

MECHANICAL AND RHEOLOGICAL PROPERTIES OF ETHYLENE  
PROPYLENE DIENE MONOMER BASED MAGNETORHEOLOGICAL  
ELASTOMERS WITH SILICA NANOPARTICLES

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## **DEDICATION**

This thesis is dedicated to my beloved husband, children, parents, parents in law and family for supporting me all the way.

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## ABSTRACT

Magnetorheological elastomer (MRE) is an emerged smart material in which its responsive moduli in term of mechanical and rheological properties are influenced by the presence of an external magnetic field. However, the low mechanical properties of existing MREs have limited its use in some engineering applications, and temperature is one of the most influential factors affecting the performance of elastomer matrix in MRE which deteriorates the required properties of MREs. Previous studies have utilized silica as a reinforcing filler around polymer composite. Furthermore, silica is also being used as one of coating material to the magnetic particle in MRE to improve the mechanical properties and thermal stability of the base material. However, the use of silica as an additive in the thermal stability of MRE has not been explored. Thus, in this study, the effect of different content of silica on the mechanical and rheological properties of ethylene propylene diene monomer (EPDM)-based MREs under various operating temperatures is investigated by using 30 wt.% carbonyl iron particles (CIPs). The microstructure analysis was examined by using field-emission scanning electron microscopy, while the thermal characterizations were studied by using a thermogravimetric analyser and differential scanning calorimetry. The tensile properties were conducted by using Instron Universal Testing Machine in the absence of magnetic field at various temperatures. Meanwhile, the rheological properties were analysed under oscillatory loadings in the influence of magnetic field, using a rotational rheometer at 25 to 65° C. The experimental results revealed that the temperature diminished the molecular chains of elastomer matrix and caused the interfacial defects between filler and matrix, thus affecting the properties of MRE, in which the tensile strength and MR effect decreased with increasing temperature. However, the presence of silica has improved the thermal stability of MRE, thus reducing the interfacial defects when under the influence of temperature. The distribution of silica within the EPDM matrix and the adhesiveness of silica into the CIPs surface that occupied the gaps between distributed CIPs within the matrix enhanced the interfacial interactions between filler and matrix. Consequently, the addition of 11 wt.% silica improved the tensile strength by 344% and maintained the MR effect compared to MRE without silica at room temperature condition. The similar trends were also observed when MRE under the influence of temperatures; the MRE containing silica had higher tensile strength compared to MRE without silica, while the presence of silica maintained the MR effect under various operating temperatures. The incorporation of silica nanoparticles as an additive in EPDM-based MRE has the potential to sustain the properties of MRE devices in various temperature conditions. Thus, the study on the temperature-dependent mechanical and rheological properties of MRE is necessary, particularly regarding its practical applications.

## ABSTRAK

Elastomer reologi magnet (MRE) ialah bahan pintar yang muncul, di mana modulus responsifnya dari segi sifat mekanikal dan reologi dipengaruhi oleh kehadiran medan magnet luaran. Walau bagaimanapun, sifat mekanikal yang rendah bagi MRE sedia ada telah mengehadkan penggunaannya dalam beberapa aplikasi kejuruteraan, dan suhu adalah salah satu faktor paling berpengaruh yang mempengaruhi prestasi matrik elastomer dalam MRE yang merosotkan sifat MRE yang diperlukan. Kajian terdahulu telah menggunakan silika sebagai pengisi pengukuhan dalam bidang komposit polimer. Tambahan pula, silika juga digunakan sebagai salah satu bahan salutan kepada zarah magnet dalam MRE untuk meningkatkan kestabilan haba pada MRE. Walau bagaimanapun, penggunaan silika sebagai bahan tambahan dalam kestabilan haba dalam MRE belum diterokai. Oleh itu, dalam kajian ini, kesan kandungan silika yang berbeza terhadap sifat mekanikal dan reologi MRE berasaskan etilena propilena diena monomer (EPDM) di bawah pelbagai suhu operasi disiasat dengan menggunakan 30 wt.% zarah besi karbonil (CIPs). Analisis mikrostruktur diperiksa dengan menggunakan Mikroskop Elektron Pengimbasan Pelepasan Medan (FESEM), manakala pencirian haba dikaji dengan menggunakan Penganalisis Termogravimetrik dan Kalorimetri Pengimbasan Pembezaan. Sifat tegangan dijalankan dengan menggunakan Mesin Pengujian Sejagat Instron tanpa medan magnet pada pelbagai suhu. Sementara itu, sifat reologi dianalisis di bawah beban berayun dalam pengaruh medan magnet, menggunakan rheometer putaran pada 25 hingga 65° C. Keputusan eksperimen mendapati bahawa haba telah mengurangkan rangkaian molekul matriks elastomer dan menyebabkan kerosakan antara muka di antara pengisi dan matriks, sekali gus menjejaskan sifat MRE, di mana kekuatan tegangan dan kesan MR berkurangan dengan peningkatan haba. Walau bagaimanapun, kehadiran silika mampu meningkatkan kestabilan haba MRE dengan meningkatkan interaksi di antara pengisi dan matriks, sekali gus mengurangkan kerosakan antara muka apabila di bawah pengaruh haba. Taburan silika dalam matriks EPDM dan kelekatan silika pada permukaan CIP yang memenuhi ruang antara CIP yang bertaburan di dalam MRE, telah meningkatkan interaksi antara muka di antara CIP dan matriks. Akibatnya, penambahan 11 wt.% silika telah meningkatkan kekuatan tegangan sebanyak 344% dan mengekalkan kesan MR berbanding MRE tanpa silika pada keadaan suhu bilik. Trend yang sama juga diperhatikan apabila MRE di bawah pengaruh suhu; MRE yang mengandungi silika mempunyai kekuatan tegangan yang lebih tinggi berbanding MRE tanpa silika, manakala kehadiran silika mengekalkan kesan MR di bawah pelbagai suhu operasi. Penggabungan nanozarah silika sebagai bahan tambahan dalam MRE berasaskan EPDM berpotensi untuk mengekalkan sifat peranti MRE dalam pelbagai keadaan suhu. Oleh itu, kajian tentang sifat mekanikal dan reologi MRE yang bergantung kepada suhu adalah keperluan, terutamanya berkenaan dengan aplikasi praktikalnya.

