of magnetic field. The effect of magnetizable particles content, excitation frequency and magnetic field will be discussed in details.

2. Experimental Procedure

2.1. Fabrication of MRE and Characterization

Epoxidized natural rubber (ENR) 25 and carbonyl iron particles (CIPs) were used as MRE matrix and magnetizable particles in this study. The ENR 25 has a glass transition temperature of -45°C , and the density of CIPs is 7.86 g/mL . Generally, the MRE samples were prepared by three stages of mixing process; (a) addition of additives that include ZnO, stearic acid, carbon black and aromatic oil, (b) addition of sulfur and CBS, and finally (c) addition of CIPs. The mixing process was performed via conventional double-roll mill. After mixing, the mixture undergo curing process without magnetic field which was carried out at 150°C for 15 minutes by compressing the mixture in a mold.

The microstructure observation was carried out using Field Emission Scanning Electron Microscopy (FESEM, Supra 40VP-31-31, Germany). The cross-section of the selected sample was observed at 500x and 2000x magnifications using voltage acceleration of 20 kV. The samples were treated with gold coating prior to the observation.

The rheological measurements were performed experimentally using a Rotational Rheometer (MCR 302, Anton Paar Company, Germany) equipped with MRD 70, which can produce the magnetic field

perpendicularly to the shear flow in the test area. A temperature control device (Viscotherm VT2, Anton Paar, Germany) was utilized for controlling the measuring temperature to be at 25 °C. The samples with diameter of 20 mm were placed between a rotary disk and a parallel base plate. In this study, an oscillatory shear was used for the entire rheological experiments.

The strain sweep test was performed in order to identify the linear viscoelastic region of a sample prior to frequency sweep measurement. The measurement was done by varying the strain amplitude from 0.1 to 10% at a constant frequency of 1 Hz for MRE sample that contained 50 wt% CIPs. The range of frequency at 0.1 to 100 Hz was applied for frequency sweep measurement. Meanwhile, the strain amplitude was kept constant at 0.4% in case of all samples. In order to analyze the effect of magnetic field, the current was increased from 0 to 730 mT by increment of approximately in the range of 150-350 mT at each measurement. The magnetic field values for each current were recorded using teslameter. The rheological measurements were conducted at room temperature.

3. Results and discussion

3.1. Microstructure Observation

Figure 1(a) and (b) illustrate the microstructure of the ENR-based MRE for 50 wt% CIPs at 500x and 2000x magnifications, respectively. Noticeably, the CIPs were available in spherical and white gray color, while the ENR matrix was black in color. From Figure 1(a), it is apparent that the CIPs were scattered uniformly in the ENR matrix, but some agglomerated CIPs were also observed. In addition, the porosities were also seen in the sample. The porosities were occurred due to a chemical reaction amongst ENR, additives and CIP during vulcanization at high temperature. Based on Figure 1(b), the average diameter of CIPs was determined to be at approximately 3-4 μm . It can be concluded that the CIPs tend to adhere to the ENR matrix and subsequently played a crucial role in inducing the magnetic flux density in the MRE samples. In other words, the magnetic forces of CIPs in the ENR-based MREs were dependent on the adhesion and distance between CIPs which will be discussed in next section.

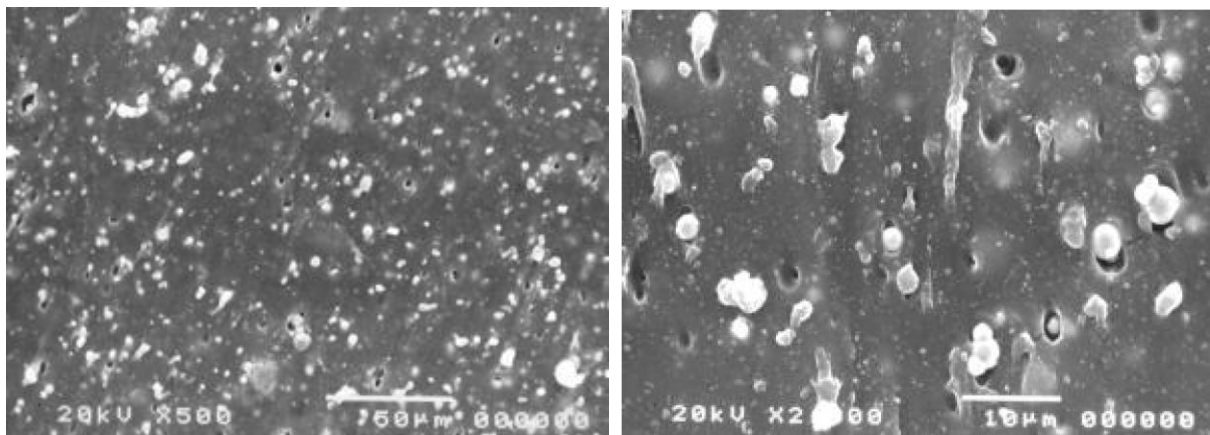


Figure 1. Microstructure observation at: (a) 500x and (b) 2000x of magnification for ENR-based MRE samples containing 50 wt%.

