

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

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UNIVERSITI SAINS MALAYSIA

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ELASTOMER BASED ON NICKEL ZINC
FERRITE AND NATURAL RUBBER**

by

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TABLE OF CONTENTS

	Page
ACKNOWLEDGEMENT	ii
TABLE OF CONTENTS	iii
LIST OF TABLES	viii
LIST OF FIGURES	ix
LIST OF SYMBOLS	xiii
LIST OF ABBREVIATIONS	xiv
ABSTRAK	xv
ABSTRACT	xvii
CHAPTER 1 INTRODUCTION	1
1.1 Background of study	1
1.2 Problem statement	3
1.3 Research objectives	6
1.4 Outline of the thesis.....	6
1.5 Scope of study	7
CHAPTER 2 LITERATURE REVIEW	8
2.1 Magnetorheological (MR) materials	8
2.1.1 Magnetorheological fluids (MRFs)	8
2.1.2 Magnetorheological elastomers (MREs)	10
2.2 Damping mechanisms in magnetorheological elastomers	11
2.2.1 Viscoelastic damping.....	11
2.2.2 Interfacial damping.....	12
2.2.3 Magnetism-induced damping	13
2.3 Magnetorheological elastomer applications.....	14
2.4 Magnetorheological elastomer components.....	16
2.4.1 Matrix material	16

2.4.1(a)	Natural rubber.....	18
2.4.1(b)	Rubber compounding ingredients.....	22
2.4.2	Magnetic particles.....	23
2.5	Fabrication process of magnetorheological elastomers.....	28
2.5.1	Mixing	28
2.5.1	Vulcanization and measurement of cure characteristics.....	29
2.5.3	Shaping and curing for magnetorheological elastomers (MREs).....	32
2.6	Factors affecting magnetorheological elastomer performance	36
2.6.1	Viscosity of rubber matrix	36
2.6.2	Surface modification of magnetic particles	37
2.6.3	Magnetic field strength.....	40
2.6.4	Other factors	40
CHAPTER 3 METHODOLOGY		42
3.1	Introduction	42
3.2	Materials.....	43
3.2.1	Rubber matrix	44
3.2.2	Zinc oxide and stearic acid	44
3.2.3	Nickel zinc ferrite	44
3.2.4	Paraffin oil	45
3.2.5	N-cyclohexyl-2-benzothiazolsulfenamide (CBS).....	45
3.2.6	Tetra-methylthiuram disulphide (TMTD)	45
3.2.7	N-isopropyl-n'-phenyl-p-phenylenediamine (IPPD)	46
3.2.8	Bis [3-(triethoxysilyl) propyl] tetrasulfide (TESPT).....	46
3.2.9	Sulphur.....	46
3.2.10	Toluene	46
3.3	Equipment	47
3.4	Formulation and preparation of magnetorheological elastomer (MREs).....	48

3.4.1	Preparation of nickel zinc ferrite	48
3.4.2	Preparation of MREs with different matrix viscosity.....	48
3.4.3	Preparation of MREs with different content of TESPT and matrix viscosity	49
3.4.4	Surface treatment of nickel zinc ferrite	50
3.4.5	Preparation of MREs with different magnetic field	51
3.4.6	Rubber compounding	51
3.4.7	Measurement of cure characteristics	53
3.4.8	Vulcanization process.....	53
3.5	Materials characterization	55
3.5.1	Particle size.....	55
3.5.2	Magnetic properties	55
3.5.3	Fourier Transform Infrared Spectroscopy	56
3.5.4	Thermal gravimetric analysis (TGA)	56
3.5.5	Swelling and crosslink density	57
3.5.6	Tensile test.....	58
3.5.7	Dynamic mechanical properties	58
3.5.8	Morphology test.....	59
CHAPTER 4 RESULTS ANALYSIS AND DISCUSSION.....		60
4.1	Characterization of waste nickel zinc ferrite.....	60
4.2	Matrix viscosity of magnetorheological elastomers.....	62
4.3	Influence of matrix viscosity on the dynamic mechanical performance of magnetorheological elastomers	63
4.3.1	Scanning electron microscope (SEM)	63
4.3.2	Curing characteristic.....	66
4.3.3	Dynamic mechanical properties	70
4.3.3(a)	Frequency sweep measurement.....	70
4.3.3(b)	Strain amplitude measurement	73

