Smart material based on magnetorheological elastomer and its 3D printing

Bastola, Anil K.

2019

Bastola, A. K. (2019), Smart material based on magnetorheological elastomer and its 3D printing. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/137581>

<https://doi.org/10.32657/10356/137581>

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Smart Material Based on Magnetorheological Elastomer and its 3D Printing

ANIL K. BASTOLA

SCHOOL OF MECHANICAL AND AEROSPACE ENGINEERING

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SCHOOL OF MECHANICAL AND AEROSPACE ENGINEERING

A thesis submitted to the Nanyang Technological University in partial
fulfilment of the requirement for the degree of

Doctor of Philosophy

2019

Statement of Originality

I hereby certify that the work embodied in this thesis is the result of original research, is free of plagiarized materials, and has not been submitted for a higher degree to any other University or Institution.

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Anil K. Bastola

Supervisor Declaration Statement

I have reviewed the content and presentation style of this thesis and declare it is free of plagiarism and of sufficient grammatical clarity to be examined. To the best of my knowledge, the research and writing are those of the candidate except as acknowledged in the Author Attribution Statement. I confirm that the investigations were conducted in accord with the ethics policies and integrity standards of Nanyang Technological University and that the research data are presented honestly and without prejudice.

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Date

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Assoc. Prof. Li Lin

Authorship Attribution Statement

This thesis contains material from following papers published/under review in the following peer-reviewed journals where I was the first author.

Part of Chapter 4 is published as A. K. Bastola, L. Li, M. Paudel “A hybrid magnetorheological elastomer developed by encapsulation of magnetorheological fluid”, *Journal of Materials Science*, 53, 7004–7016 (2018) and A.K Bastola, E. Ang, M. Paudel and L. Li “ Soft hybrid magnetorheological elastomer: gap bridging between MR fluid and MR elastomer”, *Colloids and Surfaces A: Physicochemical and Engineering Aspects*.

The contributions of the co-authors are as follows:

- Prof L. Li provided the initial project direction and edited the manuscript drafts.
- M. Paudel and I co-designed the study and modified the testing fixtures.
- E. Ang performed a part the experimental study, sample development and cyclic compression for core-shell hybrid MR elastomers
- I performed all the laboratory work and analyzed the data.
- I prepared the manuscript drafts. The manuscript was revised by M. Paudel and Prof. Li Lin.

Part of Chapter 5 is published as A. K. Bastola, T. V. Hoang, L. Li, “A Novel Hybrid Magnetorheological Elastomer Developed by 3D Printing”, *Materials & Design*, 114, 391–397 (2017), A. K. Bastola, M. Paudel, L. Li, “Development of hybrid magnetorheological elastomers by 3D printing”, *Polymer*, 149, 213–228 (2018), A.K. Bastola M. Paudel, L. Li “ Line-patterned hybrid magnetorheological elastomer developed by 3D printing”, *Journal of Magnetism and Magnetic Materials* and, A.K. Bastola M. Paudel, L. Li “Dot-patterned hybrid magnetorheological elastomer developed by 3D printing”, *Journal of Intelligent Material Systems and Structures*

The contributions of the co-authors are as follows:

- Prof L. Li provided the initial project direction and edited manuscripts.

- T. V. Hoang and I conducted materials selection for 3D printing and wrote the first manuscript.
- M. Paudel and I co-designed the experimental setup for compression testing and forced vibration testing.
- I performed all the test and conducted data analysis.
- I prepare the manuscript drafts. The manuscript was revised by M. Paudel, T.V. Hoang and prof. Li Lin.

Chapter 6 is published as Anil K. Bastola and Lin Li, “A new type of vibration isolator based on magnetorheological elastomer”, *Materials & Design*, 157, 431–436 (2018).

The contributions of the co-authors are as follows:

- Prof Li Lin provided the initial project direction and edited the manuscript.
- I performed experimental design, experiment, data collection, data analysis and prepared the draft manuscript.

02 Aug 2019

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Date

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Anil K. Bastola

List of Publications

Published Journal Articles

- [1]. **A. K. Bastola**, T. V. Hoang, L. Li, “A Novel Hybrid Magnetorheological Elastomer Developed by 3D Printing”, Materials & Design, 114, 391–397 (2017).
- [2]. **A. K. Bastola**, L. Li, M. Paudel “A hybrid magnetorheological elastomer developed by encapsulation of magnetorheological fluid”, Journal of Materials Science, 53, 7004–7016 (2018).
- [3]. **A. K. Bastola**, M. Paudel, L. Li, “Development of hybrid magnetorheological elastomers by 3D printing”, Polymer, 149, 213–228 (2018).
- [4]. **Anil K. Bastola**, L. Li, “A new type of vibration isolator based on magnetorheological elastomer”, Materials & Design, 157, 431–436 (2018).
- [5]. **A. K. Bastola**, M. Paudel, L. Li, “Magnetic circuit analysis to obtain the magnetic permeability of magnetorheological elastomers”, Journal of Intelligent Material Systems and Structures, 29, 2946–2953 (2018).
- [6]. Sijun Liu, **Anil Kumar Bastola**, and Lin Li, “A 3D Printable and Mechanically Robust Hydrogel Based on Alginate and Graphene Oxide” ACS Applied Materials & Interfaces, 47, 41473–41481, (2017).
- [7]. **A.K. Bastola** M. Paudel, L. Li “ Line-patterned hybrid magnetorheological elastomer developed by 3D printing”, Journal of Intelligent Material Systems and Structures, 31 377-388 (2019)
- [8]. **A.K. Bastola** M. Paudel, L. Li “Dot-patterned hybrid magnetorheological elastomer developed by 3D printing”, Journal of Magnetism and Magnetic Materials 494, 165825 (2020).
- [9]. **A.K. Bastola**, E. Ang, M. Paudel and L. Li “ Soft hybrid magnetorheological elastomer: gap bridging between MR fluid and MR elastomer”, Colloids and Surfaces A: Physicochemical and Engineering Aspects 583, 123975 (2019).

Conference Proceedings and Presentations

- [1]. **Anil K. Bastola**, Vin T. Hoang, Li Lin, “Magnetorheological Elastomer: A Novel Approach of Synthesis”, in 2nd International Conference, in Sports Science and Technology (ICSST2016), 12-13 December 2016, Singapore.

- [2]. **Anil K. Bastola**, Milan Paudel, Li Lin, “3D Printed Magnetorheological Elastomers”, in ASME 2017 Conference on Smart Materials, Adaptive Structures and Intelligent Systems, 18–20 September 2017, Snowbird, Utah, USA.
- [3]. **Anil K. Bastola**, Milan Paudel, Li Lin, “Patterned Magnetorheological Elastomer Developed by 3D Printing”, in 2nd International Conference on Advanced Manufacturing and Materials (ICAMM 2018), 9-11 June 2018, Tokyo, Japan.

Abstract

Intelligent or smart materials have one or more properties that can be significantly changed in a controlled fashion by external stimuli, such as temperature, pH, electric or magnetic fields, etc. Magnetorheological (MR) materials are a class of smart materials whose properties can be varied by applying an external magnetic field. Two major branches of the magnetorheological materials are MR fluids and MR elastomers. The MR fluids largely suffer from the sedimentation problem. Whereas, MR elastomers conquer the sedimentation problem, the price is to be paid for lower MR effect. For current MR elastomers, a need of magnetic field is mandatory during the fabrication process for the anisotropic configuration of the magnetic particles within the matrix material. However, the applied magnetic field does not promise a unique particle alignment. Similarly, main concerns of current MR elastomer-based applications such as vibration isolators and absorbers are a high-power requirement and bulky configuration. To this end, the focus of this study is to explore the development of hybrid MR elastomer that potentially bridges the gap between MR fluid and MR elastomer. Two different types of fabrication methods have been adopted, first is conventional molding and second is additive manufacturing also known as 3D printing. This is the first time to implement 3D printing method to develop MR elastomers, thus, feasibility and implementation of a new fabrication method, 3D printing has been studied in detail. Lastly, this study explores the techniques to lower the range of the magnetic field needed for the current MRE-based systems.

In the conventional fabrication method, a cavity of an elastomer matrix was formed by molding and MR materials were deposited into the cavity to form an MR core within the elastomer matrix. Three different types of MR cores, namely, low viscosity fluid MR core, high viscosity fluid MR core, and solid MR core were considered. On the other hand, in a 3D printing method, extrusion-based multi-material printing was implemented, where MR fluid filaments were printed within an elastomer matrix in the layer-by-layer fashion. The choice of printing materials determines the final structure of the 3D printed hybrid MR elastomer. Printing with a vulcanizing MR suspension produces the solid MR structure inside the elastomer matrix while printing with a non-vulcanizing MR suspension (MR fluid) results in the structures that the MR fluid is encapsulated inside the elastomer matrix. The 3D printability of different materials has

been studied by measuring their rheological properties and it was found that the highly shear thinning, and thixotropic properties are important for 3D printability. The quality of the printed filaments strongly depends on the key printing parameters such as extrusion pressure, initial height, feed rate and time. Two different patterned were considered in this study, namely, line-patterned and dot-patterned 3D printed MR elastomers (3DP-MREs), line-patterned samples were formed by continuous printing while dot-patterned samples were formed by discontinuous printing.

