INVESTIGATION OF DYNAMIC MECHANICAL PROPERTIES OF MAGNETORHEOLOGICAL ELASTOMER BASED ON NICKEL ZINC FERRITE AND NATURAL RUBBER

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by

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LIST OF SYMBOLS

G'	Storage modulus
G''	Loss modulus
H_c	Coercive force
mT	Militesla
m_{dry}	Dry mass of MREs
m_{wet}	Swollen equilibrium mass
M_1	Original mass of MREs rubber
M_2	MREs rubber swollen mass
M_H	Maximum torque
M_L	Minimum torque
M_s	Saturation magnetization
M_r	Remanence
t_{s2}	Scorch time
t_{90}	Cure time
$\tan \delta$	Tan delta
T_g	Glass transition temperature
V_o	Molar volume of toluene
V_p	Volume fraction of the particles
V_r	Volume fraction of MREs
ρ_r	Density of natural rubber
ρ_s	Density of toluene
X	Interaction parameter between rubber and toluene
δ	Phase angle
$[\chi]$	Crosslink density

LIST OF ABBREVIATIONS

APTES	(3-aminopropyl) triethoxy silane
ASTM	American Society of Testing and Materials
BR	Butadiene rubber
CBS	N-cyclohexyl-2-benzothyazolsulfenamide
DMA	Dynamic Mechanical Analysis
DOP	Dioctyl Phthalate
DPG	Diphenyl guanidine
EP	Epoxy resin
FESEM	Fourier emission scanning electron microscope
FTIR	Fourier Transform Infrared Spectoscopy
IPPD	N-isopropyl-n'-phenyl-p-phenylenediamine
ISO	International Standards Organisations
LVE	Linear viscoelastic
MBT	Mercaptobenzothiazole
MR	Magnetorheological
MREs	Magnetorheological elastomers
MRFs	Magnetorheological fluids
M100	Modulus at 100% elongation
PANI	Polyaniline
phr	Parts per hundred rubber
PU	Polyurethane
SEM	Scanning Electron Microscope
SMR	Standard Malaysian Rubber
TESPT	Bis [3-(triethoxysilyl)propyl] tetrasulfide
TGA	Thermal gravimetric analysis
TMTD	Tetra-methylthiuram disulphide
TSR	Technically Specified Rubber
VSM	Vibrating sample magnetometer

PENYIASATAN SIFAT MEKANIK DINAMIK ELASTOMER MAGNETOREOLOGI BERDASARKAN NIKEL ZINK FERIT DAN GETAH ASLI

ABSTRAK

Elastomer magnetorheologi (MREs) adalah kelas komposit yang terdiri daripada matrik elastomerik dengan partikel magnet tertanam. Prestasi MREs dipengaruhi oleh sifat viskoelastik matrik getah, interaksi antara muka partikel magnet dan matrik getah dan mekanisme tambahan melalui interaksi partikel magnet. Dalam kajian ini, MREs berasaskan getah asli dan sisa nikel ferit telah dihasilkan. Komponen individu dalam MREs yang mempengaruhi prestasi dan penyerapan tenaga bahan tersebut telah disiasat. MREs dengan kelikatan matrik getah yang berbeza dan sisa industri nikel zink ferit telah disediakan untuk mengkaji kesan sifat viskoelastik matrik getah kepada prestasi dinamik dan mekanikal. Hasil dari kajian ini menunjukkan bahawa, tan δ meningkat dengan peningkatan kelikatan matrik dalam julat frekuensi dan amplitud ketegangan yang diuji. Kajian ini juga mendapati kekuatan dan pemanjangan tegangan pada rehat meningkat dengan peningkatan kelikatan matrik. Mikrograf pengimbas mikroskop elektron (SEM) menunjukkan bahawa struktur kolumnar menjadi lebih panjang dan tebal dengan penurunan kelikatan matrik. Walau bagaimanapun banyak rongga kekal terhasil daripada partikel magnet yang terkeluar menunjukkan bahawa interaksi yang lemah antara sisa nikel zink ferit dan matrik getah. Untuk menilai kesan interaksi antara partikel magnet dan matrik getah terhadap prestasi dinamik dan mekanikal MREs, bis [3- (triethoxysilyl) propil] tetrasulfida (TESPT) telah digunakan untuk mengubah permukaan nikel zink ferit. Kandungan TESPT diubah pada 0, 2, 4, 6 dan 8% untuk kelikatan matrik getah yang rendah dan tinggi. Ikatan antara muka yang lebih baik telah dibuktikan oleh spektroskopi Fourier infra-merah (FTIR), peratusan ikatan cantuman, kepadatan sambung silang dan gambar SEM. Hasilnya menunjukkan bahawa tan δ meningkat lebih kurang 30% untuk julat frekuensi dan amplitud tegangan diuji untuk kelikatan matrik getah rendah dan tinggi. Interaksi yang lebih baik juga meningkatkan sifat tegangan MREs dan kandungan optimum TESPT didapati pada 6 wt%. Kesan interaksi antara partikel magnet terhadap prestasi dinamik dan mekanik MREs telah disiasat dengan pemvulkanan bahan pada medan magnet 0, 100, 150, 165 dan 200 mT. Hasilnya menunjukkan bahawa tan δ meningkat apabila medan magnet meningkat dan kekal di titik ketepuan magnet pada 165 mT. Walau bagaimanapun, kekuatan tegangan didapati menurun dengan medan magnet yang semakin meningkat disebabkan oleh arah beban tegangan berserenjang dengan penjajaran partikel magnet.