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## LIST OF ABBREVIATIONS

BIPs	-	Bare iron particles
BR	-	Polybutadiene
CB	-	Carbon black
CIPs	-	Carbonyl iron particles
CNT	-	Carbon nanotube
CPVC	-	Critical particle volume concentration
DSC	-	Differential Scanning Calorimetry
DTG	-	Differential Thermogravimetric
EDX	-	Energy Dispersive X-ray Spectroscopy
ENR	-	Epoxidized natural rubber
EPDM	-	Ethylene Propylene Diene Monomer
FESEM	-	Field Emission Scanning Electron Microscopy
Gr	-	Graphite
LVE	-	Linear viscoelastic region
MBT	-	2-Mercaptobenzothiazole
MR	-	Magnetorheological
MRE	-	Magnetorheological elastomer
MRF	-	Magnetorheological fluid
MRG	-	Magnetorheological grease
MRP	-	Magnetorheological plastomer
MWCNT	-	Multiwall carbon nanotube
NLVE	-	Nonlinear viscoelastic region
NR	-	Natural rubber
PDMS	-	Polydimethylsiloxanes
PE	-	Polyethylene
PEN	-	Poly (ethylene 2.6-naphthalate)
PTFE	-	Polytetrafluoroethylene
Phr	-	Parts per hundreds of rubbers
PU	-	Polyurethane
SEBS	-	Styrene ethylene butylene styrene

SEM	-	Scanning Electron Microscopy
SiO <sub>2</sub>	-	Silicon dioxide
SR	-	Silicon rubber
TESPT	-	Bis-(3-triethoxysilylpropyl) tetrasulphane
TGA	-	Thermogravimetric Analyzer
TMTD		Tetramethyl thiuram disulphide
VSM	-	Vibrating Sample Magnetometer
ZnO	-	Zinc Oxide

## LIST OF SYMBOLS

$M_s$	-	Magnetic saturation
$M_r$	-	Retentivity
$H_c$	-	Coercivity
$\mu$	-	Magnetic permeability
$B$	-	Flux density
$G'$	-	Storage modulus
$G''$	-	Loss modulus
$\tan \delta$	-	Loss factor
$T_g$	-	Glass transition temperature
$T_m$	-	Melting temperature
$\Delta H$	-	Enthalpy of fusion
$T_p$	-	Peak decomposition temperature



# CHAPTER 1

## INTRODUCTION

### 1.1 Research Background

Development of controllable stimuli-responsive technology in engineering applications has invented various kinds of smart materials in recent years. Magnetorheological (MR) materials are among smart materials that have propitious future in terms of fast real-time response since their rheological properties can be continuously, rapidly and reversibly altered by an external magnetic field [1–4]. MR elastomers (MREs) are recognized as a solid-state analogue of MR fluid (MRF) and a new branch of MR materials, which composed of magnetic particles embedded in a non-magnetic elastomeric matrix. The responsive interactions between magnetic particles when subjected to external magnetic fields has resulted in field-dependent material property of MRE. Having unique properties such as controllable stiffness and fast response time as well as its capability to overcome the limitations in MRF, MRE has attracted growing interest in many engineering applications such as adaptive tuned vibration absorbers [5,6], stiffness tuneable mounts [7], vehicle suspension bushing [8], and tuneable spring components [9].

Realizing the importance and potential of MRE in real applications, many studies have been conducted by manipulating the parameters in MRE fabrication especially in material selections and compositions in order to enhance the performance of MREs. In terms of matrix materials selection, many variations have been used consisting of silicone rubber (SR) [10–13], natural rubber (NR) [14–17], polyurethane (PU) [18–20], polybutadiene [21,22], polyisobutylene [23], and ethylene-propylene-diene monomer (EPDM) [24–26]. It is well known that rubber is a non-magnetic material, where the modulus is constant under any applied magnetic fields, however, with the presence of magnetic particles in MRE, the modulus can be changed tremendously according to the intensity of magnetic fields. Thus, in order to obtain the

largest changes in stiffness or known as MR effect, many researchers have employed the soft rubber matrix in MRE fabrication due to its flexible macromolecular chains [25]. Nevertheless, the requirement of high tensile strength and good MR effect in some industries have limited the application of soft rubber based MREs due to its low stiffness and strength [15,27]. In the meantime, NR-based MREs have been effectively implemented in industry especially in automotive applications due to its excellent mechanical properties. However, due to weak ageing behavior of NR [24], the presence of magnetic particles in NR-based MRE will accelerate the oxidative ageing, thus weaken the properties as well as shorten its usage. Therefore, the development of other alternative elastomeric matrices that has good thermal and aging resistance, and at the same time possess good tensile and fatigue strength will be the ideal approach for MRE applications in automotive industry. Among those listed matrices, EPDM is getting more attention recently in MRE's fabrication due to high strength, good resistance to temperature, ageing, oxidation and atmosphere, easier processing steps and low production cost compared to NR [24,25].

Aside of matrix materials selection, composition of magnetic particles in MRE fabrication is another crucial factor to determine the performance of MRE. Carbonyl iron particles (CIPs) are mostly considered as main magnetic particles in MRE fabrication compared to magnetite [28–30], nickel [31,32], iron sand [33,34] and cobalt [32] due to high permeability, high magnetic saturation and low remnant magnetization, which can contribute to actively tuned and fast respond towards the magnetic fields [2,35]. Previous studies have reported that the ideal concentration of magnetic particles in MRE fabrication was about 30 vol% (~70 wt.%) as the higher the particles content, the higher the storage modulus and MR effect [13,15,36]. Though, too high amount of particles would lead to aggregation of particles, and degradation of mechanical properties and durability [15,26,27,28]. In addition, high damping ratio and very high stiffness caused poor vibration suppression, which was not applicable for absorber and suspension applications [24,37]. Thus, some researchers have manipulated the composition of magnetic particles as low as 10 to 20 vol% (40 to 60 wt.%) to enhance the mechanical properties of MRE. Based on their findings, the optimum mechanical properties of MRE could be achieved by reducing the magnetic particles concentrations as low as 20 wt.% [10,11].

The interfacial interaction between filler and matrix is crucial in an MRE as the difference in surface characteristics between elastomer matrix and magnetic particles may weaken the required properties of MREs. In other words, the dispersion of hydrophilic magnetic particles inside MRE will get worse due to nonpolar surface properties of matrix, thus resulted in inhomogeneous dispersion of magnetic particles [6,38]. It will consequently deteriorate the mechanical properties and increase the damping properties of MRE [38–40]. Moreover, the weak filler-matrix interactions will lead to the loss of magnetic interactions at the gaps between filler and matrix, diminishing the field-dependent properties of MRE [40]. In the meantime, the modulus and strength of elastomer matrix itself are low and tend to degrade when expose to various operating temperatures. The weakened polymer molecular chains caused by the temperature will initiate the interfacial defects between filler and matrix, resulting in the decrement of the MRE's properties. Therefore, to overcome the weak interactions between filler and matrix, few studies on the surface modification of magnetic particles have been conducted to improve the filler-matrix interfacial interactions and thermal stability of the MRE [6,25,41]. On the other hand, additive utilization in MRE is another alternative that has been proposed by many researchers [14,15,17,27,33,38,42,43], mainly to strengthen the interfacial interaction between particles and matrix, to enhance the dispersion of magnetic particles as well as to improve the properties of MRE.

There are many types of additives that have been applied into MREs including plasticizers [15,20,27,44], silane coupling agents [33,34] and nanoparticles [14,17,42,45,46]. Additives have been used to enhance the properties of MRE by altering the mechanism of matrix and/or by modifying the surface properties of magnetic particles. The inclusion of nano-sized powdered additives in MRE are remarkably enhanced the mechanical properties of MRE as it can improve the bonding between filler and matrix. The nano-sized additives are adsorbed on the surface of magnetic particles, thus improves the filler-matrix interactions by occupying the gaps at the interface. Until now, the nano-sized additives such as carbon black [14,17,24,37], graphite [47,48] and carbon nanotubes [38,42] have been studied in MRE fabrication to enhance the mechanical and rheological properties in dynamic automotive applications such as vibration absorbers and suspension bushings. Meanwhile, numerous studies on the polymer composite have demonstrated that the

reinforcement of silica nanoparticles could improve the mechanical and thermal stability of the polymer material [49–54]. Therefore, the additives manipulation using silica nanoparticles should be considered to cater the issue related to interfacial interactions between its raw materials and low thermal stability of MRE under various operating temperatures.