The cyclic compression and forced vibration experimental testing show that both conventionally developed, and the 3D printed MR elastomers could change their elastic and damping properties when exposed to an external magnetic field. Furthermore, both core-shelled and 3D printed MR elastomers also exhibit the anisotropic behavior when the direction of the magnetic field is changed with respect to the orientation of the printed filaments or printed layers.

Core-shell hybrid MR elastomer with high viscosity fluid core avoid the sedimentation issue of the current MR fluid and show higher MR effect than that of current MR elastomers and also exhibits anisotropic MR effect. Similarly, the sedimentation and settling of magnetic particles are highly unlikely within the small MR fluid filaments in 3DP-MREs. The 3D printing method can develop different configurations of MR fluid or magnetic particles without applying a magnetic field. Putting together, 3DP-MREs or core-shell hybrid MREs with high viscosity fluid MR core are bridge materials between MR fluid and MR elastomers. Moreover, core-shell hybrid MR elastomer and 3DP-MREs also offer the low working range of magnetic field, the increase in the stiffness is much gentler after 300 mT magnetic field.

Lastly, a new method was introduced for the development of MRE based vibration isolator by the simultaneous application of preloading and magnetic field. The vibration isolator can work in the low magnetic field range. The preloading effect was noteworthy for significant enhancing the performance of vibration isolator even in lower magnetic field strength.

Acknowledgments

It is my pleasure to acknowledge following people whose advice, guidance, talks, and supply are extremely helpful for the successful completion of this incredible journey.

I would like to express my deep sense of gratitude to my supervisor associate professor Li Lin for his continued support of my doctoral research, for his encouragement, guidance, suggestions and unselfish help and support. I am so thankful to my supervisor for reading and reviewing my manuscripts and thesis.

I am very grateful to my co-worker and a good friend, Mr. Vinh Hoang Tan, who helped me a lot during my first year of Ph.D. Vinh's help and support were invaluable for the initial work on 3D printing. I am so thankful to my very close friend, Milan Paudel, for his endless support in every step throughout the Ph.D. journey. I am always indebted to his advice and helps in the concept development, experiment design and proofreads of the manuscripts. I also appreciate my group members Li Huijun, Feng Han and Ying Zhen Low, for their help.

Thanks to the School of Mechanical and Aerospace Engineering, NTU for all the research facilities. I am thankful for the financial support from the SINGA scholarship, the Singapore government for my Ph.D. study. I am also thankful to Institute for Sports Research (ISR) at NTU for providing me a cubicle. I am grateful to thank all the laboratory technical staffs especially at Bio Lab, Materials Lab 1, and Mechanics of Machine (MOM) for their kindness and continuous support. I owe special thanks to the technical staffs at MOM, Mrs. Hali and Mr. Poon for helping to arrange shaker, accelerometers and other devices for the force vibration testing setup.

On the personal side, I am so thankful to my wife Shirish Ranabhat who always supported and encouraged me, loved me and always made me smile. I am also so thankful to Geet and Milan and the time we spent together in Singapore always remains as one of the most memorable moments of my life. I am so thankful to my mom for her care and endless love. I am also thankful to all my family members, special thanks to my brother Ramchandra for taking care of everything at home. Lastly, I would like to thank my friends Purna, Rakesh, Arjun, Rinoj, Arun and all the friends who helped me directly or indirectly in my Ph.D. journey.

Devoted to My Son

Shrian Bastola (30.10.2019)

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Chapter 1: Introduction

This chapter provides the background, problem statement, objectives and the structure of the thesis.

1.1 Background

Material technologies have remarkably influenced the human civilization and hence distinct time period can be found, being profoundly dominated by a specific class of materials used in various eras. One of the very well-known classes of materials is smart materials. The smart materials have one or more properties that can be significantly changed in a controlled fashion by external stimuli, such as temperature, pH, electric or magnetic fields, etc. The smart materials are first recognized in the late 1980s, and since then a number of studies have been devoted to achieving high performance of such smart materials. To date, several smart materials have been developed and their capability “as smart” has been successfully validated. The field of smart materials is very board and large, very general smart materials are illustrated as piezoelectric materials, shape memory alloys or polymers, electroactive, and magnetic materials.

Magneto-rheological (MR) materials belong to the category of smart materials, and their rheological and mechanical properties can be changed with respect to an externally applied magnetic field. MR materials can be a fluid, gel or solid-like elastomer. Depending upon the type of matrix material, MR fluid, ferrofluid, MR foams, and MR elastomer can be distinguished. The magnetorheological fluids (MRFs) and magnetorheological elastomers (MREs) are the two main branches of the MR materials. In MR fluids, magnetic particles are suspended in a carrier fluid such as silicone oil, whereas magnetic particles are locked in a place within a polymeric elastomer matrix in MREs. They undergo rheological and mechanical changes when an external magnetic field is applied. In particular, MR fluids are known for a large stress enhancement, whereas, MR elastomers are typically known for changing their viscoelastic modulus under the magnetic field.

1.2 Problem Statement

MR fluids have been commercialized and employed in a wide range of applications such as dampers, valves, polishing, and brakes [1-4]. However, MR fluid devices still come with inherent problems such as settling or precipitation of magnetic particles and leakage of MR fluids. One of the biggest characteristic problems of the MR fluids is the sedimentation of magnetic particles because of the large density mismatch between the magnetic microparticles and the carrier fluid. A number of efforts have been made in minimizing the sedimentation problem. One of the most widely practiced methods is the use of a higher off-state (i.e. no magnetic field) viscosity carrier fluid [1, 4, 5]. The use of higher off-state viscosity carrier fluids has been found to be effective in minimizing the sedimentation, but the high off-state viscosity is not welcomed in most of the MR fluid applications because of the handling problem and a need of higher magnetic field. Therefore, various kinds of other methods apart from changing the carrier fluid such as particles coating, the addition of nano spherical and nanowire particles, surfactants, thixotropic agent have been considered to improve the stability of the MR fluids [1, 6-10]. Although a high viscosity carrier fluid has not been used for the development of MR fluid devices it might be suitable to develop another type of magnetorheological devices which has not been explored yet.

On the other hand, the solid counterpart of MR fluids, MR elastomers have been found to be a candidate material for several applications including vibration absorbers, vibration isolators, smart actuators, and sandwich beams [2, 11-14]. But most of the applications are based on laboratory experiments only. An MR elastomer also consists of magnetically polarizable particles as in MR fluid but locked within a matrix material. Therefore, the distribution of the magnetic particles within the elastomer matrix influences the MR effect (MR effect is defined as the changed in properties when the magnetic field is applied) of MR elastomers. Typically, the distribution of magnetic particles within the elastomer matrix can be isotropic or anisotropic. The magnetic particles are randomly distributed in an isotropic MR elastomer whereas the particles are aligned in a specific direction in an anisotropic MR elastomer. It has been proven that the anisotropic MR elastomer exhibits a higher MR effect than that of isotropic MR elastomer when the direction of the applied magnetic field is parallel to the chains of the magnetic particles [11-18]. A magnetic field is always required for anisotropic

configuration of the magnetic particles during the crosslinking process in all fabrication methods reported to date. One of the efficient ways to avoid the need of a magnetic field during the crosslinking process is to make magnetic particles moveable within the matrix materials. Thus, when a magnetic field is applied, they can form chains depending on the direction of a magnetic field to show an anisotropic behavior.

