ELECTRICAL CONDUCTIVITY AND RHEOLOGICAL PROPERTIES OF MAGNETORHEOLOGICAL PLASTOMER USING CONDUCTIVE MATRIX AND GRAPHITE FOR SENSOR APPLICATIONS

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DEDICATION

This thesis is dedicated to my beloved family member, especially my father and mother, who always taught me that the most value treasure in the world is knowledge which bring to the blissful and successful life.

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ABSTRACT

Researchers have attempted to identify the best approach to utilize the Magnetorheology (MR) material such as in resistivity sensing-based devices. Magnetorheological Elastomer (MRE) material has attracted researchers due to its broad applications that require high electrical conductivity. However, the properties of MRE in a solid-like form is too rigid to be molded into certain devices, making the particles inside the matrix structure to remain trapped, thus resulting in poor electrical conductivity. Therefore, Magnetorheological Plastomer (MRP) is introduced which offers a new sensing capability due to its flexibility, soft nature, responsiveness to external magnetic field and simultaneously, conducts electricity. In this study, the rheology and resistance properties of Graphite (Gr) based Magnetorheological Plastomer (MRP) have been studied to enhance the electrical conductivity while maintaining the rheological properties. Even though previous studies have proven the capability of Gr in MRP in electrical and rheological properties, the effects of Gr in MRP are still low due to the use of non-conductive material as a matrix. Therefore, Polyvinyl Alcohol (PVA) material was used as a conductive matrix together with MRP with various content of Gr, from 0 to 10 wt.%, and the magnetic field-dependent electrical property was tested. The morphological aspect of Gr-MRP was identified using environmental scanning electron microscopy (ESEM). Besides, the magnetic property of MRP and Gr-MRP was tested using a vibrating sample magnetometer (VSM). The resistance value of Gr-MRP was assessed using a test rig under various applied magnetic flux densities. The results showed that the resistance of Gr-MRP decreased with the increase of Gr content up to 10 wt.%. The resistivity value reached a plateau at 400mT, with a value of 1.35×104 kΩ.m, possibly caused by the movement of Gr and CIPs assisted by an external magnetic field. Hence, the possible particle movement mechanism related to Gr and CIPs was also discussed. Moreover, the samples' electrical conductivity showed a proportional response to the addition of Gr value. The electrical conductivity of 10 wt.% Gr–MRP material was found to be the highest, approximately 178.06% as compared to 6 wt.%. It was also observed that with the addition of Gr, the conductivity properties were improved with the increasing of magnetic flux densities, while maintaining the storage modulus verified by using rheometer. This could contribute to the practicality of this material as a sensing detection device.

ABSTRAK

Para penyelidik telah cuba mengenal pasti kaedah terbaik melalui penggunaan magnetorheologi (MR) untuk kegunaan dalam bidang peranti berasaskan pengesanan rintangan. Bahan Elastomer Magnetorheologi (MRE) telah menarik minat para penyelidik kerana bahan tersebut mempunyai konduktiviti elektrik yang tinggi untuk pelbagai aplikasi. Walaubagaimanapun, sifat MRE dalam bentuk pepejal adalah keras untuk diubah bentuk dan dimasukkan ke dalam beberapa peranti tertentu, menyebabkan zarah-zarah di dalam maktriks kekal terperangkap, mengakibatkan konduktiviti elektrik yang rendah. Untuk mengatasinya, bahan Plastomer Magnetorheologi (MRP) telah diperkenalkan kerana kebolehan fleksibiliti, boleh dibentuk, bertindakbalas dengan magnet dan dapat mengalirkan arus elektrik. Dalam kajian ini, sifat reologi dan rintangan bagi bahan Grafit (Gr) berasaskan Plastomer Magnetorheologi (MRP) telah dikaji untuk meningkatkan kekonduksian elektrik sambil mengekalkan sifat rheologi. Walaupun kajian telah membuktikan kebolehan penggunaan Gr dalam MRP untuk pengaliran dan sifat reologi, kesan Gr dalam MRP masih kekal rendah disebabkan penggunaan bahan bukan konduktif sebagai matriks. Oleh itu, bahan Polyvinyl Alkohol (PVA) telah digunakan sebagai matriks konduktif bersama dengan Gr untuk meningkatkan konduktiviti elektrik sambil mengekalkan sifat rheologi. Sampel MRP dan pelbagai kandungan Gr, dari 0 hingga 10 wt.%, telah disediakan, dan sifat daya medan magnet yang bergantung kepada arus elektrik telah dinilai secara eksperimen. Aspek morfologi Gr-MRP telah dikenalpasti menggunakan peralatan mikroskop elektron pengimbasan alam sekitar (ESEM). Di samping itu, sifat daya medan magnet MRP dan GR-MRP telah dinilai menggunakan peralatan magnetometer sampel bergetar (VSM). Nilai rintangan Gr- MRP telah diuji menggunakan rig ujian dengan pelbagai ketumpatan fluks magnet. Hasil pengujian telah menunjukkan bahawa nilai rintangan Gr-MRP menurun dengan peningkatan Gr sehingga 10 wt.%. Pada 400mT, daya rintangan menurun hingga 1.35×10^4 kΩ.m, disebabkan oleh pergerakan Gr dan CIP yang dibantu daya medan magnet luar. Oleh itu, mekanisme pergerakan zarah yang berkaitan dengan Gr dan CIP juga telah dibincangkan. Selain itu, konduktiviti elektrik menunjukkan kenaikan dengan pertambahan Gr. Konduktiviti elektrik bagi bahan 10 wt.% Gr-MRP didapati tertinggi, 178.06% berbanding 6 wt.%. Hasil eksperimen juga mendapati dengan penambahan Gr, sifat konduktiviti elektrik telah bertambah baik dengan peningkatan intensiti magnetik, sementara mengekalkan modulus dengan menggunakan reometer. Perkara ini dapat menyumbang kepada potensi kegunaan bahan-bahan ini sebagai peranti alat pengesanan.