## 1.2 Motivation of Research

Previous studies have utilized nanoparticles additive such as carbon black, carbon nanotube and graphite to improve the mechanical and rheological properties of MRE by strengthening the interfacial interactions between filler and matrix. However, the demand on high stiffness and tensile strength with good thermal stability of MRE has attracted attention from MRE potential applications and has become an emerging research topic [1,3,4,35]. MREs normally operated in the multiple ranges of frequency, strain amplitude, magnetic field and temperature, where the temperature is considered as one of the most influential factors affecting the performance of elastomer matrix in MRE [55–57]. The temperature changes in MRE may cause by the environmental temperature variations and/or internal temperature rise during energy dissipation [25,57,58], which can tremendously affect the MRE performance. Few studies on the temperature-dependent rheological properties of MRE have reported that the initial storage modulus and MR effect decreased with increasing temperature owing to the relative movement between polymer molecular chains and rotation of the particle chains inside the MRE [22,25,55–60]. In addition, the most critical factor that affecting the stiffness of MRE under temperature intervention is the interaction between the magnetic particles and elastomer matrix, showing that the temperature would significantly affect the properties of elastomer matrix in MRE [59]. Thus, it is crucial to investigate the effect of temperature on the mechanical and rheological properties of MRE in order to meet the requirements of their practical applications.

Realizing the significant effect of temperature on the performance of MRE, Qi *et al.* [25] has utilized a good thermal resistance elastomer matrix like EPDM rubber as a polymer blend with SR-based MRE to enhance the tensile strength and thermal

stability of MRE. The results revealed that the incorporation of EPDM has improved the tensile strength with excellent temperature stability, while maintaining high MR effect. Even though the incorporation of EPDM as polymer blend has significantly improved the mechanical, rheological and thermal stability of SR-based MRE, but somehow the interfacial interactions between its raw materials is still a concern that to be studied. Due to that limitation, the surface of CIP has been modified with silica coating to improve the interfacial interaction and dispersion of CIP in elastomer matrices, while the silane coupling agent was added to improve the compatibility between its raw materials. In fact, the precipitated silica has been used as one of the ingredients, but the effect of precipitated silica as an additive on the properties of MRE was not fully investigated. Hence, the usage of silica as an additive in MRE fabrication should be studied as an alternative to enhance the interfacial interactions between filler and matrix, so that the prior properties will be improved without additional surface treatment on magnetic particles.

Meanwhile, the silica has been recognized as one of the effective coating materials to magnetic particles in order to improve the interfacial interactions between magnetic particles and elastomer matrix in MRE fabrication. According to Malecki *et al.* [41], the coated CIPs exhibited a higher mechanical, thermal stability and MR effect than pure CIPs in the styrene ethylene butylene styrene (SEBS)-based MRE. The strong affinity between CIPs surface and silica has led to a good adhesion between CIPs and matrix, which consequently increased the mechanical properties and thermal stability of MRE as also reported elsewhere [6,25]. In the meantime, the used of silica as the reinforcing filler has been recognized by referring to the growing number of publications in rubber processing [49–54,61–64]. Generally, when using silica with elastomers, the resultant material would have unique characteristics such as improved hardness, mechanical strength, and thermal stability along with the process ability and the interfacial interaction between filler and matrix itself. Mokhothu *et al.* [50] has reported that the addition of 30 wt.% of treated nano-silica had improved the thermal stability by 10 °C and 23% in tensile strength. Based on the findings, it could be concluded that the utilization of silica as coating material and reinforcing filler managed to enhance the mechanical and thermal stability of the base materials where it could be an indicator to study the temperature dependent properties of MRE. Moreover, the characteristics of nano-sized particles were believed could

simultaneously improve the interactions between magnetic particles and elastomer matrix even under the influence of temperature. Therefore, a comprehensive study on the effect of silica nanoparticles as an additive in EPDM-based MRE would be worthwhile in providing very useful guidelines on the proper selection of MRE with good tensile strength and thermal stability while maintaining appropriate MR effect.

### **1.3 Problem Statement**

Many studies have utilized various kinds of nano-sized additives to improve the interfacial interactions between filler and matrix, resulting in the properties enhancement of MRE. However, the findings on the enhancement in mechanical and rheological properties of MRE under various operating temperatures are rather limited. Generally, temperature is considered as one of the most influential factors that affects the modulus of elastomer matrix in MRE, thus decreases the properties of MRE. Due to that concern, the effect of temperature on the properties of MRE should be considered for further studies as the MRE devices commonly operate in multiple temperature ranges, which require MRE materials to undergo repetitious loadings under various working environment [55–58]. Meanwhile, the utilization of silica as reinforcing filler in polymer composite, in general, has demonstrated an improvement in mechanical, as well as thermal stability of polymer composite. Despite that, silica has also been implemented as a coating material in MRE to improve the interfacial interactions and thermal stability of the MRE. However, the use of silica as an additive in thermal stability of MRE has not been fully discussed and needs to be explored more for further understanding of its influence on the temperature dependent properties of MRE. The addition of silica nanoparticles in MRE not only can increase the mechanical properties by improving the interfacial interactions between filler and matrix, but compared to other additives, it can also enhance the thermal stability of MRE [65]. Therefore, the introduction of a good thermal resistance material like silica as an additive in MRE will improve the thermal stability of MRE, thus increase the tensile strength and maintain the MR effect of MRE when under the influence of temperature.

## **1.4 Research Objectives**

The aim of the research is to enhance the tensile strength and maintain MR effect of EPDM-based MRE at various temperatures by using silica nanoparticles. The following are the specific objectives for this research:

- (a) To characterize the physicochemical properties of EPDM-based MRE in terms of morphological, magnetic, mechanical and thermal properties.
- (b) To analyse the rheological properties of the EPDM-based MRE for dynamic shear under absence and presence of various magnetic fields.
- (c) To evaluate the temperature-dependent on mechanical and rheological properties of EPDM-based MRE under various operating temperatures.

## **1.5 Research Scopes**

The scopes of this study are specified on the experimental investigation of silica as an additive in EPDM-based MRE as well as comprehensive characterization to evaluate the resultant properties to be utilized in potential practical applications. The scopes in this study include:

- (a) The fabrication of MRE consists of EPDM rubber with constant amount of 30 wt.% CIP and various silica contents (0, 3, 6, 9, and 11 wt.%).
- (b) The morphological, elemental, magnetic and thermal analysis of EPDM-based MRE were conducted using FESEM, EDX, VSM and TGA/DSC, respectively.
- (c) The rheological test was performed in the absence and presence of magnetic fields under oscillatory shear mode loadings. The experiments include input parameters of strain sweep and magnetic fields sweep to investigate the effect of storage modulus, loss factor and MR effect changes upon different contents of silica in EPDM-based MRE.

- (d) The studies regarding chemical reaction between EPDM and silica polar functional groups with magnetic particles or other additives are not included as the aim of this research focuses more on mechanical and field-dependent rheological properties of EPDM-based MRE under various operating temperatures.
- (e) The investigation on mechanical properties of EPDM-based MREs including only tensile strength and elongation at break, while the tensile modulus, tear strength and compression are not included in this study.