The solid analog of MR fluid, MR elastomer overcomes the sedimentation problem of MR fluid. However, the consequence is a much lower MR effect when compared to the MR fluid because of the locked magnetic particles in the MR elastomer [19, 20]. Various efforts have been devoted in order to enhance the MR effect of the conventional MR elastomer such as the development of porous MR elastomer [21] and the addition of additives such as carbon black [22], carbon nanotubes [23, 24], magnetic nanoparticles [25] or using other techniques such as utilization of hard magnetic particles and soft elastomeric matrix [26-29]. However, the MR effect is still lower compared to the MR fluids. Therefore, there is a need for the development of bridge materials between MR fluids and MR elastomers. The bridge material must be able to overcome the problem of sedimentation of MR fluids and at the same time should avoid the need of magnetic field for the configuration of magnetic particles within the elastomeric matrix. Some efforts have been made to investigate the hybrid MR materials such as thermo-responsive polymer-based MR composite [20], where rheological properties can be controlled with respect to the temperature changes. Likewise, MR fluid impregnated composites [30, 31] and MR fluid encased MR elastomers have also been proposed [32-36]. The MR fluid encased MR elastomers comprise an MR fluid element as an interior and a non-magnetic elastomer casing as an exterior. Such MR fluid encased MR elastomers avoid the requirement of a magnetic field during crosslinking. However, those studies have not focused on investigating the behavior of the encased fluid regarding the concentrations of magnetic particles, stability, and viscosity of carrier fluid. On the other hand, to avoid the need of a magnetic field for fabricating anisotropic MR elastomer researchers have also developed patterned magnetorheological elastomer using conventional molding or manual patterning [37]. However, all methods used in the literature to fabricate MR elastomers are unable to exactly control the dispersion and distribution of magnetizable particles. Hence, an innovative fabrication technique has to be looked for, which must be able to easily control the configuration of magnetic particles in MR elastomer without applying a magnetic field.

In recent decades, additive manufacturing or three-dimensional (3D) printing has been substantially used in several fields including but not limited to construction, footwear, automotive, aerospace, dental and medical industries. 3D printing technology is a fabrication method where the structures are constructed in a layer by layer fashion. The fast and precise manufacturing process, suitable to produce highly customizable products, has also opened the door for additive manufacturing of smart materials and structures [38-41]. The additively fabricated smart materials or structures can alter their shape or properties over time. Therefore, the fabrication of smart materials or structure via 3D printing is often regarded as 4D printing [41, 42]. One of the biggest advantages of additive manufacturing is its ability to exactly control where and what to be deposited on a substrate from a printing head. Thus, 3D printing or additive manufacturing could be the only and best way to add magnetizable particles exactly to the pre-desired locations in an MR elastomer to be fabricated without applying a magnetic field.

From the application point of view, current MRE-based devices such as vibration isolators/absorbers have bulky configuration and require higher power if electromagnets are used. In such devices, in order to achieve a high performance and MR effect, the magnetic field (>700 mT) needs to be applied until the magnetic saturation of the magnetic particles is reached [11, 12]. On the other hand, the range of the magnetic field is much lower for MR fluid devices (<300 mT) [1]. Thus, there is also a need for an innovative technique to efficiently lower the range of magnetic field required for the development of compact MRE-based devices/systems. Hence, one of the main significances of this study is to lower the range of the magnetic field required for the current MRE-based devices.

1.1 Objectives

The focus of this study is to explore the development of hybrid MR elastomers that potentially bridge gap the between MR fluid and MR elastomer by means of conventional and 3D printing methods. Secondly to explore the technique to lower the range of magnetic field needed for the current MRE-based devices.

The main objectives to be achieved in this study are to:

- Develop a hybrid MR elastomer to potentially bridge a gap between conventional MR elastomer and MR fluid by forming a core-shell structure
- Study the MR effect of core-shell structure MR elastomer by considering particle concentration, carrier medium, and viscosity of the core medium
- Introduce 3D printing technology for fabrication of MR elastomers and develop various kinds of 3D printed MR elastomers with a unique configuration of magnetic suspension or magnetic particles without applying a magnetic field
- Characterize the magneto-mechanical properties of different kinds of hybrid MR elastomers in the presence and absence of a magnetic field
- Study the dynamic behavior of 3D printed MR elastomers under uniaxial deformation
- Develop a new type of an active MRE-based vibration isolator that works with the lower range of a magnetic field
- Fundamentally explore and understand the mechanisms behind the structure-properties results.

1.2 Structure of the Thesis

This thesis is organized to consist of seven chapters. The main contents of each chapter are given below:

Chapter 1 provides the background to magnetorheological materials and also includes problem statement and objective of the study and the structure of the thesis.

Chapter 2 is a literature review on the various research works conducted on MR materials including MR elastomers. The review provides a brief introduction to MR materials, thereafter, focuses on MR elastomer with a historical overview, choice of materials and fabrication technique and methods to improve the MR elastomer performance. Various potential applications of MR elastomers are also discussed.

Chapter 3 describes the materials and fabrication processes used to develop various kind of hybrid MR elastomers. In this project, different hybrid MR elastomers are fabricated using conventional and 3D printing methods. Furthermore, the design and development of experimental setups to characterize the hybrid MR elastomers without and with the application of a magnetic field are also presented in this chapter.

Chapter 4 contains experimental results from the core-shelled hybrid MR elastomers. This chapter focuses on the MR effect of core-shell hybrid MR elastomers that is related to the type of MR core, concentrations of magnetic particles within the MR core, stability and viscosity of carrier fluid. Similarly, the enhancement of the MR effect by the addition of magnetic nanoparticles (MNPs) is also presented.

Chapter 5 provides the detail and challenges when developing hybrid MR elastomers by a 3D printing method. Various MR fluid filaments were precisely and accurately configured and encapsulated in the elastomeric matrix. The MR effects shown by 3D printed various patterned MR elastomers are also investigated. Lastly, the chapter presents the dynamic behavior of 3D printed MR elastomer under uniaxial deformation in the absence and presence of a magnetic field through a forced vibration technique.

Chapter 6 delineates the development of a new type of MRE-based vibration isolation. The isolator has been developed by implementing the preload and magnetic field simultaneously. This chapter provides a new method to develop MRE-based isolator where the behavior of isolator is enhanced by applying a preload together with a magnetic field.

Chapter 7 summarizes the contents of all the chapters together with concluding remarks and also provides the suggestion for the potential future works.

Chapter 2: Literature Review

This chapter provides a brief overview of magnetorheological materials in the beginning and thereafter focuses on the progress of MR elastomers with the historical overview, materials used, synthesis process, magneto-mechanical characterization and current applications of MREs.

2.1 Overview of Magnetorheological Materials

Magnetorheological (MR) materials consist of magnetic particles loaded in a non-magnetic matrix. The magnetic interaction between particles in these composites can provide various interesting magneto-mechanical phenomena. Such phenomena are magnetic field dependent and can be regulated rapidly and reversibly, therefore, MR materials belong to a class of smart materials. The MR materials can be found in different forms and are distinguishable based on the non-magnetic matrix such as MR fluid (MRF), MR elastomer (MRE), MR foam, MR grease (MRG), MR polymer gel (MRPG) and MR plastomer (MRP) [11, 12, 43-45]. Among all the MR materials, MRF is the first and the most popular MR material, and the popularity is attributed to its dramatic and fast changes of rheological properties, easy preparation procedure as well as insensitivity to the contamination. The latter MR materials, MR foam, MRG, MRPG, and MRP can be perceived as new MR materials as the studies on such materials are limited as well as in the early stage. On the other hand, MRE is neither as new as the latter MR materials nor as old as MRF.

Figure 2.1: Publications on MR materials since 1972, Engineering Village © as per 26 Dec 2018, (Compendex database only).

Figure 2.1 shows an overview of the annual research increment of refereed journals articles on MR materials to date, data collected based on web information services as per December 26, 2018, from the Engineering Village© (Compendex database only). Even though new categories of MR materials are emerging, MRF and MRE are found to be the major categories of MR materials, thus, research and development of these two categories have been steadily increasing. Of course, such a huge number of studies is attributed to many advantages of these MR materials.

Table 2.1: Summary of the various properties of MR materials.