INVESTIGATION OF DYNAMIC MECHANICAL PROPERTIES OF MAGNETORHEOLOGICAL ELASTOMER BASED ON NICKEL ZINC FERRITE AND NATURAL RUBBER

ABSTRACT

Magnetorheological elastomers (MREs) are a class of composite that consist of elastomeric matrix with embedded magnetic particles. The performance of MREs can be ascribed to viscoelastic properties of rubber matrix, interfacial interaction at the interface between the rubber matrix and magnetic particles as well as additional mechanism through interparticle magnetic particle interaction. In this research, MREs based on natural rubber and waste nickel zinc ferrites were prepared. Individual components in MREs that contribute to the performance and energy absorption of the materials were investigated. MREs with different natural rubber matrix viscosities and industrial waste nickel zinc ferrite were prepared in order to study the effect of viscoelastic properties of rubber matrix on the dynamic and mechanical performance of the materials. The results revealed that the tan δ increased with increasing matrix viscosity over the whole range of frequency and strain amplitude explored. It was also found that the tensile strength and elongation at break increased with increasing matrix viscosity. The scanning electron microscope (SEM) micrographs revealed that the columnar structures became longer and thicker with a decrease in matrix viscosity. However, numerous cavities remained due to particle pull out, suggesting poor interaction between waste nickel zinc ferrite and rubber matrix. For assessing the effect of interfacial interaction between rubber matrix and magnetic particles on dynamic and mechanical performance of the MREs, Bis-(3-triethoxysilylpropyl) tetrasulphane (TESPT) was utilized to modify the surface of nickel zinc ferrite. The content of

TESPT was varied at 0, 2, 4, 6, and 8 wt% for the low and high viscosity rubber matrix. The improved interfacial bonding was evidenced by Fourier transform infrared spectroscopy (FTIR), grafting percentage, crosslink density and SEM images. The result revealed that the tan δ improved approximately 30% over the frequency and strain amplitude explored for both low and high viscosity rubber matrix. Stronger interfacial interaction also improved the tensile properties of the MREs and the optimum content of TESPT was found to be at 6 wt%. The effect of interparticle magnetic particle interaction on dynamic and mechanical performance of the MREs was investigated by curing the materials at 0, 100, 150, 165 and 200 mT magnetic field. It was found that the tan δ increased as the magnetic field increased and level off at magnetic saturation point of 165 mT. However, the tensile strength was found to decrease with increasing magnetic field due to the tensile load direction is perpendicular to the magnetic particles alignment.