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LIST OF ABBREVIATIONS

BORAX	-	Sodium Tetraborate Decahydrate
CCPI	-	Copolymide
CFs	-	Carbon Microfibers
CIP	-	Carbonyl Iron Particle
CNTs	-	Carbon Nanotubes
CPCs	-	Conductive Polymer Composites
DMSO	-	Dimethyl Sulphoxide
EDX	-	Energy Dispersive X-ray Spectroscopy
ESEM	-	Environmental Scanning Electron Microscope
FESEM	-	Field Emission Scanning Electronic Microscopy
FGP	-	Flake Graphite Particle
Gr	-	Graphite
Gr-MRP	-	Graphite Magnetorheological Plastomer
LV-SEM	-	Low Vacuum Scanning Electronic Microscopy
MR	-	Magnetorheological
MRE	-	Magnetorheological Elastomer
MRF	-	Magnetorheological Fluid
MRG	-	Magnetorheological Grease
MRP	-	Magnetorheological Plastomer
MRPG	-	Magnetorheological Plastomer Graphite
PP20	-	Parallel Plate 20
PU	-	Polyurethane
PVA	-	Polyvinyl Alcohol
SEM	-	Scanning Electronic Microscope
TDI	-	Toluene Diisocyanate
TEM	-	Transmission Electron Microscopy
VSM	-	Vibrating Sample Magnetometer

LIST OF SYMBOLS

%	-	Percentage
˚C	-	Degree Celcius
µm	-	Micronmeter
C	-	Carbon
F	-	Fareinheit
Fe	-	Iron
g/cm^3	-	Gram per Cubic Centimeter
G’	-	Storage Modulus
H_c	-	Coercivity
K	-	Kelvin
kA/m	-	Kilo Ampere per Meter
kV	-	Kilo Voltmeter
KΩ	-	Kilo Ohm
kΩ.m	-	Kiloohmmeter
M_r	-	Magnetic Retentivity
M_s	-	Magnetic Saturation
mT	-	Millitesla
O	-	Oxygen
T	-	Tesla
Wt.%	-	Weight Percentage
ρ	-	Resistivity
σ	-	Conductivity
Ω	-	Ohm

CHAPTER 1

INTRODUCTION

1.1 Research Background

Flexible electronic sensors have attracted a lot of attention in recent years because of their remarkable flexibility and potential applications. This kind of sensor has been widely applied nowadays in many practical applications such as in automotive [1], aerospace [2], manufacturing [3], and even human motion monitoring [4]. Conventional electronic sensors made of semiconductors and metallic materials, as well as flexible strain devices, offer great sensing sensitivity [5], outstanding stretchability [6], and the ability to detect both small and big movement [7]. In particular, a number of smart materials is getting considerable attention by gaining a lot of interest in a broad range of applications. Potential smart materials include electrorheological materials, electroactive polymers, and magnetorheological (MR) materials. MR materials are one example of a material that could be employed in smart devices.