Parameters	MRF	MRE	MR foam	MRPG	MRP
Carrier media	Oils	Elastomers/natural rubbers	Silicone oils	Liquid or suspension	Plasticine-like polymer
Magnetic particle types	CIPs, iron, cobalt, nickel	CIPs, Cobalt, Nickel	CIPs	CIP/ferrous particles	Iron/CIPs
Particle size	0.1-10 μm	0-100 μm / even >100 μm	Micron size	Micron-sized	Micron-sized
Additives as carrier media	Surfactant, Thixotropic Mineral Oil, Hydrocarbon Oil, Silicone Oil	Plasticizers/silicone oil	Glycerol, DABCO-33LV, L-568, and water	1-methyl-2-pyrrolidone	-
Additives as magnetic or non-magnetic particles	Particles Coating MNPs, Magnetite, CNTs, Nanowire Particles	MNPs, CNTs, graphite powder	-	-	-
Volume fraction	0.1-0.5	0.1-0.5	0.35	0.3-0.9	-
Response time	< 1 ms	Millisecond	10 ms	Millisecond	Millisecond
Maximum Field	150-250 kA/m	-	-	-	-

MRF-magnetorheological fluid, **MRE**-magnetorheological elastomer, **MRPG**-magnetorheological polymer gels, **MRP**-magnetorheological plastomer, **CIPs**-carbonyl iron powders, **MNPs**- magnetic nanopowders, **CNTs**-carbon nanotubes

As given in Figure 2.1, MR fluid is the most common MR material and it is a suspension of magnetic particles in a non-magnetic carrier medium. The constituents and various properties of MR fluid and other MR materials are summarized in Table 2.1. In 1948, Jacob Rabinow [46] introduced MR materials by developing MR fluids. However, the studies devoted to MR fluids started to flourish in the late 1980s. In the presence of a magnetic field, such MR fluid changes its form to a semi-solid state from a liquid state within a few milliseconds. However, its physical state is immediately recovered upon the removal of a magnetic field. MR fluids are widely known for their reversible, instantaneous and large stress enhancement upon the application of a magnetic field. Therefore, MR fluids have been commercialized and widely used in various applications

including but not limited to the development of dampers, vibration absorbers/isolators, valves, clutches, polishing and even in prosthetic devices. However, problems such as sedimentation of magnetic particles because of the density mismatch between magnetic particles and the carrier medium, leakage of MR fluid and a need of a storage device limit the utilization of MR fluid in various applications. Among these problems, the biggest problem is the sedimentation and it affects the performance of the MR devices as well as the redispersion of magnetic particles in MR fluid. In order to overcome the aforementioned problems of MR fluid, various solid counterparts of MR fluids such as MRE, MR foam, MRPG, and MRP have been proposed. Among them, MREs have attracted the most attention, which can be visualized from Figure 2.1 as well.

MR elastomers (MREs) can be regarded as the solid analog of MR fluid. In 1983, Rigbi and Jilken [47] introduced the MRE, where ferrite particles are loaded to the elastomer. Later in 1996, Jolly et al [48] started to comprehensively study the behavior of MREs. Thereafter, the various properties of MREs have been explored in a considerable amount. This is due to the appealing advantages of MRE over MR fluid, such as no leakage, no issue of sedimentation as well as no requirement of container or storage device. Usually, MRE is a rubber-like solid material which is loaded with magnetic particles and sometimes additives such as surfactants/silicone oil are also added. In MRE, magnetic particles are locked within the matrix materials, therefore, there is no issue of the sedimentation of the magnetic particles. Because of the locked magnetic particles within the matrix materials, distribution of the magnetic particles could be either isotropic or anisotropic. In the presence of a magnetic field, the magnetic interaction between the neighboring particles lead to the change in the properties, usually, MREs are known for changing their viscoelastic modulus. If soft matrix material is used the MRE can also deform along the direction of a magnetic field: the phenomenon can be regarded as magneto-deformation or magneto-striction [49, 50]. Due to the various advantages of MRE over MRF, MRE is also a potential candidate material in the various applications where stiffness changeability is desired such as vibration absorbers, vibration isolators, and sandwich beams [11, 12]. In addition, MRE has also attracted researchers in developing actuators and sensors such as electronic devices, energy converters, pressure sensors [51-54]. However, the major downside of MRE is the small relative MR effect as compared to MR fluid.

Apart from MRF and MRE, there are other types of MR materials such as MR foam, MR grease, MR polymer gels, and MR plastomer. The main aims of the introduction of new MR materials are to address the sedimentation problem of MRF as well as to seek the implementation of MR materials in new applications. In MR grease, magnetic particles are suspended in a highly viscous carrier medium [55]. It has been reported that the magnetic particles do not move in the absence of the magnetic field in MR grease, thus sedimentation is not the problem. In addition, in 2002 Wilson et al [56] proposed another category of MR material known as MR polymer gel (MRPG), where magnetic particles are distributed in a gel. MRPGs are to improve the stability of the system by controlling initial viscosity and the sedimentation of magnetic particles. In 2004, MR foams are reported for the first time and utilized as an acoustic absorber. MR foams are porous structure, and thus, can be used in a field where light weight is desired for example aerospace [57]. In the early 2010s, a very new category of MR materials called MR plastomer (MRP) was introduced by Xu et al [58]. MRP consists of magnetic particles distributed in a plasticine-like polymer matrix [12, 58-60]. MRP also overcomes the sedimentation problem of MRF as well as it has been reported that MRP showed higher MR effect than that of MRE because of the highly mobile magnetic particles [61-63]. However, the MRPs are much softer as compared to MREs and are not desired in applications such as vibration absorbers or isolators. Nonetheless, MRPs could find a new type of application.

Although various types of solid counterparts of MR fluid have been developed and are continuously increasing to overcome the sedimentation issue of MR fluid which might also lead to the new applications, MRE remains the most popular MR materials after MR fluid because of various appealing advantages. MRE offers specific responses categorized as alternation of elastic and viscous properties in a time domain such as the change in stiffness, storage modulus, stress and strain, and complex viscosity as well as the change in electrical resistance and capacitance. Thus, it is worthy to investigate and enhance the properties of an MRE, a solid counterpart of MR fluid. Hereafter, advancement on MR elastomer, based on materials used, synthesis process, characterization methods, potential applications and various efforts to enhance the MR effects are presented.

2.2 A Brief History of MRE

As described in Section 2.1, the MR materials were introduced by Rabinow [46] in 1948, who demonstrated the MR effects from MR fluids. However, MR elastomers do not have a long history. The preliminary research on MRE was piloted after few decades in 1983 by Rigbi and Jilken [47]. After about 20 years, starting from 2002 the interest in MREs' has increased significantly. There are several terms that have been used to acknowledge the solid analog of MR fluid such as magneto-active elastomer/polymer (MAE/ MAP), magneto-rheological visco-elastomer (MRVE), soft magnetic elastomer (SME) [64-68]. These terms have been derived based on the structural appearance and functional behavior of developed materials, but, the fundamental constituents (magnetic particle and non-magnetic matrix) remain the same. For most of the researcher's usage, the term MRE, hereafter, the same name is used in this study. The initial investigation of MREs was on small strain properties, and the change of storage modulus and the natural frequency have been extensively reported [47, 48]. Since 2002, researchers also started exploring the MREs as smart materials by studying magnetostrictive behavior [69]. Shape changing capacity determines the magnetostrictive property. The maximum stretch of 10% for an MRE was demonstrated by Diguel et al [70]. Nonetheless, the shape-changing property is not considered as a major property alike changes in modulus and natural frequency. Therefore, the property-changing functional behavior of MREs has been extensively explored in recent years [11, 12]. The functional behavior of MRE includes the changes of storage modulus, stiffness, natural frequency, damping, as well as magnetostrictive which have been extensively reported. Thus, MRE makes a natural candidate when stiffness or modulus changeability is desired as needed in vibration isolators or absorbers and sandwich beams. On the other hand, MREs have been considered as a potential material in sensing and actuation areas. Thus, resistance and capacitance and other sensing capacities of MREs are also studied [71, 72]. Starting from 2013 and later, studies devoted to fatigue behavior of MREs can be found in the literature [73, 74]. In 2013, Krolewicz et al [75] studied the fatigue behavior in a shear mode, while Zhou et al [73] studied in an equi-biaxial tension mode.

Similarly, the magnetic permeability is very different when an MRE structure is isotropic or anisotropic. A very first magnetization curve for MREs was reported by Boczkowska and Awietjan [76] in 2012. The magnetic permeability of isotropic and anisotropic

MREs was also studied by Kallio in 2005 [77], Zeng et al [78] in 2013 and Schubert and Harrison [79] in 2016. The anisotropic structure has higher magnetic permeability compared to the isotropic structure as the particle interaction is greater along the particle chain. Very recently, in 2018, Bastola et al [80] also investigated both isotropic and anisotropic permeability of MREs by means of closed magnetic circuit analysis. Similarly, in 2019, Tao et al [81] also investigated the magnetic permeability of MREs by simulations and experiments.

In recent years, the addition of magnetic and non-magnetic additives to MREs has been considered to enhance both zero-field properties and MR effects. Different additives include such as the plasticizer, carbon-based additives and magnetic nanoparticles (MNPs) based on iron. Additives are to prevent the agglomeration of magnetic particles and increase the compatibility of the matrix material with the particles, therefore to enhance both zero-field and magnetic field dependent properties. Consideration of additives such as mineral oils, Phthalate esters, and silicone-based natural esters has started from as early as 2003 [82] and been continuously increasing [83-86]. However, carbon black, CNTs and MNPs were studied starting from 2008. For the first time, use of carbon black was reported in 2008 by Chen et al [87], and the use of CNT was reported in 2011 by Li et al [88] by means of utilizing the multiwall CNTs (MCNTs) and it is continuously increasing [86]. Nickel-based nanoparticles and nanochains were used for the first time in 2013 by Landa et al [89], and additionally, use of MNPs based on iron oxides have also been reported recently in 2018 [25], and 2019 [90].