## **1.6 Significance of the Research**

This fundamental study is believed to contribute to the development of MRE material, which possess better tensile properties while maintaining the MR effect, particularly for applications under various operating temperatures. In reviewing the literature, many studies reported on the enhancement in mechanical and rheological properties of MRE by utilizing the nano-sized additives through various characterization methods and mathematical modelling. However, limited studies have focused on the employment of good thermal resistance nano-sized additive that could provide the enhancement in interfacial interaction between its raw materials and at the same time could improve the thermal stability of MRE under the influence of temperature. Thus, the utilization of silica nanoparticles is believed to cater on the major issues related to interfacial interactions between its raw materials and deterioration of MRE properties caused by various operating temperatures.

In addition, the present study provides important findings and gaining an attention to the importance of considering nano-sized additive instead of implementation of additional processing steps such as coating method to magnetic particles in order to improve the interfacial interaction between magnetic particles and elastomer matrix in MRE fabrication. This study also delivers additional evidence to the current literature and has gone some way towards enhancing understanding of raw materials selection and fabrication subjected to better MRE performance in MRE applications that often work in various operating temperatures. Therefore, it is



significant to utilize the silica nanoparticles in MRE fabrication as an additive to enhance the tensile strength while maintaining appropriate MR effect of MRE under various operating temperatures to meet the requirements of their practical applications.

## 1.7 Research Outlines

This research consists of five chapters including this introduction chapter. The relevant information, achievements and findings are highlighted in each chapter and end with a brief summary. The outline of the chapters is established as below:

**Chapter 1** introduces the idea of research by providing the general information related to the research background, motivation of the research, problem statement and significance of the research that clearly identifies the gap, objectives and scopes of research.

**Chapter 2** is the literature review related to previous research work of MR materials especially on the development of MREs. The literature review covers on MRE materials, additives, fabrication, characterization, and applications. Several scientific published and patented of potential applications are also summarized. The feasibility studies on EPDM and silica are explained in detail.

**Chapter 3** represents the methodology and experimental section. A brief explanation is given on the step-by-step procedures of the research to achieve the objectives. This chapter also describes the materials and detailed fabrication of the isotropic EPDM-based MREs. The experimental setup involved in the MRE characterizations and rheological examinations are also clearly explained, including facility details.

**Chapter 4** describes the results and discussions of physicochemical characterizations and rheological properties. Physicochemical characterizations include morphological observation, elemental determination, magnetic properties, mechanical properties and thermal properties analysis. Rheological properties as functions of several parameters

which include strain amplitude, magnetic field and temperature are explained based on dynamic loadings.

**Chapter 5** concludes and summarizes on the experimental findings that answered the objectives of this research. In addition, the contributions of this research towards real engineering applications also mentioned in this chapter. The future works for this research also proposed at the end of this chapter to further explore and enhance the existing issue arise in this research.

## REFERENCES

1. Morillas JR, de Vicente J. Magnetorheology: A Review. *Soft Matter*, 2020 **16**(42): 9614–9642.
2. Ubaidillah, Sutrisno J, Purwanto A, Mazlan SA. Recent Progress on Magnetorheological Solids: Materials, Fabrication, Testing, and Applications. *Advanced Engineering Materials*, 2015 **17**(5): 563–597.
3. Samal S, Škodová M, Abate L, Blanco I. Magneto-Rheological Elastomer Composites. A Review. *Applied Sciences*, 2020 **10**(14): 4899.
4. Hafeez MA, Usman M, Umer MA, Hanif A. Recent Progress in Isotropic Magnetorheological Elastomers and Their Properties: A Review. *Polymers*, 2020 **12**(12): 3023.
5. Lerner AA, Cunefare KA. Performance of MRE-Based Vibration Absorbers. *Journal of Intelligent Material Systems and Structures*, 2008 **19**(5): 551–563.
6. Chen D, Yu M, Zhu M, Qi S, Fu J. Carbonyl Iron Powder Surface Modification of Magnetorheological Elastomers for Vibration Absorbing Application. *Smart Materials and Structures*, 2016 **25**(11): 115005.
7. Song RM, Mazlan SA, Johari N, Imaduddin F, Aziz SAA, Fatah AYA, *et al.* Semi-Active Controllable Stiffness Engine Mount Utilizing Natural Rubber-Based Magnetorheological Elastomers. *Frontiers in Materials*, 2022 **9**(April): 1–12.
8. Li Y, Li J, Li W, Du H. A State-of-the-Art Review on Magnetorheological Elastomer Devices. *Smart Materials and Structures*, 2014 **23**(12): 123001.
9. Jung HS, Kwon SH, Choi HJ, Jung JH, Kim YG. Magnetic Carbonyl Iron/Natural Rubber Composite Elastomer and Its Magnetorheology. *Composite Structures*, 2016 **136**: 106–112.
10. Perales-Martínez IA, Palacios-Pineda LM, Lozano-Sánchez LM, Martínez-Romero O, Puente-Cordova JG, Elías-Zúñiga A. Enhancement of a Magnetorheological PDMS Elastomer with Carbonyl Iron Particles. *Polymer Testing*, 2017 **57**: 78–86.
11. Puente-Córdova J, Reyes-Melo M, Palacios-Pineda L, Martínez-Perales I, Martínez-Romero O, Elías-Zúñiga A. Fabrication and Characterization of

- Isotropic and Anisotropic Magnetorheological Elastomers, Based on Silicone Rubber and Carbonyl Iron Microparticles. *Polymers*, 2018 **10**(12): 1343.
12. Yu M, Ju B, Fu J, Liu X, Yang Q. Influence of Composition of Carbonyl Iron Particles on Dynamic Mechanical Properties of Magnetorheological Elastomers. *Journal of Magnetism and Magnetic Materials*, 2012 **324**(13): 2147–2152.
  13. Gong XL, Zhang XZ, Zhang PQ. Fabrication and Characterization of Isotropic Magnetorheological Elastomers. *Polymer Testing*, 2005 **24**(5): 669–676.
  14. Nayak B, Dwivedy SK, Murthy KSRK. Fabrication and Characterization of Magnetorheological Elastomer with Carbon Black. *Journal of Intelligent Material Systems and Structures*, 2015 **26**(7): 830–839.
  15. Chen L, Gong X long, Jiang W quan, Yao J jing, Deng H xia, Li W hua. Investigation on Magnetorheological Elastomers Based on Natural Rubber. *Journal of Materials Science*, 2007 **42**(14): 5483–5489.
  16. Wahab NAA, Mazlan SA, Ubaidillah, Kamaruddin S, Ismail NIN, Choi SB, *et al.* Fabrication and Investigation on Field-Dependent Properties of Natural Rubber Based Magneto-Rheological Elastomer Isolator. *Smart Materials and Structures*, 2016 **25**(10): 107002.
  17. Fan L, Wang G, Wang W, Lu H, Yang F, Rui X. Size Effect of Carbon Black on the Structure and Mechanical Properties of Magnetorheological Elastomers. *Journal of Materials Science*, 2019 **54**(2): 1326–1340.
  18. Hu Y, Wang YL, Gong XL, Gong XQ, Zhang XZ, Jiang WQ, *et al.* New Magnetorheological Elastomers Based on Polyurethane/Si-Rubber Hybrid. *Polymer Testing*, 2005 **24**(3): 324–329.
  19. Wu JJK, Gong XLX, Fan YCY, Xia H. Anisotropic Polyurethane Magnetorheological Elastomer Prepared through in Situ Polycondensation under a Magnetic Field. *Smart Materials and Structures*, 2010 **19**(10): 105007.
  20. Wu J, Xia XG, Hesheng YF. Improving the Magnetorheological Properties of Polyurethane Magnetorheological Elastomer through Plasticization. *Journal of Applied Polymer Science*, 2011 **123**(4): 2476–2484.
  21. Sun TL, Gong XL, Jiang WQ, Li JF, Xu ZB, Li WH. Study on the Damping Properties of Magnetorheological Elastomers Based on Cis-Polybutadiene Rubber. *Polymer Testing*, 2008 **27**(4): 520–526.
  22. Gong X, Fan Y, Xuan S, Xu Y, Peng C. Control of the Damping Properties of