MR materials belong to an important part of the field of smart materials, which their rheological properties change rapidly with magnetic field. More specifically, when the external stimulus such as magnetic field is exerted, the rheological properties of MR materials can be quickly tuned or controlled due to the chain formation structure according to the field direction [8]. MR materials can be classified into many groups such as MR fluid (MRF), MR elastomer (MRE), MR foam, MR grease (MRG), MR gel and MR plastomer (MRP) [9-14]. Recently, MRP has received growing attention due to better stability and higher MR effect than other MR materials [15,16]. Unlike conventional MR gel, MRP is a solid-like gel that functions like a plasticine. It canbe made into different forms and the shapes can be held for a long time. At an early stage, MRP, which normally consists of non-magnetic responsive iron particles (3-5 μm) dispersed into a low-crosslinking polymer matrix has become a promising candidate to substitute the traditional MR materials [17]. Due to their high saturation magnetization of metallic elements, high permeability, and relatively low magnetization, carbonyl iron particles (CIPs) are the most extensively utilized magnetic material.

Due to the tunable electrical properties of MR material, the study of electrical properties on MR material has drawn a variety of scholars up to this point. The material can nevertheless conduct electricity because CIPs are insulating ferromagnetic particles dispersed in a conductive matrix [16,17]. Therefore, in on-state condition, MR material can transform from an electric insulator to an electric conductor[18-20]. In the absence of a magnetic field, magnetic particles are dispersed at random throughout the medium or matrix and no conductive channels are formed, resulting in the material acting as an insulator [21]. Magnetic particles form a chain parallel tothe applied magnetic field in on-state condition. The stronger the applied magnetic field is, the longer and tighter the chain is formed. The material becomes an electrical conductor because of these complete chains, which create conductive paths for electron transfer [22-24].

1.2 Motivation of Research

MRPs can be classified into several categories by matrix types. As indicated by Xuan et al. [25], MRPs is one of the plastic MR materials that can be categorized into three main groups, which are a hydrogel [26,27], swollen polymer gels [28,29] and pure polymer gels [30-32]. Amongst these groups of MRPs, the fabrication of hydrogel based MRPs are the least demanding and conservative to be rehearsed. Magnetic particles are embedded inside a polymer matrix network that has been swelled by a liquid solution such as water or a water-miscible organic solvent [33]. Hydrogel MRP can also be categorized under solid-like MR gel. Furthermore, these MRPs have a better stability and higher MR performance [15,16,34]. MRP is normally consists of micron size of iron particles (usually 3-5 micron) dispersed in a non-magnetic solid, liquid or gel-like matrix. CIPs are the most commonly utilized magnetic material in MR materials due to their high saturation magnetization of metallic elements, as well as their high permeability and low residual magnetization [34].

It is known that poor electrical conductivity, high resistivity and low sensitivity of conventional electronic sensors have limited their use in the diverse applications. Thus, many attempts have been made to find viable candidates to overcome these limitations by utilizing different types of matrix; polyurethane (PU), polyacrylamide and polystyrene, and the introduction of additives in the material; carbon nanotubes (CNTs) [35], carbon nanofibers (CFs) [36] and graphite (Gr) [37] to further improve mechanical, chemical and electrical properties of MR materials. As such, until to date, PU has been used widely due to its better sedimentation stability [38]. However, PU has some drawbacks such as poor compatibility with hydrocarbon matrix resulting in low electrical conductivity [39]. Recently, polyvinyl alcohol (PVA) has been introduced to overcome PU issues to accomplish better mechanical performance, biocompatible and good electrical conductivity, making it very useful for the development of sensor application systems [30]. As a result, PVA has been used as a conductive matrix, which is believed to improve electrical conductivity [40]. In another study, PVA was used as a matrix with the addition of Gr oxide additive to examine electrical conductivity. However, their research was focused the rheological properties only. In another study, Hapipi et al. [41] utilized PVA as a matrix together with CIP as a magnetic particle to investigate on the rheological properties.

On the other hand, the use of Gr in MR materials particularly MRE has been proven to enhance the electrical properties. Besides, Gr is the only non-metal material that can conduct electricity despite its low density. Bica [42] revealed that MRE was capable to become electroconductive with the addition of 14% Gr micro particles. The finding was supported by Huang et al. [43], who discovered that the Gr could change the function of MRE to either insulator (<14%) or electrical conductor (>14%). The problem comes when the solid-state MRE is too rigid to be utilized into specific devices such as soft sensor. When the material is rigid, it is difficult to be shaped and despite has low conductivity. Therefore, in MRP, Pang et al. [44] studied the rheological and conductivity properties of PU based MRP with the addition of flake Gr. The results showed that the electrical conductivity of MRP was increased by 10000 times relative to pure MRP when 15 wt.% Gr flakes were added to it.