CHAPTER 1

INTRODUCTION

1.1 Background of study

Magnetorheological elastomer (MRE) is a class of composite material that consists of magnetic particles suspended within an elastomer matrix. The magnetic particles used in MRE are carbonyl iron, iron oxides and other soft-magnetic particles without magnetic hysteresis, and suitable elastomer matrix materials include natural rubber (Yoon et al., 2013), silicone rubber (Shiga et al., 1995), nitrile rubber (Tian et al., 2013), polybutadiene rubber (Fuchs et al., 2007) and polyurethane rubber (Ju et al., 2016). MREs could be constructed as isotropic and anisotropic MREs (Zhou, 2003, Boczkowska et al., 2012). The isotropic MRE is characterized by its homogeneous dispersion of magnetic particles in a natural rubber matrix. The anisotropic MRE formed chain-like magnetic particle structures within a rubber matrix as a result of subjecting the material to an external magnetic field during curing (Farshad and Benine, 2004). Formation of such chain-like structures relies on the mechanism such that when individual particles are exposed to an applied magnetic field, magnetic dipole moments pointing along the field direction are induced within them. A magnetic force will cause the north pole of one particle to attract the south pole of its neighbour, resulting in the formation of chains and columnar structures inside the matrix (Shuib, 2015, Shuib and Pickering, 2016).

Eventually, when the matrix is cured, the particle structure set in place. Anisotropic MRE is found to produce materials with larger stiffness and better damping performance compared to isotropic MRE or conventional rubber composites (Ginder et al., 1999). Here damping is mainly promoted by energy absorption by friction between the molecule chains in the rubber matrix and damping provided by matrix-filler interface as with conventional rubber composites, but inclusion of magnetic particles in rubber enables additional damping through magnetism-induced damping (Kaleta et al., 2011). Jung et al. (2016) investigated the MR performance of isotropic and anisotropic MRE systems prepared using natural rubber (NR) and carbonyl iron (CI) particles and found that the anisotropic MRE possessed a larger storage modulus than the isotropic one, which was explained as due to the reason that the chain-like structure formed by aligned particles along the field direction acts as a rod-like filler. Similarly, Lu et al. (2012) reported that for MRE consisting of thermoplastic poly (styrene-b-ethylene-co-butylene-b-styrene) rubber and CI particles, the anisotropic MRE showed an even higher initial storage modulus because the filler effect resulting from the chain-like structure of the particles enhanced the magnetic permeability of the MRE.

MREs hold promise in a large variety of engineering fields for vibration control and vibration isolation systems, including the automotive industry (Cao and Deng, 2009), machinery (Xu et al., 2010) and earthquake resistance (Yang et al., 2016). The formation of columnar structures in anisotropic MREs has huge potential implication to practical applications. For instance, current seismic bearings are large and heavy which typically consists of rubber-reinforced metal plates installed for a mid-sized building. In order to apply the technology of seismic isolation for public housing and low cost buildings, columnar structures of anisotropic MREs provide micro reinforcement to the rubber matrix and offer huge potential to be applied as seismic bearing. Dyke et al. (1996) investigated the performance of a semiactive control system-based newly developed magnetorheological fluid (MRF) dampers. Khimi and Pickering, (2015) compared the performance of anisotropic MREs with conventional antivibration rubber for potential application in vibration damping. The results revealed the performance of MREs were comparable with conventional antivibration rubber.

1.2 Problem statement

The most commonly used rubber matrix for MREs is natural rubber (Yoon et al., 2013, Chung et al., 2015) and synthetic rubbers such as silicone rubber (Shiga et al., 1995, Sedlacik et al., 2016, Perales-Martínez et al., 2017), nitrile rubber (Tian et al., 2013), and polyurethane rubber (Chokkalingam et al., 2011, Ju et al., 2016). Natural rubber is preferred matrix, because it has good elasticity and damping properties. Furthermore, natural rubber does not corrode and is able to withstand abrasive substances such as salt water, acids, and corrosive liquids (Le Gac et al., 2015, Chandrasekaran, 2010). It can also bond well to metal parts and is relatively easy to process. The most magnetic particles for MREs are carbonyl iron and iron oxides (Jang et al., 2005). Carbonyl iron is frequently used due to their high magnetization (up to 2.1 Tesla), low residual magnetization, high magnetic permeability and soft magnetic characteristics. However, the price of carbonyl iron particles is too high, at $13─$20/kg in bulk (Goodman, 2019).