- Magnetorheological Elastomers by Using Polycaprolactone as a Temperature-Controlling Component. *Industrial & Engineering Chemistry Research*, 2012 **51**(18): 6395–6403.
23. Wang Y, Hu Y, Wang Y, Deng H, Gong X, Zhang P, *et al.* Magnetorheological Elastomers Based on Isobutylene–Isoprene Rubber. *Polymer Engineering & Science*, 2006 **46**(3): 264–268.
  24. Burgaz E, Goksuzoglu M. Effects of Magnetic Particles and Carbon Black on Structure and Properties of Magnetorheological Elastomers. *Polymer Testing*, 2020 **81**: 106233.
  25. Qi S, Yu M, Fu J, Zhu M, Xie Y, Li W. An EPDM/MVQ Polymer Blend Based Magnetorheological Elastomer with Good Thermostability and Mechanical Performance. *Soft Matter*, 2018 **14**(42): 8521–8528.
  26. Plachy T, Kratina O, Sedlacik M. Porous Magnetic Materials Based on EPDM Rubber Filled with Carbonyl Iron Particles. *Composite Structures*, 2018 **192**: 126–130.
  27. Khairi MHA, Fatah AYA, Mazlan SA, Ubaidillah, Nordin NA, Ismail NIN, *et al.* Enhancement of Particle Alignment Using Silicone Oil Plasticizer and Its Effects on the Field-Dependent Properties of Magnetorheological Elastomers. *International Journal of Molecular Sciences*, 2019 **20**(17): 4085.
  28. Masłowski M, Zaborski M. Smart Materials Based on Magnetorheological Composites. *Materials Science Forum*, 2012 **714**: 167–173.
  29. Brezoi V. Magnetic Elastomers Based on Nanocrystalline Magnetite Particles. *The Scientific Bulletin of Valahia University – Materials and Mechanics*, 2014 **9**(9): 53–56.
  30. Ubaidillah, Imaduddin F, Li Y, Mazlan SA, Sutrisno J, Koga T, *et al.* A New Class of Magnetorheological Elastomers Based on Waste Tire Rubber and the Characterization of Their Properties. *Smart Materials and Structures*, 2016 **25**(11): 115002.
  31. Kchit N, Bossis G. Piezoresistivity of Magnetorheological Elastomers. *Journal of Physics: Condensed Matter*, 2008 **20**(20): 204136.
  32. Padalka O, Song HJ, Wereley NM, Filer JA, Bell RC. Stiffness and Damping in Fe, Co, and Ni Nanowire-Based Magnetorheological Elastomeric Composites. *IEEE Transactions on Magnetics*, 2010 **46**(6): 2275–2277.
  33. Pickering KL, Shuib RK, Ilanko S. The Effect of Silane Coupling Agent on Iron

- Sand for Use in Magnetorheological Elastomers Part 1: Surface Chemical Modification and Characterization. *Composites Part A: Applied Science and Manufacturing*, 2015 **68**: 377–386.
34. Shuib RK, Pickering KL. The Effect of Silane Coupling Agent on the Dynamic Mechanical Properties of Iron Sand/ Natural Rubber Magnetorheological Elastomers. *Composites Part B: Engineering*, 2016 **90**: 115–125.
  35. Kang S, Choi K, Nam JD, Choi H. Magnetorheological Elastomers: Fabrication, Characteristics, and Applications. *Materials*, 2020 **13**(20): 4597.
  36. Qiao X, Lu X, Gong X, Yang T, Sun K, Chen X. Effect of Carbonyl Iron Concentration and Processing Conditions on the Structure and Properties of the Thermoplastic Magnetorheological Elastomer Composites Based on Poly(Styrene-*b*-Ethylene-*Co*-Butylene-*b*-Styrene) (SEBS). *Polymer Testing*, 2015 **47**: 51–58.
  37. Chen L, Gong XL, Li WH. Effect of Carbon Black on the Mechanical Performances of Magnetorheological Elastomers. *Polymer Testing*, 2008 **27**(3): 340–345.
  38. Poojary UR, Hegde S, Gangadharan K V. Experimental Investigation on the Effect of Carbon Nanotube Additive on the Field-Induced Viscoelastic Properties of Magnetorheological Elastomer. *Journal of Materials Science*, 2018 **53**(6): 4229–4241.
  39. Yang J, Gong X, Deng H, Qin L, Xuan S. Investigation on the Mechanism of Damping Behavior of Magnetorheological Elastomers. *Smart Materials and Structures*, 2012 **21**(12): 125015.
  40. Wang YLY, Hu Y, Chen L, Gong XLX, Jiang WQW, Zhang PPQ, *et al.* Effects of Rubber/Magnetic Particle Interactions on the Performance of Magnetorheological Elastomers. *Polymer Testing*, 2006 **25**(2): 262–267.
  41. Małecki P, Królewicz M, Hiptmair F, Krzak J, Kaleta J, Major Z, *et al.* Influence of Carbonyl Iron Particle Coating with Silica on the Properties of Magnetorheological Elastomers. *Smart Materials and Structures*, 2016 **25**(10): 105030.
  42. Aziz SAA, Mazlan SA, Ismail NIN, Ubaidillah, Choi SB, Khairi MHA, *et al.* Effects of Multiwall Carbon Nanotubes on Viscoelastic Properties of Magnetorheological Elastomers. *Smart Materials and Structures*, 2016 **25**(7): 077001.