4.3.4	Tensile properties	76
4.3.4(a)	Tensile strength.....	76
4.3.4(b)	Elongation at break.....	77
4.3.4(c)	Modulus at 100% elongation (M100).....	78
4.3.5	Glass transition temperature (T_g).....	79
4.3.6	Thermogravimetric analysis (TGA)	80
4.4	Effect of silane coupling agent on dynamic mechanical performance of plasticized anisotropic magnetorheological elastomers	82
4.4.1	Characterization of surface modified nickel zinc ferrite particles....	82
4.4.2	Curing characterization of MREs	89
4.4.3	Morphology	91
4.4.4	Dynamic mechanical properties	93
4.4.4(a)	Frequency sweep measurement.....	93
4.4.4(b)	Strain amplitude measurement	96
4.4.5	Tensile properties	99
4.4.5(a)	Tensile strength.....	99
4.4.5(b)	Elongation at break.....	100
4.4.5(c)	Modulus at 100% elongation (M100).....	101
4.5	Effect of magnetic field on dynamic mechanical properties of magnetorheological elastomers	103
4.5.1	Morphology	103
4.5.2	Dynamic mechanical properties	105
4.5.2(a)	Frequency sweep measurement.....	105
4.5.2(b)	Strain amplitude measurement	108
4.5.3	Glass Transition Temperature (T_g).....	111
4.5.4	Tensile properties	112
4.5.4(a)	Tensile strength.....	112
4.5.4(b)	Elongation at break.....	114

4.5.4(c)	Modulus at 100% elongation (M100).....	115
CHAPTER 5 CONCLUSION AND FUTURE RECOMMENDATIONS		116
5.1	Conclusion.....	116
5.1.1	To characterize the properties of waste nickel zinc ferrite	116
5.1.2	To assess the effect of matrix viscosity on the formation of magnetic particle alignment in magnetorheological elastomers (MREs)	116
5.1.3	To evaluate the role of interfacial damping through weakly bonded interfaces and strongly bonded interfaces between nickel zinc ferrite particle and natural rubber matrix	117
5.1.4	To investigate the effect of magnetic field on magnetic particle interaction and dynamic mechanical performance of MREs.....	117
5.2	Recommendations for future works	119
REFERENCES.....		120
LIST OF PUBLICATIONS		

LIST OF TABLES

	Page
Table 2.1	List of matrix types commonly used in MREs..... 17
Table 2.2	Physical properties of common types of rubber 18
Table 2.3	Chemical composition of fresh latex 20
Table 2.4	Specifications for natural rubber grades 21
Table 2.5	Common additives used in rubber compounding..... 22
Table 2.6	List of particle types commonly used in MREs 26
Table 2.7	Methods of common surface modification used 38
Table 3.1	List of materials with their manufacturers and commercial names ... 43
Table 3.2	Properties of SMR L 44
Table 3.3	List of equipment 47
Table 3.4	Formulation of MREs with different matrix viscosity 48
Table 3.5	Formulation of MREs with different content of TESPT and matrix viscosity 50
Table 3.6	Formulation of MREs with different magnetic field..... 51
Table 3.7	The mixing sequence of rubber compounding..... 52
Table 4.1	Elemental composition of nickel zinc ferrite particles..... 61
Table 4.2	VSM data of nickel zinc ferrite particle..... 61
Table 4.3	Viscosity of Anisotropic MREs with different plasticizer contents... 63

LIST OF FIGURES

	Page
Figure 2.1	MRF structure; (a) in the absence of magnetic field and (b) under influence of magnetic field9
Figure 2.2	Cyclic stress-strain viscoelastic curve split up into viscous and elastic components 12
Figure 2.3	Constrained and free rubber in an agglomerate 13
Figure 2.4	Prototype of (a) tuned vibration absorption system using MRE, (b) MRE isolator with variable stiffness and damping, (c) MRE-based force sensor and (d) tuneable spring element..... 16
Figure 2.5	Structure of linear poly(cis-1,4-isoprene) 19
Figure 2.6	Mapping of MREs based on magnetizable particle; (a) based on particle size, (b) based on particle shape.....24
Figure 2.7	Waste nickel zinc ferrite particles27
Figure 2.8	MREs fabrication process28
Figure 2.9	Two roll mill29
Figure 2.10	Sulphur vulcanisation of cis-polyisoprene31
Figure 2.11	Schematic diagram of compression moulding34
Figure 2.12	Magnet mould for MRE fabrication.....34
Figure 2.13	Electromagnetic-heat coupled device for MRE fabrication.....35
Figure 3.1	The flow chart of research methodology43
Figure 3.2	Molecular structure of TESPT46
Figure 3.3	Permanent magnet magnetic mould for anisotropic curing54
Figure 3.4	Electronic magnetometer55
Figure 4.1	SEM micrograph of nickel zinc ferrite particles60
Figure 4.2	Particle size distribution of nickel zinc ferrite61