3.2. Rheological properties

3.2.1. Effect of Strain Amplitude

The MREs are classified as viscoelastic materials. In the viscoelastic materials, some part of the shear deformation energy is stored temporarily which could be recovered after each cycle, and the rest of the energy is dissipated as heat. The ability of storing the energy temporarily is referred as stiffness and called as storage modulus (G'). In the meantime, the viscous properties or loss modulus (G'') represents the capability of dissipating the deformation energy in the form of heat. Fundamentally, the G' and G''

are parameters which describe the rheological properties of MREs. In this paper, selected plots of storage and loss moduli as a function of strain amplitude and oscillation frequency are described without and with the application of magnetic field.

Figure 2 displays the influence of strain amplitude on the storage and loss moduli of ENR-based MRE sample with 50 wt% of CIPs content. Based on this figure, it is apparent that the ENR-based MREs were dependent on strain. Similar to other viscoelastic materials, the storage modulus and loss modulus of the ENR-based MREs was constant up to certain strain amplitude which is signified as linear viscoelastic region. Subsequently, the storage modulus was diminished while the loss modulus was increased as shear strain amplitude decreased. At this point, the structure of polymer network in the samples started to disrupt and this region is called as non-viscoelastic region. Therefore, it is significant to determine the viscoelastic region to avoid from measuring the frequency sweep at region whereby the structure was damaged, and MRE materials become more viscous rather than elastic solid. The viscoelastic region of ENR-based MRE samples was up to approximately 0.4% of strain amplitude.

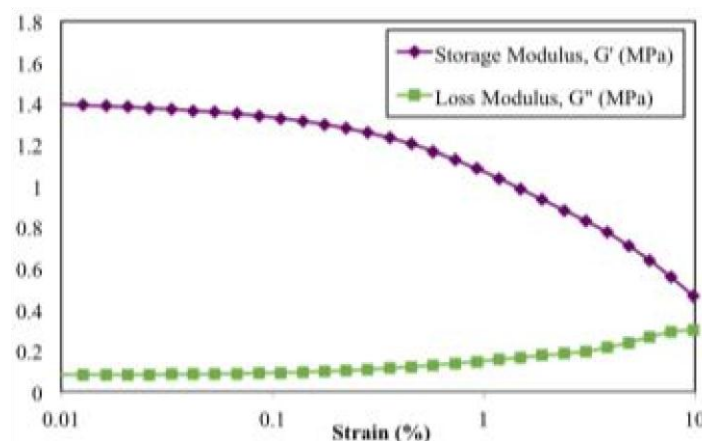


Figure 2. The effect of strain amplitude on the storage and loss modulus of ENR-based MRE for 50 wt% CIPs

3.2.2. Effect of Frequency

Figure 3 (a)-(d) demonstrate the effect of frequency on the storage modulus and loss modulus at various CIPs content and magnetic field density. These figures apparently depicted that both moduli were increased with increasing of frequency. As frequency increased from 0.1-100 Hz, the storage modulus was augmented non-linearly from about 0.65 to 0.82 MPa, 0.71 to 0.92 MPa and 0.86 to 1.18 MPa for MRE samples containing 10, 30 and 50 wt% of CIPs, respectively. Similarly, the loss modulus was remarkably increased from approximately 0.025 to 0.12 MPa, 0.026 to 0.14 and 0.05 to 0.17 MPa for 10, 30 and 50 wt% of CIPs, respectively. This is due to the fact that high shear stress was formed at the surface and in the MRE samples during shear deformation particularly at high excitation frequency. Consequently, the ENR-based MRE samples were stiffened, and resulted in increasing of storage modulus. Furthermore, the friction that occurred at the interface between CIPs, and CIPs with the ENR matrix was rose with frequency, leading to increase of loss modulus.

In the meantime, the CIPs content and magnetic field density also significantly augmented both of the moduli. Based on Figure (a) and (c), the small increment of storage and loss moduli were observed as CIPs content rose from 10 to 30 wt%. Meanwhile, both moduli were increased significantly when CIPs content increased to 50 wt%. The same trend of the graph was observed in Figure 3 (b) and (d), in which the storage and loss moduli were enhanced steadily and remarkably as the magnetic field increased from 383 to 483 mT and 483 to 730 mT, accordingly. Besides that, the storage and loss moduli were detected to be the lowest without application of magnetic field. These results indicated that adjusting the applied magnetic field could control the storage and loss moduli. The growth of the moduli with CIPs content and applied magnetic field indicated that the dipole-dipole interparticle magnetic forces among

CIPs in the samples were enlarged. In other words, the magnetic forces were enhanced due to the mitigation of interparticle distance among CIPs as more CIPs were loaded.

Moreover, the increment of the applied magnetic field on the samples resulted in more magnetic field density induced in the samples. Therefore, addition of CIPs and applied magnetic field also contributed to increment of storage and loss moduli as similar to the excitation frequency. The rise of the storage and loss moduli for different wt% of CIPs could be guidance in manufacturing the ENRbased MRE samples with the desirable properties.