43. Yang J, Gong X, Zong L, Peng C, Xuan S. Silicon Carbide-strengthened Magnetorheological Elastomer: Preparation and Mechanical Property. *Polymer Engineering and Science*, 2013 **53**(12): 2615–2623.
44. Khairi MHA, Mazlan SA, Ubaidillah, Ahmad KZK, Aziz SAA, Yunus NA. Effect of Sucrose Acetate Isobutyrate Ester on the Epoxidised Natural Rubber Based Magnetorheological Elastomers. *Journal of Physics: Conference Series*, 2016 **776**(1): 012034.
45. Bica I, Anitas EM, Chirigiu L. Magnetic Field Intensity Effect on Plane Capacitors Based on Hybrid Magnetorheological Elastomers with Graphene Nanoparticles. *Journal of Industrial and Engineering Chemistry*, 2017 **56**: 407–412.
46. Fan L, Wang G, Wang W, Shi G, Yang F, Rui X. Investigations on the Properties of NH<sub>4</sub>HCO<sub>3</sub> Filled Natural Rubber Based Magnetorheological Elastomers (MREs). *Materials Research Express*, 2018 **5**(4): 045307.
47. Tian TF, Li WH, Alici G, Du H, Deng YM. Microstructure and Magnetorheology of Graphite-Based MR Elastomers. *Rheologica Acta*, 2011 **50**(9–10): 825–836.
48. Tian TF, Li WH, Deng YM. Sensing Capabilities of Graphite Based MR Elastomers. *Smart Materials and Structures*, 2011 **20**(2): 025022.
49. Morselli D, Bondioli F, Luyt AS, Mokhothu TH, Messori M. Preparation and Characterization of EPDM Rubber Modified with in Situ Generated Silica. *Journal of Applied Polymer Science*, 2013 **128**(4): 2525–2532.
50. Mokhothu TH, Luyt AS, Messori M. Reinforcement of EPDM Rubber with in Situ Generated Silica Particles in the Presence of a Coupling Agent via a Sol–Gel Route. *Polymer Testing*, 2014 **33**: 97–106.
51. Shin Y, Lee D, Lee K, Ahn KH, Kim B. Surface Properties of Silica Nanoparticles Modified with Polymers for Polymer Nanocomposite Applications. *Journal of Industrial and Engineering Chemistry*, 2008 **14**(4): 515–519.
52. Azizi S, Momen G, Ouellet-Plamondon C, David E. Performance Improvement of EPDM and EPDM/Silicone Rubber Composites Using Modified Fumed Silica, Titanium Dioxide and Graphene Additives. *Polymer Testing*, 2020 **84**(November 2019): 106281.
53. Khan H, Amin M, Yasin M, Ali M, Ahmad A. Effect of Hybrid-SiO<sub>2</sub> Particles

- on Characterization of EPDM and Silicone Rubber Composites for Outdoor High-Voltage Insulations. *Journal of Polymer Engineering*, 2017 **37**(7): 671–680.
54. Samaržija-Jovanović S, Jovanović V, Marković G, Konstantinović S, Marinović-Cincović M. Nanocomposites Based on Silica-Reinforced Ethylene–Propylene–Diene–Monomer/Acrylonitrile–Butadiene Rubber Blends. *Composites Part B: Engineering*, 2011 **42**(5): 1244–1250.
  55. Wen Q, Shen L, Li J, Xuan S, Li Z, Fan X, *et al.* Temperature Dependent Magneto-Mechanical Properties of Magnetorheological Elastomers. *Journal of Magnetism and Magnetic Materials*, 2020 **497**(May 2019): 165998.
  56. Zhang W, Gong X, Xuan S, Jiang W. Temperature-Dependent Mechanical Properties and Model of Magnetorheological Elastomers. *Industrial & Engineering Chemistry Research*, 2011 **50**(11): 6704–6712.
  57. Wan Y, Xiong Y, Zhang S. Temperature Dependent Dynamic Mechanical Properties of Magnetorheological Elastomers: Experiment and Modeling. *Composite Structures*, 2018 **202**(March): 768–773.
  58. Ju B, Tang R, Zhang D, Yang B, Yu M, Liao C. Temperature-Dependent Dynamic Mechanical Properties of Magnetorheological Elastomers under Magnetic Field. *Journal of Magnetism and Magnetic Materials*, 2015 **374**: 283–288.
  59. Xiang C, Gao P, Liu H, Zhou H. Experimental and Theoretical Study of Temperature-Dependent Variable Stiffness of Magnetorheological Elastomers. *International Journal of Materials Research*, 2018 **109**(2): 113–128.
  60. Yunus NA, Mazlan SA, Ubaidillah, Aziz SAA, Shilan ST, Wahab NAA. Thermal Stability and Rheological Properties of Epoxidized Natural Rubber-Based Magnetorheological Elastomer. *International Journal of Molecular Sciences*, 2019 **20**(3): 746.
  61. Feng J, Yan Y, Chen D, Ni W, Yang J, Ma S, *et al.* Study of Thermal Stability of Fumed Silica Based Thermal Insulating Composites at High Temperatures. *Composites Part B: Engineering*, 2011 **42**(7): 1821–1825.
  62. Ashtiani M, Hashemabadi SH. The Effect of Nano-Silica and Nano-Magnetite on the Magnetorheological Fluid Stabilization and Magnetorheological Effect. *Journal of Intelligent Material Systems and Structures*, 2015 **26**(14): 1887–1892.



63. Torbati-Fard N, Hosseini SM, Razzaghi-Kashani M. Effect of the Silica-Rubber Interface on the Mechanical, Viscoelastic, and Tribological Behaviors of Filled Styrene-Butadiene Rubber Vulcanizates. *Polymer Journal*, 2020 **52**(10): 1223–1234.
64. Mokhothu TH, Luyt AS, Morselli D, Bondioli F, Messori M. Influence of in Situ -Generated Silica Nanoparticles on EPDM Morphology, Thermal, Thermomechanical, and Mechanical Properties. *Polymer Composites*, 2015 **36**(5): 825–833.
65. Saiz-Arroyo C, Escudero J, Rodríguez-Pérez MA, De Saja JA. Improving the Structure and Physical Properties of LDPE Foams Using Silica Nanoparticles as an Additive. *Cellular Polymers*, 2011 **30**(2): 63–78.
66. Lokander M, Stenberg B. Improving the Magnetorheological Effect in Isotropic Magnetorheological Rubber Materials. *Polymer Testing*, 2003 **22**(6): 677–680.
67. Ravishankar S, Mahale R. A Study on Magneto Rheological Fluids and Their Applications. *International Research Journal of Engineering and Technology*, 2015 **2**(4): 2023–2028.
68. Spaggiari A. Properties and Applications of Magnetorheological Fluids. *Journal of Achievements in Materials and Manufacturing Engineering*, 2006 **18**(1–2): 127–130.
69. Aziz SAA, Mazlan SA, Nordin NA, Rahman NANA, Ubaidillah U, Choi SB, *et al.* Material Characterization of Magnetorheological Elastomers with Corroded Carbonyl Iron Particles: Morphological Images and Field-Dependent Viscoelastic Properties. *International Journal of Molecular Sciences*, 2019 **20**(13): 3311.
70. Wu JK, Gong XL, Fan YC, Xia HS. Anisotropic Polyurethane Magnetorheological Elastomer Prepared through in Situ Polycondensation under a Magnetic Field. *Smart Materials and Structures*, 2010 **19**(10): 105007.
71. Lu X, Qiao X, Watanabe H, Gong X, Yang T, Li W, *et al.* Mechanical and Structural Investigation of Isotropic and Anisotropic Thermoplastic Magnetorheological Elastomer Composites Based on Poly(Styrene-*b*-Ethylene-Co-Butylene-*b*-Styrene) (SEBS). *Rheologica Acta*, 2012 **51**(1): 37–50.
72. Lokander M, Stenberg B. Performance of Isotropic Magnetorheological Rubber Materials. *Polymer Testing*, 2003 **22**(3): 245–251.
73. Bokobza L. Natural Rubber Nanocomposites: A Review. *Nanomaterials*, 2018