Equally, a plethora of studies dealing with the numerical investigations and modeling of the MREs' properties are also available in the literature. The very first modeling approach was proposed by Jolly et al [91] in 1996, where MREs were modeled in a similar manner as MRF. To date, various types of modeling approaches such as particle interaction-based models, magnetoelastic response, magneto-viscoelastic response, the effect of environmental conditions and fatigue as well as phenomenological models for the response of MRE-based devices/systems are available in the literature, which are well summarized in a review article by Cantera et al [92] in 2017. Such modeling approaches provide an in-depth understanding of MREs' behavior under various loading conditions as well as help to forecast the MRE behavior for long-term use.

To date, a tremendous number of works devoted to both theoretical and experimental investigations have been performed to study various properties of MREs such as a change in storage and loss moduli, change in damping and elastic properties, and change in electrical resistance, capacitance, and magnetic permeability. Yet, there is plenty of room for the research work to be conducted for a better understanding and the enhancement of MRE properties. Details and progress of different features of MREs' research and development are presented in the subsequent sections.

2.3 MRE Materials

The MRE materials composed of non-magnetic elastomer matrix materials loaded with magnetic particles and the additives. The features of the matrix materials and the addition of additives highly influence the appearance of MREs, and thus, MRE can be soft or hard solid as well as porous or other microstructures. MRE can be classified based on the various parameters such as matrix materials, particle types, and distribution of particles [76] as summarized in Table 2.2. Even though MRE can be categorized into various types, the fundamental materials used remain similar.

Table 2.2: Classification of MRE based on matrix materials, particle types and distribution of particles.

Parameter	Matrix Materials	Particles' Magnetic Properties	Distribution of Particles
MRE Type	Solid Matrix	Soft	Isotropic
	Porous Matrix	Hard	Anisotropic

2.3.1 Matrix Materials

Matrix materials used are a non-magnetic component and the selection of matrix materials significantly influences the various properties of MREs such as hardness, porosity, initial modulus, magnetic field dependent modulus, MR effect etc. Various matrix materials have been used to fabricate MREs, such as silicone rubber, vinyl rubber (VR), polyurethane (PU), thermosets/thermoplastics elastomers and natural/synthetic rubber [11, 12, 43, 45, 86, 93, 94].

Silicone rubber is a saturated elastomer and is the most widely used material among others. The main reasons for the silicone rubber to have attracted the highest interest are firstly because silicone rubber is in the liquid state, which allows a homogenous dispersion and easy suspension of magnetic particles during the synthesis process as

well as because low viscosity of silicone rubber resin allows magnetic particles to easily travel to form chains along the magnetic flux direction to produce anisotropic MREs. In addition, such silicone-based matrices can be cured rapidly at elevated temperature as well as room temperature and they are also non-toxic and nonflammable [95, 96].

Blending with magnetic particles can become a difficult task in the case that the silicone rubber resin possesses high initial viscosity or higher concentration of particles is used. Thus, researchers have utilized additives such as silicone oil to improve the fluidity and plasticity of the matrix materials [97-101]. Likewise, other types of additives such as methyl tri-methoxysilane, dimethyl silicone, silane clear, tert-buthyl perbenzoat, toluene-benzoil peroxide, catalyst 60R/Rhone-Poulenc-stearic acid, and acetic initiator have also been considered to improve the zero-field properties and MR effect of MREs [12, 48, 102-105].

Even though silicone-based rubber has been widely used for the fabrication of MREs, it also has downsides. One of the major downsides of silicone-based rubber is poor mechanical performance such as low strength and shorter fatigue life.

Thermosets and thermoplastic have also attracted a number of attention to be used as an MRE matrix [58, 106-122]. Usage of the thermoplastic elastomers has several advantages over thermoset elastomers including the easy and fast manufacturing process, low energy consumption as well as the possibility of waste recycling. Thermoset elastomers achieve their strength via irreversible crosslinking process upon the application of heat and pressure, whereas thermoplastic can be melted by reheating. The most widely used thermoplastic-based elastomer is polyurethane (PU) compared to other types of elastomers owing to its good mechanical stability [111, 122].

Another type of matrix material of MREs includes natural or synthetic rubber [83, 87, 94, 102, 123-129]. Owing to their complexity of manufacturing (need of more equipment), raw materials (solid phase) and resistance to thermal degradation, natural rubber (NR) or synthetic rubber are the least used matrix materials for the fabrication of MREs. However, utilization of such natural or synthetic rubbers does offer better mechanical properties such as higher tensile and rupture strength as well as higher heat resistance compared to other matrices [94]. The crosslinking of natural rubber should be assisted by other supplements/additives such as plasticizers, crosslinking/vulcanizing

agents (such as zinc oxide, stearic acid, sulfonamide, sulfur) [12, 87, 94, 102, 105, 126, 129]. Examples of the synthetic rubbers used to develop MREs are ethylene propylene rubber, acrylonitrile rubber, cis-polyisoprene/polyisoprene, styrene butadiene rubber, and chloroprene rubber [102, 130-137]. These synthetic rubbers also need additives like natural rubbers during the crosslinking process.

2.3.2 Magnetic Particles

Magnetic particles are a magnetic component of MREs and are responsible for the magnetic field dependent properties (i.e. MR effect). Carbonyl iron powders (CIPs) are the most widely used magnetizable particles. Owing to their high magnetic saturation, low remnant, softness, and high magnetic permeability, CIPs are considered to be the best choice to produce MREs [12, 82]. Utilization of other magnetic particles such as Cobalt, Nickel, and Nd-Fe-B have also been considered. Nonetheless, other particles are found to be not as good as CIPs because magnetic remnant value for these particles is higher as compared to that of CIPs.

Most of the CIPs are reported to be spherical in shape and have an average size below 10 μm . The underlying reason for the utilization of smaller particles is that the smaller particle size offers a higher effective area of interfacial friction between magnetic particles and the matrix materials, which leads to a good damping factor. Bigger particles ranging from 10-100 μm or even higher up to 200 μm have also been reported. Bigger particles are found to be more irregular in shape. The influence of magnetic particles size is significant on MREs properties such as modulus and loss factor. In the absence of a magnetic field, modulus of MREs is smaller for larger particle size. In the presence of a magnetic field, an increase in modulus is found to be significant with smaller particles [12].

On the other hand, the distribution of particles in MREs also affects the MR effect. Particles can be distributed in the isotropic or anisotropic manner in MREs, in which anisotropic configuration of magnetic particles leads to a better MR effect when the direction of an applied magnetic field is parallel to the chains of particles than that of isotropic configuration. The detail of the production of isotropic and anisotropic MREs will be presented in Section 2.4. Similarly, the content of particles also affects the MR effect. In the literature, volume fraction or weight fraction has been adopted, where

volume fraction can range from 0 to 60% and correspondingly weight fraction ranges from 0 to 85%. For anisotropic MREs, volume fraction about 30% has been reported to be the best concentration. For isotropic MREs, volume fraction can be as high as critical volume concentration ($\cong 68\%$). Generally, the MR effect is found to increase with increasing concentration of magnetic particles. The increase in particle concentration decreases the amount of matrix material, so that the MRE sample may not be able to hold the toughness of the matrix rubber. Excessive increase in the concentration of magnetic particles can increase the zero-field modulus/stiffness but not the MR effect.

Figure 2.2: Examples of modification of CIPs.

Apart from the CIPs, other particles such as hard NdFeB particles have also been utilized as a magnetic filler in MREs [27, 140, 141]. The particle size of NdFeB particles ranges from 1-100 μm and is irregular in shape. Use of such hard-magnetic filler provided high-damping of MREs as reported by Yu et al [140]. Similarly, the use of iron sand has also been reported in a few studies [142-144]. Such iron sand particles have a size in the range of 45-56 μm . Morphological study of using iron sand provided evidence that the use of iron sand does not interfere with the formation of the magnetic chain during the crosslinking process.

In recent years, modifications of CIPs have attracted considerable interest among researchers. The modification includes a surface coating, use of nano-flakes, and use of flower-like CIPs [138, 139, 145-150], as given in Figure 2.2. Such modifications are generally to improve the interaction between the particles and the matrix material to obtain an effective damping property. Additionally, such modifications also increase the particle dispersion and wettability between particles and matrix materials during

processing. For examples, magnetic particles are coated with poly (tetrafluoro-propyl methacrylate), (3-aminopropyl) triethoxy silane (APTES), silica coated, polyaniline-modified, siloxane-modified [[12](#), [138](#), [145](#), [148](#)].