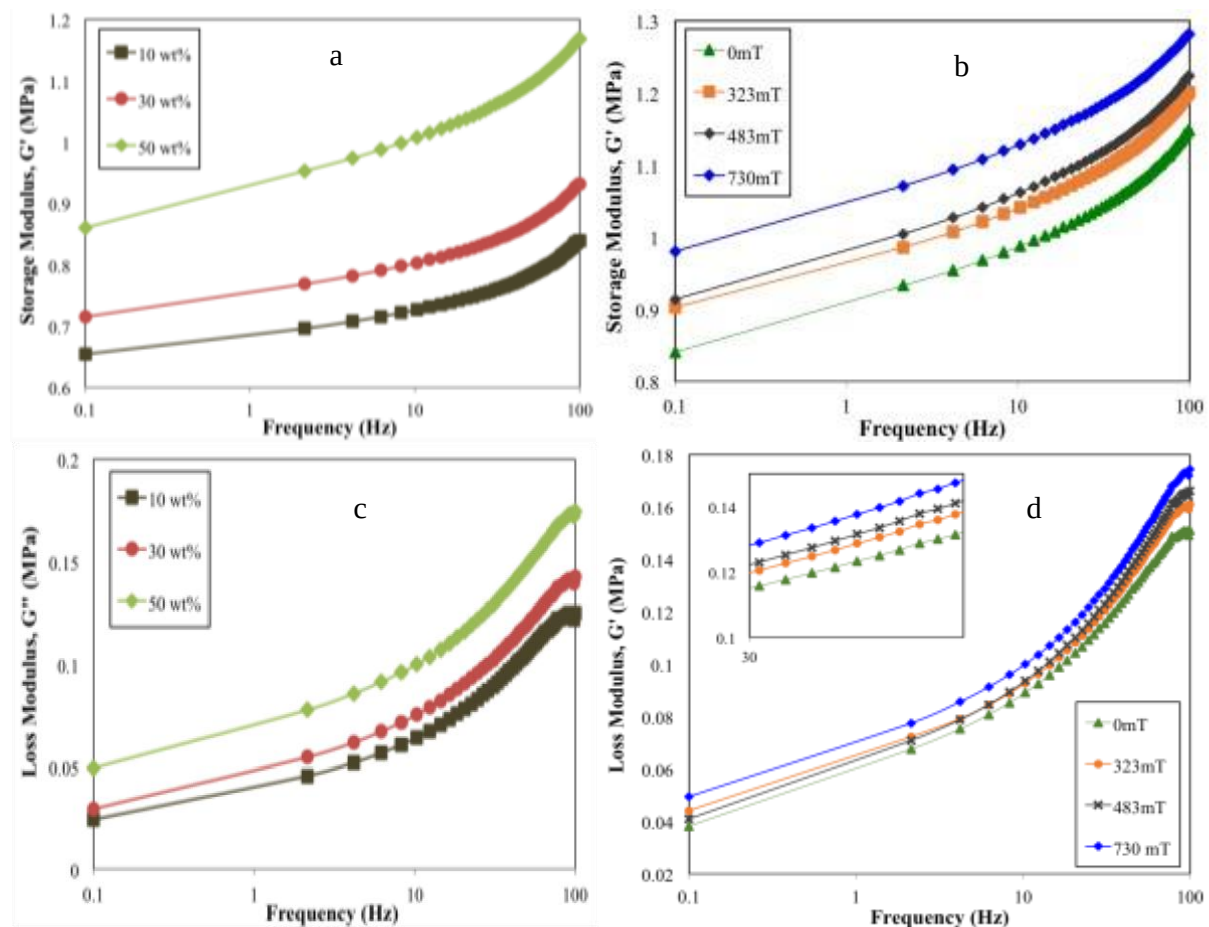


Figure 3. Frequency effect on the storage modulus and loss modulus at various: (a) and (b) CIPs content, and (c) and (d) magnetic field density

4. Conclusion

In this study, the new type of rubber known as epoxidized natural rubber (ENR) was employed as MRE matrix. The microstructure of the ENR-based MREs proved that the CIPs were dispersed uniformly in the ENR matrix which referring to isotropic MRE characteristics. The ENR-based MREs were dependent on strain, and the viscoelastic region of ENR-based MRE sample was 0.4% of strain amplitude. Determination of the viscoelastic region is important to prevent from performing the frequency sweep measurement at the region whereby the network structure in the MRE samples was disrupted. The results of frequency sweep indicated that the storage and loss moduli were dependent on the frequency, magnetic field and essentially relied on the CIPs content. Other than that, both moduli were obviously increased with increasing of these parameters. In other words, the rise of dipole-dipole interparticle magnetic forces and friction between CIPs contributed to the increment of both moduli. The outcomes indicated that adjusting the parameters could control the magnetic field-dependent modulus of

ENR-based MREs. Other than that, the results related to CIPs content could be guidance in manufacturing the ENR-based MRE samples to suit certain requirements for applications.

5. Acknowledgement

The authors gratefully acknowledge the financial funded by the Ministry of Higher Education Malaysia PRGS Grant (Vote No: 4L667) and Universiti Teknologi Malaysia under GUP (Vote No: 13H55).

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