- 9(1): 12.
74. Shuib RK, Pickering KL, Mace BR. Dynamic Properties of Magnetorheological Elastomers Based on Iron Sand and Natural Rubber. *Journal of Applied Polymer Science*, 2015 **132**(8): 41506.
  75. Aziz SAA, Mazlan SA, Ismail NIN, Ubaidillah, Choi SB, Nordin NA, *et al.* A Comparative Assessment of Different Dispersing Aids in Enhancing Magnetorheological Elastomer Properties. *Smart Materials and Structures*, 2018 **27**(11): 117002.
  76. Aziz SAA, Mazlan SA, Ismail NIN, Choi SB, Ubaidillah, Yunus NA. An Enhancement of Mechanical and Rheological Properties of Magnetorheological Elastomer with Multiwall Carbon Nanotubes. *Journal of Intelligent Material Systems and Structures*, 2017 **28**(20): 3127–3138.
  77. Ali M, Choudhry MA. Preparation and Characterization of EPDM-Silica Nano/Micro Composites for High Voltage Insulation Applications. *Materials Science- Poland*, 2015 **33**(1): 213–219.
  78. Akpinar Borazan A. Preparation and Characterization of Ethylene Propylene Diene Monomer (EPDM) Rubber Mixture for a Heat Resistance Conveyor Belt Cover. *Anadolu University Journal of Science and Technology A - Applied Sciences and Engineering*, 2017 **18**(2): 507–520.
  79. Habieb AB, Milani F, Milani G, Cerchiaro R. Rubber Compounds Made of Reactivated EPDM for Fiber-Reinforced Elastomeric Isolators: An Experimental Study. *Iranian Polymer Journal (English Edition)*, 2020 **29**(11): 1031–1043.
  80. Yunus NA, Mazlan SA, Ubaidillah, Choi SB, Imaduddin F, Aziz SAA, *et al.* Rheological Properties of Isotropic Magnetorheological Elastomers Featuring an Epoxidized Natural Rubber. *Smart Materials and Structures*, 2016 **25**(10): 107001.
  81. Tian T, Nakano M. Fabrication and Characterisation of Anisotropic Magnetorheological Elastomer with 45° Iron Particle Alignment at Various Silicone Oil Concentrations. *Journal of Intelligent Material Systems and Structures*, 2018 **29**(2): 151–159.
  82. Ubaidillah, Mazlan SA, Sutrisno J, Yahya I, Imaduddin F. Physicochemical Properties and Stress-Strain Compression Behaviors of a Waste Based Magnetorheological Elastomers. *Scientia Iranica*, 2016 **23**(3): 1144–1159.

83. Ubaidillah, Choi HJ, Mazlan SA, Imaduddin F, Harjana. Fabrication and Viscoelastic Characteristics of Waste Tire Rubber Based Magnetorheological Elastomer. *Smart Materials and Structures*, 2016 **25**(11): 115026.
84. Farshad M, Benine A. Magnetoactive Elastomer Composites. *Polymer Testing*, 2004 **23**(3): 347–353.
85. Boczkowska A, Awietj S. Microstructure and Properties of Magnetorheological Elastomers. In: Boczkowska A, editor. *Advanced Elastomers - Technology, Properties and Applications*, InTech; 2012.
86. Agirre-Olabide I, Berasategui J, Elejabarrieta MJ, Bou-Ali MM. Characterization of the Linear Viscoelastic Region of Magnetorheological Elastomers. *Journal of Intelligent Material Systems and Structures*, 2014 **25**(16): 2074–2081.
87. Wang W, Guo J, Long C, Li W, Guan J. Flaky Carbonyl Iron Particles with Both Small Grain Size and Low Internal Strain for Broadband Microwave Absorption. *Journal of Alloys and Compounds*, 2015 **637**: 106–111.
88. Burhannuddin NL, Nordin NA, Mazlan SA, Aziz SAA, Kuwano N, Jamari SKM, *et al.* Physicochemical Characterization and Rheological Properties of Magnetic Elastomers Containing Different Shapes of Corroded Carbonyl Iron Particles. *Scientific Reports*, 2021 **11**(1): 868.
89. Kaleta J, Królewicz M, Lewandowski D. Magnetomechanical Properties of Anisotropic and Isotropic Magnetorheological Composites with Thermoplastic Elastomer Matrices. *Smart Materials and Structures*, 2011 **20**(8): 085006.
90. Boczkowska A, Awietjan SF, Wroblewski R. Microstructure–Property Relationships of Urethane Magnetorheological Elastomers. *Smart Materials and Structures*, 2007 **16**(5): 1924–1930.
91. Xu Y, Gong X, Xuan S, Zhang W, Fan Y. A High-Performance Magnetorheological Material: Preparation, Characterization and Magnetic-Mechanic Coupling Properties. *Soft Matter*, 2011 **7**(11): 5246–5254.
92. Xu Z, Wu H, Wang Q, Jiang S, Yi L, Wang J. Study on Movement Mechanism of Magnetic Particles in Silicone Rubber-Based Magnetorheological Elastomers with Viscosity Change. *Journal of Magnetism and Magnetic Materials*, 2020 **494**(August 2019): 165793.
93. Li J, Gong X, Xu Z, Jiang W. The Effect of Pre-Structure Process on Magnetorheological Elastomer Performance. *International Journal of*

- Materials Research*, 2008 **99**(12): 1358–1364.
94. Tian TF, Zhang XZ, Li WH, Alici G, Ding J. Study of PDMS Based Magnetorheological Elastomers. *Journal of Physics: Conference Series*, 2013 **412**(1): 012038.
  95. Zając P, Kaleta J, Lewandowski D, Gasperowicz A. Isotropic Magnetorheological Elastomers with Thermoplastic Matrices: Structure, Damping Properties and Testing. *Smart Materials and Structures*, 2010 **19**(4): 045014.
  96. Barman H, Hegde S. Comprehensive Review of Parameters Influencing the Performance of Magnetorheological Elastomers Embedded in Beams. *Materials Today: Proceedings*, 2020 **26**: 2130–2135.
  97. Li WH, Nakano M. Fabrication and Characterization of PDMS Based Magnetorheological Elastomers. *Smart Materials and Structures*, 2013 **22**(5): 055035.
  98. Gordaninejad F, Wang X, Mysore P. Behavior of Thick Magnetorheological Elastomers. *Journal of Intelligent Material Systems and Structures*, 2012 **23**(9): 1033–1039.
  99. Ge L, Gong X, Fan Y, Xuan S. Preparation and Mechanical Properties of the Magnetorheological Elastomer Based on Natural Rubber/Rosin Glycerin Hybrid Matrix. *Smart Materials and Structures*, 2013 **22**(11): 115029.
  100. Pauline U, Austine N, Rosemary U, Joy N. Effects of Natural Fillers on Some Properties of Polystyrene. *International Journal of Scientific and Research Publications*, 2014 **4**(7): 5–8.
  101. Khairi MHA, Mazlan SA, Ubaidillah, Ahmad KZK, Choi SB, Aziz SAA, *et al.* The Field-Dependent Complex Modulus of Magnetorheological Elastomers Consisting of Sucrose Acetate Isobutyrate Ester. *Journal of Intelligent Material Systems and Structures*, 2017 **28**(14): 1993–2004.
  102. Najam M, Hussain M, Ali Z, Maafa IM, Akhter P, Majeed K, *et al.* Influence of Silica Materials on Synthesis of Elastomer Nanocomposites: A Review. *Journal of Elastomers & Plastics*, 2020 **52**(8): 747–771.
  103. Das A, De D, Naskar N, Chandra Debnath S. Effect of Vulcanization Technique on the Physical Properties of Silica-Filled EPDM Rubber. *Journal of Applied Polymer Science*, 2006 **99**(3): 1132–1139.
  104. Park SJ, Cho KS. Filler–Elastomer Interactions: Influence of Silane Coupling