2.3.3. Additives

The main role of additives in MREs is to enhance the field-dependent properties, fundamentally known as MR effect as well as other sensing capabilities such as resistance and capacitance. Additives can be categorized as non-magnetic and magnetic additives: plasticizer and carbon-based materials are popular non-magnetic additive while chromium-based particles and other magnetic nanoparticles (MNPs) fall under the category of magnetic additives.

The most common additives for matrix materials are plasticizers. Common plasticizers used in MREs include silicone oil, mineral oil, phthalate Esters and silicone/natural-based Esters. Such plasticizers are added into matrix materials in order to augment their flowability, flexibility, and workability, thus, to assist the MRE' materials processing. A plasticizer can be dissolved in the matrix materials such as silicone rubber by means of mechanical mixing or vulcanization. The additives, plasticizers provide ease to glide molecular chains of rubbers by acting as lubricants, thus adhesiveness of the matrix rubber decreases. Therefore, the molecular chains of rubber can easily glide back and forth upon stretching and releasing. Similarly, the plasticizers also soften the matrix materials, which eases the alignment of the magnetic particles along the direction of a magnetic field for the production of anisotropic MREs as well as help to increase the MR effect. In MREs, the most commonly used plasticizer is silicone oil. The addition of silicone oil has been reported ever since the interest in MREs gained its attraction from 2002. For example, the MR effect up to 600% has been reported by the addition of silicone oil up to 20% by weight. Similarly, the addition of esters-based plasticizers such as rosin glycerin, sucrose acetate Isobutyrate (SAIB) has also been added to natural rubber based MREs. The weight percentage of such additives can range up to 18% and the enhancement of MR effect by 10% have been reported by Khairi et al by addition of 7.5% SAIB [[83](#), [86](#), [151](#)].

Carbon-based materials are another common type of additives used in MREs. The significance of the addition of these additives is to enhance the mechanical properties of

matrix materials during processing, provide better mechanical properties after being cured and enhance MR effect as well as provide sensing capabilities. The first type of carbon-based additive is carbon black (CB) and reported by Chen et al. in 2008 [87] for the first time. The addition of 7 wt% CB provided an increase of MR effect to 104% from 88%. Similarly, development of anisotropic MREs has been also reported with the addition of CB (7.4 wt%), where MR effect was enhanced by 21%, likewise, an increment of MR effect by addition of CB has been also reported by Nayak et al [22] and Wang et al [152] in 2015. Percentage of the addition of CB remains below 10% and still be considered as one of the effective additives to enhance MR effect. The CB is known as a helping agent to modify the properties of MRE by providing better bonding with matrix materials and hence reported to be reliable additives to enhance the MR effect of MREs.

Another member-of carbon-based additives is carbon nanotubes (CNTs) because of its advantages such as high aspect ratio, high surface to mass ratio and lightweight. The first use of CNTs was reported by Li et al [88] in 2011. Both isotropic and anisotropic MREs containing 1% multi-wall CNTs by weight were developed. In 2014, by the same authors, an addition of CNTs up to 3.5% have also been reported, where the increase of MR effects up to 70% was reported. In 2016, even addition of 0.1wt% functionalized multi-wall CNTs (COOH-MWCNTs and OHMWCNTs) provided a noticeable increment of MR effect (up to 13.7%) as reported by Aziz et al [23]. The author reported that the CNTs enhanced the zero-field properties as well as the MR effect.

Similarly, the addition of graphite micro-particles and graphene nanoparticles has also been considered in order to study the sensing behavior of MREs [65]. The main role of graphene particles in MREs is to provide the electroconductive property. The electrical resistance of graphene loaded MRE was found to be decreased with increasing magnetic field strength and compression force. Even though graphite could decrease electric resistance of MREs, but excess addition of graphite powder would lower the MR effect as reported by Tian et al [153].

Nano-sized iron oxides and nickel or cobalt-based nanowires are another class of additives and known as magnetic additives. In 2009, Song et al [154] developed Fe and Co nanowire-based MREs for the first time, where they reported that the nanowire-based MREs exhibited higher specific modulus than that of MREs with spherical particles. In

2014, Maslowski fabricated MREs with micro CIPs and nano $\gamma\text{-Fe}_2\text{O}_3$, where the ionic liquid was utilized to homogeneously disperse the nanopowders and reported that the tensile properties were enhanced by the addition of nano-powders. Similarly, nano-flakes Fe particles have been utilized to develop MREs: with the addition of Fe nano-flakes by 6 wt%, the increase in loss factor was 1.56 times higher than that MREs with micron-sized CIPs only [146]. Similarly, in 2018, rod-shaped $\gamma\text{-Fe}_2\text{O}_3$ nanoparticles were added to conventional MREs and the increase in MR effect about 25% was reported [25].

In conclusion, the addition of small amounts of additives provides an improvement in both zero-field and magnetic field dependent properties. The quantity and quality of additives also influence the MR effect, and additionally, such additives are also known to improve the processing ability of the MRE materials during fabrication. The most common additives are plasticizer, carbon black, CNTs and MNPs.

2.4 Synthesis of MRE

Fabrication of MREs is similar to that of common polymer-based materials processing. Figure 2.3 demonstrates the conventional method of MREs fabrication. The most widely used matrix material, silicone rubber is in a liquid state, and thus, MREs can be produced simply by mixing silicone rubber and magnetic particles with the addition of other additives. The mixing is usually performed until completed at room temperature and thereafter the mixture is allowed to cure inside a mold. Similarly, thermoset or thermoplastic polymers can easily be mixed with magnetic particles in a similar manner to that of silicone rubber but at a high temperature ($> T_g$ or T_m). On the other hand, natural rubber which can be found either immiscible polymer blends or in a solid phase has also been used to develop MREs. Generally, such solid raw materials have to be transformed into a liquid or plastic state before mixing. For such cases, double roll mill and extruder have been utilized in order to have better dispersion of magnetic particles and matrix materials.

Figure 2.3: Method for fabrication of both isotropic and anisotropic MREs.

After proper mixing, the mixture is allowed to crosslink or cure or vulcanize in a mold. Therefore, magnetic particles are locked within the polymer network after the mixture is fully cured. The mold defines dimensions such as thickness and width of MRE to be developed. Generally, mixing is performed at room temperature and curing is performed at a higher temperature in order to accelerate the crosslinking process. If the matrix material is of a type of room temperature vulcanizing (RTV), curing can also be performed at room temperature. During the crosslinking process, a magnetic field is also applied to configure magnetic particles in a specific direction. If the magnetic particles are aligned in a specific direction, such MREs are known as anisotropic MREs. Otherwise, if the mixture is cured without applying a magnetic field, isotropic MREs are obtained. In an isotropic MRE, magnetic particles are expected to be uniformly distributed.

In an anisotropic MRE, the chains of magnetic particles or so-called pre-structuring or forming of columnar structures are found to be affected by factors such as the strength of the magnetic field, exposure time, and temperature. Li et al [155] investigated the effect of variation of these parameters for the development of anisotropic MREs. Application of a higher magnetic field would result in the samples with higher initial modulus. The increase in pre-structuring time would help to restrain rubber and columnar structure. When the microstructure of rubber and magnetic particles stops to move, that point of time can be regarded as saturation time. Increased temperature decreases the viscosity of matrix materials thus allowing the easier movement of

magnetic particles to form columnar structures. Similarly, additives such as plasticizer can help to reduce the viscosity of matrix materials to facilitate the formation of columnar structure.

The anisotropic MREs are formed by applying external energy, which is from the magnetic force, the force that makes the chains of magnetic particles aligned parallel to the direction of the applied magnetic field, so in this way, magnetic particles attempt to reach the minimum energy state [114]. Thus, deformation of anisotropic MREs along the direction of particle chains needs additional force to overcome stronger magnetic interaction force between particles compared to that of isotropic MREs where particles are distributed uniformly in the spatial location. Similarly, the magnetic interaction would be higher when the magnetic field is parallel to the chains of anisotropic MREs and that would result in higher relative MR effect [156]. However, the essential requirement of anisotropic MREs production is the need for a magnetic field during the cross-linking process, so a relatively higher power is required to generate higher magnetic flux. Similarly, controlling the arrangement of the magnetic particles within the matrix material is a difficult task as the magnetic particles can only be configured in the direction of magnetic flux. Thus, a unique configuration is tough to obtain, and it has not been reported to date.

On the other hand, isotropic MREs offer some advantages to its counterpart, such as no pre-structuring process, no need of a magnetic field during the crosslinking process and uniform absorption property along a different direction. However, the MR effect is lower compared to the anisotropic MREs.