1.3 Problem Statement

In general, MRE has attracted researchers interested in studying the conductivity properties of MR materials for a wide range of applications that require high electrical conductivity, especially in sensors. The solid-state MRE, on the other hand, is too rigid to be moulded or fitted into certain devices. When the materials are rigid, the magnetic particles inside the low crosslinked matrix are mobile and not permanently trapped making the particles are difficult to form and have poor electrical conductivity. As a result, it is not suitable for some applications. A semi-solid form of MR material, such as MRP, is the best candidate for research since it is soft and flexible, has high conductivity, and good magnetic properties, and can generate electricity. Previous researchers utilized PU as a matrix in MRP to maintain the flexibility of material. However, PU based MRP has low conductivity since PU is an insulator and has some drawbacks such as poor compatibility with hydrocarbon matrix resulting in low electrical conductivity. Therefore, a comprehensive investigation has to be undertaken to gain a fundamental understanding of the effect of Gr concentration towards the magnetization, morphological, and electrical conductivity properties by using PVA as a conductive matrix in MRP.

1.4 Research Objectives

The main objective of this research is to investigate the electrical performance of PVA based Gr-MRP. The primary objectives for this research are:

(a) To characterize the PVA based MRP and Gr-MRP in terms of morphological and magnetic properties.

(b) To analyse the resistivity and electrical conductivity of Gr-MRP at different magnetic field strengths.

(c) To evaluate the effect of Gr on the rheological properties of MRP and Gr-MRP at different magnetic field strengths.

1.5 Research Scopes

The scope of this thesis is specified on the experimental investigation of graphite as an additive in MRP fabrication as well as fundamental characterization to indicate their potential ability in a real application. The scopes are listed.

(a) Gr-MRP is fabricated using various weight percentage of graphite additive (0, 2, 4, 6, 8 and 10 wt.%).

(b) The physicochemical characteristics including magnetic properties, size of Gr and distribution of CIPs and Gr in MRP are carried out through ESEM and VSM at off-state and on-state conditions.

(c) The arrangement of CIPs and Gr in plastomer matrix under influence of magnetic fields is examined via ESEM by pre-treated the sample with 0.1T magnetic fields for 5 minutes.

(d) Resistance and electrical conductivity value of MRP is measured through proper set up with absence and presence of magnetic fields (0 to 400 mT).

(e) The rheological properties of the MRP under oscillatory mode are investigated using rheometer. The effect of frequency sweep on MRP and Gr-MRP is presented in terms of storage modulus.

1.6 Significance of Research

The significance of this research is summarized in the following points:

(a) This research can provide phenomenology study on the physicochemical, electrical conductivity, resistivity and rheological properties of Gr-MRP.

(b) The resistivity and electrical properties performance of MRP can be improved by the introduction of additives in MRP. This is due to the features owned by Gr, led in developing a great interaction between the conductive matrix and CIPs thus results in creation of strong polarized chain structure within the matrix with the influence of magnetic fields. Therefore, the utilization of Gr in MRP with high conductivity range can be potentially used in sensing application.

1.7 Thesis Outline

This thesis consists of five chapters. The related information, achievements, and findings are highlighted in each chapter. The outline of the chapters is established as:

Chapter 1	is the introduction of the thesis. A brief introduction of the research is undertaken including the research background, problem statement, research objectives, research scope and significance of the research.
Chapter 2	comprises a brief literature review on the MR materials, MRP and materials characterization that related to previous information and understanding of MRP.
Chapter 3	explains the methodology and experimental flow of the research. The experimental methodology is divided by two stages; the fabrication of MRP samples that incorporates with various concentrations of Gr. The Gr-MRP is characterized in terms of magnetic properties, morphological properties, elemental composition and electrical properties.
Chapter 4	presents the results and discussion on the materials characterizations and electrical properties of Gr-MRP samples. All the data are demonstrated in forms of image, table and plotted graph.
Chapter 5	summarizes the highlight of this thesis that corresponds to the objectives. Finally, some recommendations for future works are presented as an extension of the existing research.

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