- Agent on Crosslink Density and Thermal Stability of Silica/Rubber Composites. *Journal of Colloid and Interface Science*, 2003 **267**(1): 86–91.
105. Zhang W, Gong XL, Jiang WQ, Fan YC. Investigation of the Durability of Anisotropic Magnetorheological Elastomers Based on Mixed Rubber. *Smart Materials and Structures*, 2010 **19**(8): 085008.
  106. Ng HM, Saidi NM, Omar FS, Ramesh K, Ramesh S, Bashir S. Thermogravimetric Analysis of Polymers. *Encyclopedia of Polymer Science and Technology*, Hoboken, NJ, USA: John Wiley & Sons, Inc.; 2018.
  107. Mokhothu TH, Luyt AS, Messori M. Preparation and Characterization of EPDM/Silica Composites Prepared through Non-Hydrolytic Sol-Gel Method in the Absence and Presence of a Coupling Agent. *Express Polymer Letters*, 2014 **8**(11): 809–822.
  108. Hussein M. Effects of Strain Rate and Temperature on the Mechanical Behavior of Carbon Black Reinforced Elastomers Based on Butyl Rubber and High Molecular Weight Polyethylene. *Results in Physics*, 2018 **9**: 511–517.
  109. Agirre-Olabide I, Elejabarrieta MJ, Bou-Ali MM. Matrix Dependence of the Linear Viscoelastic Region in Magnetorheological Elastomers. *Journal of Intelligent Material Systems and Structures*, 2015 **26**(14): 1880–1886.
  110. Yu M, Zhao L, Fu J, Zhu M. Thermal Effects on the Laminated Magnetorheological Elastomer Isolator. *Smart Materials and Structures*, 2016 **25**(11): 115039.
  111. Du H, Li W, Zhang N. Semi-Active Variable Stiffness Vibration Control of Vehicle Seat Suspension Using an MR Elastomer Isolator. *Smart Materials and Structures*, 2011 **20**(10): 105003.
  112. Watson JR. U.S Patent 005609353A 1997(19).
  113. Kallio M, Lindroos T, Aalto S, Järvinen E, Kärnä T, Meinander T. Dynamic Compression Testing of a Tunable Spring Element Consisting of a Magnetorheological Elastomer. *Smart Materials and Structures*, 2007 **16**(2): 506–514.
  114. Li Y, Li J, Li W, Samali B. Development and Characterization of a Magnetorheological Elastomer Based Adaptive Seismic Isolator. *Smart Materials and Structures*, 2013 **22**(3): 035005.
  115. Opie S, Yim W. Design and Control of a Real-Time Variable Modulus Vibration Isolator. *Journal of Intelligent Material Systems and Structures*, 2011

- 22(2): 113–125.
116. Liao GJ, Gong XL, Xuan SH, Kang CJ, Zong LH. Development of a Real-Time Tunable Stiffness and Damping Vibration Isolator Based on Magnetorheological Elastomer. *Journal of Intelligent Material Systems and Structures*, 2012 **23**(1): 25–33.
  117. Yang C, Fu J, Yu M, Zheng X, Ju B. A New Magnetorheological Elastomer Isolator in Shear–Compression Mixed Mode. *Journal of Intelligent Material Systems and Structures*, 2015 **26**(10): 1290–1300.
  118. Gudmundsson KH, Jonsdottir F, Thorsteinsson F. A Geometrical Optimization of a Magneto-Rheological Rotary Brake in a Prosthetic Knee. *Smart Materials and Structures*, 2010 **19**(3): 035023.
  119. Van Duin M, Orza R, Peters R, Chechik V. Mechanism of Peroxide Cross-Linking of EPDM Rubber. *Macromolecular Symposia*, 2010 **291–292**(1): 66–74.
  120. Retamal Marín R, Babick F, Lindner GG, Wiemann M, Stintz M. Effects of Sample Preparation on Particle Size Distributions of Different Types of Silica in Suspensions. *Nanomaterials*, 2018 **8**(7): 454.
  121. Rashid RZA, Johari N, Mazlan SA, Aziz SAA, Nordin NA, Nazmi N, *et al.* Effects of Silica on Mechanical and Rheological Properties of EPDM-Based Magnetorheological Elastomers. *Smart Materials and Structures*, 2021 **30**(10): 105033.
  122. Geethamma VG, Sampath V. Rubber as an Aid to Teach Thermodynamics. *Resonance*, 2019 **24**(2): 217–238.
  123. Kim JY, Kim SH, Kang SW, Chang JH, Ahn SH. Crystallization and Melting Behavior of Silica Nanoparticles and Poly(Ethylene 2,6-Naphthalate) Hybrid Nanocomposites. *Macromolecular Research*, 2006 **14**(2): 146–154.
  124. Meng F, Elshahati M, Liu J, Richards RF. Thermal Resistance between Amorphous Silica Nanoparticles. *Journal of Applied Physics*, 2017 **121**(19): 194302.
  125. Agirre-Olabide I, Elejabarrieta MJ. Effect of Synthesis Variables on Viscoelastic Properties of Elastomers Filled with Carbonyl Iron Powder. *Journal of Polymer Research*, 2017 **24**(9): 139.
  126. Sorokin V V., Ecker E, Stepanov G V., Shamonin M, Monkman GJ, Kramarenko EY, *et al.* Experimental Study of the Magnetic Field Enhanced

Payne Effect in Magnetorheological Elastomers. *Soft Matter*, 2014 **10**(43):  
8765–8776.

## LIST OF PUBLICATIONS

### Journal with Impact Factor

1. **Rusila Zamani Abd Rashid**, Norhasnidawani Johari, Saiful Amri Mazlan, Siti Aishah Abdul Aziz, Nur Azmah Nordin, Nurhazimah Nazmi, S N Aqida and Mohd Aidy Faizal Johari. Effects of silica on mechanical and rheological properties of EPDM-based magnetorheological elastomers. *Smart Materials and Structures*. 30, 10 (2021) (Q1, IF: 3.748) PUBLISHED
2. **Rusila Zamani Abd Rashid**, Nurul Azhani Yunus, Saiful Amri Mazlan, Norhasnidawani Johari, Siti Aishah Abdul Aziz, Nur Azmah Nordin, Muntaz Hana Ahmad Khairi and Mohd Aidy Faizal Johari. Temperature Dependent on Mechanical and Rheological Properties of EPDM-Based Magnetorheological Elastomers Using Silica Nanoparticles. *Materials*. 15, 2556 (2022) (Q1, IF: 4.13) PUBLISHED

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1. Mechanism of Rubber-Particle Interaction in EPDM-silica based Magnetorheological Elastomers (LY202005095)
2. Mechanism of Incorporation Nano Particles within the Matrix (LY202005099)
3. Procedure for Durability Evaluation of Magnetorheological Elastomer under Oscillatory Shear Load (LY202004884)
4. Process of Nano Coating on Metal Substrate (LY202005098)
5. Formation Mechanism of Proposed Superhydrophobic Zinc Oxide/Epoxy Coating with Hierarchical Rough Surface (LY202005097)
6. Schematic Illustration of the Fabrication Steps of Superhydrophobic Zinc Oxide/Epoxy Surface on Metal Substrate (LY202005096)
7. Procedure for Large Strain Evaluation of Magnetorheological Elastomers Under Tension Load (LY2022W00748)