In order to reduce the cost of MREs, waste nickel zinc ferrite was selected in this study as the magnetic particles. Waste nickel zinc ferrite has a number of advantages including high magnetic permeability, high electrical resistivity, good chemical stability, and low cost (Mathew and Juang, 2007). It is a product of the excess raw material from ferrite industries, which manufacture electronic inductors, power transformer cores, antennas, and transponders. These industries generate 3─10 tonnes of waste nickel zinc ferrite per month (Hossen et al., 2015). The waste nickel zinc ferrite from manufacturing is usually abandoned although it contains 70% ferrite. The waste also cannot be recycled because of its complex compositions (Ismail et al., 2007). Inorganic impurities might also be present on the surface of the nickel zinc ferrite which is generated during the cutting and machining process (Pereira et al., 1999). Furthermore, the waste nickel zinc ferrite contains heavy metal elements such as zinc and nickel, which could harm health of the human and the environment if not treated properly prior to disposal (Kmita et al., 2016). The introduction of waste nickel zinc ferrite used in MREs is one of the interesting perspectives that could lead to a more sustainable environment, recycling and profit earning.

The fabrication of MREs requires in-depth consideration, not only the selection of the magnetic particles and the type of rubber matrix, but also depends on matrix viscosity. The matrix viscosity of MREs is believed to significantly impact the abilities of the magnetic particles to orientate along the magnetic field direction during curing, which, in turn, affect the structural formation of columnar structure within anisotropic MREs (Oh et al., 2014). Therefore, it is sensible to optimize the matrix viscosity of MREs to improve the formation of columnar structures in MREs, which could possibly increase the dynamic mechanical performance of the materials through magnetic particle interactions.

Development of MREs based on industrial nickel zinc ferrite and natural rubber in this work sets a challenge as the inorganic magnetic fillers are inherently incompatible with organic rubber matrix which leads to poor adhesion and wettability between the rubber matrix and magnetic filler. Therefore, surface modification of nickel zinc ferrite magnetic fillers is an attractive approach in order to promote adhesion and enhance the dispersion of magnetic fillers within the rubber matrix to ensure that the production of final product having high performance.

Surface modification of inorganic particles nickel zinc ferrite was employed by bifunctional coupling agent treatment. This treatment uses silane based coupling agent, which is the most effective and low cost treatment. Silane based coupling agent are silicon-based chemicals which consist of hydrolysable groups (for example methoxy, acetoxy or ethoxy) at one end that react with inorganic materials and organofunctional groups (for example amino or sulphide) at the other end which can react with the rubber matrix. Thus, it is expected that the silane coupling agent can couple the inorganic and organic materials and improve compatibility and interfacial interaction of MREs.

Additional important feature that affect the damping performance of MRE is applied magnetic field during curing. Magnetism-induced damping in MREs possibly due to increased energy absorbed to defeat interparticle magnetic interaction between neighbouring particles. The highest possible increase in damping between magnetic particle interactions occurs when the aligned particles develop magnetically saturated. Chen et al. (2007) stated that for MREs based on natural rubber having 60 wt% carbonyl iron particles, saturation happened around 400 mT and Qiao et al. (2012) also mentioned for MREs based on thermoplastic elastomer matrix having modified carbonyl iron, the saturation happened at around 500 mT. The results also revealed that, more particles combined each other and the chain like columnar structures became thicker and longer as the magnetic field strength increased to provide much larger damping. However, none has assessed the magnetic saturation for nickel zinc ferrite embedded in natural rubber matrix. Therefore, a substantial study on magnetism induced damping provided by waste nickel zinc ferrite particles in natural rubber based MREs is essential in order to understand the relative importance of this mechanisms in magnetorheological elastomers (MREs).