Figure 4.3	Storage modulus versus strain to define the LVE region.....	62
Figure 4.4	Loss modulus versus strain to define the LVE region	62
Figure 4.5	SEM images of the MREs at different matrix viscosity, a) MV1, b) MV2, c) MV3, d) MV4, e) MV5 and f) MV6.....	65
Figure 4.6	Thickness and length of waste nickel zinc ferrite chains at different matrix viscosity	66
Figure 4.7	(a) scorch and cure time at different matrix viscosity (b) Torque different at different matrix viscosity.....	68
Figure 4.8	Crosslink density of anisotropic MREs at different matrix viscosity	69
Figure 4.9	Shear stress of anisotropic MREs at different matrix viscosity	69
Figure 4.10	Tan δ (a) storage modulus (b) and loss modulus (c) of MREs with different matrix viscosity against frequency.	73
Figure 4.11	Tan δ (a) storage modulus (b) and loss modulus (c) of MREs with different matrix viscosity against strain amplitude.	75
Figure 4.12	Tensile strength of MREs at different matrix viscosity	77
Figure 4.13	Elongation at break of MREs at different matrix viscosity.....	78
Figure 4.14	Modulus (M100) at different matrix viscosity.....	79
Figure 4.15	Tan δ at different matrix viscosity	80
Figure 4.16	TGA curves of MREs with different matrix viscosity in the nitrogen	81
Figure 4.17	Schematic illustration of the reactions of TESPT with waste nickel zinc ferrite particles.....	83
Figure 4.18	FTIR spectra of waste nickel zinc ferrite particles at different TESPT contents.....	84
Figure 4.19	TGA curves for waste nickel zinc ferrite particles at different TESPT contents.....	85
Figure 4.20	Silane grafting percentage of waste nickel zinc ferrite at different TESPT contents.....	86

Figure 4.21	Schematic diagram of formation of polymeric siloxane oligomer.....	86
Figure 4.22	Schematic illustration of the reaction mechanisms of tetrasulphane group of TESPT with the natural rubber.....	88
Figure 4.23	Crosslink density of MREs with different TESPT.....	89
Figure 4.24	Scorch and cure time for MRE/LV and MRE/HV with different contents of TESPT	90
Figure 4.25	Torque different for MRE/LV and MRE/HV at different TESPT content.....	91
Figure 4.26	SEM images of fracture of MREs with (a) unmodified and (b) modified waste nickel zinc ferrite (treated with 6 phr of TESPT).....	92
Figure 4.27	SEM images of fracture (a) MRE/LV and (b) MRE/HV.....	93
Figure 4.28	(a) Tan δ of MRE/HV (b) Tan δ of MRE/LV (c) Storage modulus (G') of MRE/HV (d) Storage modulus (G') of MRE/LV (e) Loss modulus (G'') of MRE/HV (f) Loss modulus (G'') of MRE/LV vs. frequency with different TESPT contents.....	95
Figure 4.29	(a) Tan δ of MRE/HV (b) Tan δ of MRE/LV (c) Storage modulus (G') of MRE/HV (d) Storage modulus (G') of MRE/LV (e) Loss modulus (G'') of MRE/HV (f) Loss modulus (G'') of MRE/LV vs. strain amplitude with different TESPT contents.....	98
Figure 4.30	Tensile strength of MRE/LV and MRE/HV at different TESPT content.....	100
Figure 4.31	Elongation at break of MRE/HV and MRE/LV at different TESPT content.....	101
Figure 4.32	Modulus M100 of MRE/HV and MRE/LV at different TESPT content.....	102
Figure 4.33	SEM images of fractured surface at different magnetic field; (a) 0 mT, (b) 100 mT, (c) 150 mT, (d) 165 mT and (e) 200 mT.....	104
Figure 4.34	Swelling percentage of MREs at different magnetic field.....	105
Figure 4.35	(a) Tan δ (b) Storage modulus (c) Loss modulus versus frequency for MREs at different magnetic field	108

Figure 4.36	(a) Tan δ (b) Storage modulus (c) Loss modulus versus frequency for MREs at different magnetic field	110
Figure 4.37	Tan δ of MREs at different magnetic field	111
Figure 4.38	Tensile strength of MREs at different magnetic field.....	113
Figure 4.39	Comparison of loading modes on particle separation, (a) tensile mode, (b) shear mode.....	113
Figure 4.40	Elongation at break of MREs at different magnetic field	114
Figure 4.41	Modulus (M100) of MREs at different magnetic field	115

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-benzothiazolsulfenamide
DMA	Dynamic Mechanical Analysis
DOP	Dioctyl Phthalate
DPG	Diphenyl guanidine
EP	Epoxy resin
FESEM	Fourier emission scanning electron microscope
FTIR	Fourier Transform Infrared Spectroscopy
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, $\tan \delta$ 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 $\tan \delta$ 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 $\tan \delta$ 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.

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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 $\tan \delta$ 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 $\tan \delta$ 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 $\tan \delta$ 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).