The magnetic particles can be aligned in the different directions such as 0° , 30° , 45° , 60° and 90° as shown in Figure 2.4. Boczkowska and his group [76] studied the effect of aligning particles in different directions and found that the higher MR effect was obtained with the samples when particles are aligned in 60° . In such methods, the mixture is placed in a non-magnetic mold, and the mold is placed in between an electromagnet at a 45° angle or any other angle as shown in Figure 2.4. The magnetic flux lines cause the CIPs to form chains aligning to the magnetic field in the defined path. The MR elastomer will then be left to cure, solidifying and trapping the chains in their respective positions.

Figure 2.4: Synthesis and application of anisotropic MREs with different direction particle alignment. (a) application of a magnetic field in order to align particles in 45° [157]. (b) application of a magnetic field for different types of anisotropic MREs [76].

On the other hand, some efforts for the development of patterned MREs without applying a magnetic field can be found in the literature. First patterned MREs were developed by Zhang et al [37], in 2008, given in Figure 2.5. In their study, two new MREs have been developed namely a lattice and a BCC structure MREs as shown in Figure 2.5. The work was achieved by utilizing bigger particles (avg. diameter $400\ \mu\text{m}$). Similarly, another study [113] can be found where patterning of MREs without application of a magnetic field was performed. Using a low-cost wax-cast molding technique for structuring ultra-soft matrix, agglutinative MRE was developed as shown in Figure 2.6. The process is almost similar to that was presented by Zhang et al [37], who firstly utilized patterned molds to develop various patterned samples. The MRE structures from a few mm to the μm range with highly reproducible results have been reported claiming that such patterned MREs can be used in biomedical engineering and microfluidic applications. The common point of developing patterned MREs without applying a magnetic field is that a pre-defined mold is necessary. Even though such processes avoid the need for the application of a magnetic field, they require additional steps and processes.

Figure 2.5: The synthesis process of patterned MREs without applying a magnetic field. (a) patterned mold (b) synthesis steps (c) lattice structure (d) BCC structure [37].

Similarly, Lee et al [158] fabricated microcantilevers by a molding process using MREs, as shown in Figure 2.7. Cantilever patterns were fabricated on an aluminum specimen by using anodization and photolithography. Even though this fabrication method slightly differs from the conventional method, the basic technique is the molding and magnetic field was also applied.

Figure 2.6: Fabrication of the mold for wax-cast-molding of MREs and developed MREs thereof. (a) the fabrication process, (b) different types of MRE columns on a transparent substrate and (c) top views of different patterned MREs [113].

On the other hand, in 2005, a hybrid MRE was patented by Gordaninejad et al [159] by encasing an MR fluid within the elastomeric casing and further studies were also carried out later in subsequent studies [35, 160]. The proposed system was similar to an MR elastomer. Nonetheless, the CIPs are confined within a void of an elastomer casing and do not need the magnetic field for the alignment during the curing process as with typical MR elastomers. The particles could easily be moved within the encased MR fluid and thus provided higher damping capacity than that of conventional MR elastomers. However, further detailed studies are needed for these kinds of elastomers in terms of viscosity of the encapsulated fluid, the concentration of magnetic particles and volume of encapsulation.

Figure 2.7: Fabrication procedure and an optical image of an MRE microcantilever [158].

To summarize the synthesis process, apart from the conventional method, a few other types of fabrication methods for MREs are available in the literature including manual patterning. Nonetheless, a basic technique is molding. For the synthesis of anisotropic MREs, a magnetic field is always required. However, a precise and accurate configuration of the magnetic particles is difficult to obtain by simply applying a magnetic field during the crosslinking process. Thus, it is worthy to explore a new method that is able to precisely and accurately arrange the magnetic particles or suspension in the desired fashion without applying a magnetic field.

2.5 Mechanical Tests and MR Effects of MRE

There are a number of testing methods available in the literature in order to characterize the viscoelastic and magneto-mechanical properties of MREs. The most widely used test

mode is shear mode and other modes can be uniaxial compression or tension mode as well as biaxial test mode. The test approaches reported are both static and dynamic tests. Universal testing machine/dynamic mechanical analyzer (DMA) equipped with a magnetic device to apply magnetic field or laboratory customized setups are used to investigate the various magneto-mechanical properties of MREs. The MREs are characterized to determine dynamic moduli (storage and loss), and finally the MR effect.

The absolute and relative MR effects are used to report the MR effect of the MREs. If E_0 is modulus at zero fields and E_1 is the modulus when the magnetic field is applied, then absolute MR effect is a simple difference between these two moduli (absolute MR effect = $E_1 - E_0$) and relative MR effect is the relative difference between the moduli (relative MR effect = $(E_1 - E_0) / E_0$). The relative MR effect is also expressed as a percentage.

2.5.1 Uniaxial Compression Test

The compression test is one of the most popular methods to investigate the properties of MREs both in static and dynamic modes. A magnetic device developed using either electromagnets or permanent magnets is equipped with the testing machine. A typical result of static compression is given in Figure 2.8. Figure 2.8(a) shows a result of the stress-strain curves as obtained by Kallio [77], which were obtained at a strain level of 6.5%, by applying a magnetic flux density up to 1 Tesla. Figure 2.8(b) shows results of compressive modulus versus strain at various magnetic flux densities and up to 20% strain as obtained by Gordaninejad et al [161]. Both stress or modulus versus strain at various magnetic flux densities is used to report the compressive behavior of MREs.

Table 2.3 provides a few results of compression testing as summarized based on MRE types, test conditions, and MR effects. As can be seen from Table 2.3, the testing conditions such as particle concentration, magnetic flux density and the direction of magnetic flux are different among different studies. Thus, a direct comparison between different studies is inappropriate. Particle concentrations ranging from 4.45% to 33% by volume can be observed. Similarly, the absolute MR effect reported vary from 38.9 kPa to 5.5 MPa. The highest relative MR effect reported is 223% by Abramchuk et al [162]. In order to obtain the MR effect, modulus at zero magnetic field and final modulus upon the application of a magnetic field are used.

Figure 2.8: Results of uniaxial compression tests performed by (a) by Kallio [77] and (b) by Gordaninejad et al [161]. For MRE types and magnetic field please refer to Table 2.3.

Table 2. 3: Results of a few experimental investigations via uniaxial compression testing on MREs. Summarized based on type, the concentration of MREs and testing conditions with maximum absolute and relative MR effects.

Study	Materials		Strain level	Test	MR effect	
	PVC	Type			Absolute	Relative
Kallio [77]	30 %	Aniso @1000 mT	6.50%	700 mT loading alignment	1.75 MPa	100%
Verga et al [19]	5.45 %	Aniso @400 mT	40%	100 mT loading alignment	32kPa	58%
Abramchuk et al [162]	9.20 %	Iso	30%	230 mT ⊥ loading	38.9 kPa	223%
Boczkowska et al [112]	33%	Aniso @ 100 mT	30%	300 mT	80 kPa	4.50 %
Gudmundsson et al [163]	27%	Aniso	15%	700 mT loading alignment	5.5 MPa	120%
Gordaninejad et al 2012 [161]	23.90 %	Aniso @1000 mT	20%	710 mT loading alignment	550 kPa	73%
Schubert and Harrison [15]	30.00 %	Aniso @400 mT	50%	450 mT loading alignment	3.65 MPa	111%

PVC: particle volume concentration, Iso: Isotropic, Aniso: Anisotropic, ||: parallel, ⊥: normal

2.5.2 Uniaxial Tensile Test

The tensile test setups are very similar to the compression test, the difference is only that the MRE sample is stretched. Typical results of tensile tests are given in Figure 2.9. As given in Figure 2.9, in a tensile test, when a magnetic field is applied, the increase in the stress is much pronounced in the small strain region. MR effects were obtained with the tangent moduli from the stress-strain curve. It has been reported that the MR effect decreases rapidly with increasing strain level in the tensile test.

Figure 2.9: Results of uniaxial tensile test performed by (a) Stepanov et al [164] and (b) Schubert and Harrison [15].

Table 2.4 summarizes a few experimental investigations on MREs via tensile testing. In the literature testing up to 100% strain is also available but MR effect is higher at low strain. This is because the distance between magnetic particles increased when MRE is stretched, so the MR effect decreased. The maximum MR effect of 3000% has been reported by Stepanov et al [164], which was also observed at the low strain level. Such high MR effect is attributed to the soft matrix material.

Table 2.4: Results of a few experimental investigations via uniaxial tensile testing on MREs. Summarized based on type, the concentration of MREs and testing conditions with maximum absolute and relative MR effect.

Study	Materials		Test	MR effect		
	PVC	Type		Magnetic flux and direction	Absolute	Relative
Bellan et al [165]	15%	Aniso @ 250 mT	10.00%	150 mT loading alignment	-	-
Stepanov et al [164]	37%	Iso	60%	335 mT ⊥ loading	400 kPa	3000%
Schubert et al [15]	30%	Aniso @ 400 mT	50%	289 mT loading alignment	12.17 MPa	284%
Mordina et al [166]	10% wt.	Aniso @ 200 mT	1%	1089 mT	9.6 kPa	7%

2.5.3 Simple Shear Test

Shear mode testing is the most popular mode that has been adopted to characterize the MRE properties. The shear test can be of two types; single lap shear or double lap shear. The results are also similar to that of compression/tensile testing, however, strain rate cannot be as high as that of tensile testing (strain level can be higher than 100% in tensile

testing). An example of a shear test result is given in Figure 2.10, performed by Schubert and Harrison [15]. The MR effect was found to be higher at smaller strain level and decreased with increasing strain level.

Figure 2.10: Results of pure shear test performed by Schubert and Harrison [15]. (a) stress-strain results at various and (b) relative MR effect versus strain, MR effect was obtained with the tangent moduli.

Table 2.5: Results of a few experimental investigations via shear testing on MREs. Summary based on type, the concentration of MREs and testing conditions with maximum absolute and relative MR effect.

Study	Materials		Strain level	Test	MR effect	
	PVC	Type			Absolute	Relative
Shen et al [167]	25%	Aniso @ 400 mT	12%	395 mT $\perp$ loading $\parallel$ alignment	327 kPa	64%
Stepanov et al [164]	24%	Aniso @ 400 mT	20%	80 mT $\perp$ loading	150 kPa	750%
Yu and Wang [168]	33%	Aniso	120%	47.9 mT $\parallel$ alignment	1.75 MPa	25%
Zajac et al [120]	35%	Iso	13%	163 mT $\perp$ loading	-	70%
Hu et al [100]	24%	Iso	100%	400 mT	125 kPa	500%
Gordaninejad et al [161]	24%	Aniso @ 1000 mT	15%	700 mT $\perp$ loading $\parallel$ alignment	130 kPa	37%
Schubert and Harrison [15]	20%	Aniso @ 400 mT	25%	290 mT $\parallel$ loading $\parallel$ alignment	2.05 MPa	57%

Table 2.5 summarizes a few investigations on MREs via shear test. Yet again, we can see that the testing conditions and the outcome are not exactly similar. The maximum absolute MR effect 2.05 MPa and 750% relative MR effect have been achieved.

2.5.4 Equi-Biaxial Tests

In 2016, Schubert and Harrison [169] performed the equi-biaxial test for both isotropic and anisotropic MREs with the application of a magnetic field for the first time. A

bespoke test rig was designed to conduct the equi-biaxial test. The magnetic field can be applied in two different directions either parallel or perpendicular to the direction of particle alignment of anisotropic MRE. The relative MR effect up to 74% at 67.5 mT magnetic flux density was reported for anisotropic MRE when the applied magnetic field is parallel to the chains of particle alignment.

On the other hand, Zhou et al [73] have investigated the equi-biaxial tests on MRE without applying a magnetic field using a bubble inflation method. Zhou et al [170, 171] also studied the fatigue behavior of MREs in the same mode. It was reported that the fatigue life decreased with increasing strain amplitude and the modulus decreased with the increasing number of cycles. Fatigue behavior of MREs was also studied by Gorman et al [172, 173].

2.5.5 Dynamic Test

The dynamic tests include compression/squeeze mode and shear mode. The dynamic test aims to study the viscoelastic behavior of MRE as related to excitation frequency, strain amplitude, and magnetic field strength. Researchers have adopted two ways for dynamic characterization of MRE: one group of people study MRE samples using rheometer or DMA only while another group of people studies the dynamic behavior of MRE via the development of vibration isolators or absorbers. In the second approach, force vibration testing is adopted to study the dynamic behavior of MREs. In vibration testing, a single degree of freedom system is developed where the MRE sample acts as a spring element. Accelerometers are the main sensor in vibration testing.

The linear viscoelastic model can describe the behavior of MR elastomers in the viscoelastic region with a small strain amplitude. When the viscoelastic material is subjected to a sinusoidal loading, the strain is either in the lagging phase or leading phase with the stress. The applied instantaneous stress can be expressed as a sinusoidal function of maximum stress amplitude σ_o :

$$\sigma = \sigma_o \sin(\omega t + \delta) = \sigma_o \sin \omega t \cos \delta + \sigma_o \cos \omega t \sin \delta \quad (2.1)$$

where ω is the angular frequency, t is time and, δ is a phase angle between strain and stress, Equation 2.1 can be expressed as:

$$\sigma = \gamma_o (G' \sin \omega t + G'' \cos \omega t) \quad (2.2)$$

where, γ_0 is the maximum strain amplitude, G' is the storage modulus and G'' is the loss modulus. The storage and loss modulus represent the ability material to store and to dissipate the energy of distortion respectively. These two moduli are express as a complex quantity. The complex modulus G^* of material and loss modulus of the viscoelastic material are expressed as follows:

$$G^* = G' + iG'' \quad (2.3)$$

$$\tan \delta = \frac{G''}{G'} \quad (2.4)$$

where $\tan \delta$ is a loss tangent angle or loss factor. In vibration damping, these two parameters loss modulus and loss factor are used to describe the damping capacity of the material.

Figure 2. 11: Dynamic force-displacement loops with increasing current to the electromagnet, at 1 Hz of sinusoidal frequency, performed by Kallio et al [93].

An example of dynamic compression testing on MRE is given in Figure 2.11. Force-displacement loops with an increasing magnetic field can be recorded as shown in Figure 2.11, as performed by Kallio et al [93]. Such hysteric cycles can also be expressed as stress-strain loops. The hysteric loops provide the viscoelastic information of MREs such as storage modulus, loss modulus or loss factor.

Figure 2.12: Results of dynamic shear test performed by Hu et al [100]. (a) the storage modulus and (b) loss modulus versus strain at various magnetic flux density.

In the shear mode, the stress-strain or force-displacement loops also look similar as in Figure 2.11. An example of storage and loss moduli obtained for MRE in a shear mode is given in Figure 2.12, performed by Hu et al [100]. However, these results are obtained from vibration testing. In the dynamic test, the general trend is that both storage modulus and loss modulus increase with increasing magnetic field strength and excitation frequency while decreased with increasing strain level.

2.5.6 Summary of Mechanical Testing and MR Effects

There are a number of testing methods adopted to characterize MRE's properties, such as uniaxial compression, tension and shear and multi-axial testing in the absence and presence of a magnetic field. Both static and dynamic measurements have been found in the literature.

The type of MRE materials (matrix, CIP concentration, isotropic/anisotropic) and the testing conditions (mode, strain level, a way of application and strength of magnetic field) and even data analysis methods differ in each investigation. Thus, a direct comparison is not reliable, and is a difficult task, though a certain trend can be developed.

The following trends are observed in the experimental studies on MREs:

- MR effects increase with increasing concentration of magnetic particles (i.e. CIPs).

- Softer matrix materials (with low no-field moduli) lead to higher relative MR effects, but not necessarily the larger absolute MR effects.
- Addition of small quantities of additives such as CNTs and MNPs also increases the MR effect.
- Anisotropic MREs shows higher MR effects than that of isotropic MREs. The MR effects are the highest when loading and magnetic flux direction are parallel to the chains of particles alignment.
- MRE material saturates above 700 mT magnetic induction and MR effects do not increase with increasing magnetic flux.
- Shear mode and compression mode are the most popular testing modes. The compression test reveals lower MR effects than other deformation modes.
- For static tests, MR effects are observed to be higher at a lower strain level.
- Dynamic modulus increases with increasing magnetic field strength and frequency. However, a concrete conclusion is difficult to be drawn regarding the applied strain level.
- Finally, the MR effects shown by the MREs remain lower compared to that of MR fluids.

2.6 Current Applications of MRE

Before considering the potential applications of MR elastomers, the working modes of MR elastomers must be familiarized. The general working modes for MR elastomers can be categorized as squeeze/compression mode, tension mode, shear mode and field active mode. They are illustrated in Figure 2.13.

Understanding the working modes of an MRE is a key step to successfully design the MRE-based devices. The direction of force determines the working mode of MR elastomers and the magnetic field can be applied in various directions in the same working mode. The most popular working modes for MREs are squeeze and shear mode from the application point of view. The shear mode can be a double lap or single lap shear mode and even rotational shear mode. Tension mode is mostly used to understand the mechanical properties of MREs and limited to the laboratories test only. The field active mode is generally perceived as the lowest interest because the stretching or deformation of MRE purely under the magnetic field is very small.

Figure 2.13: Basic working/operation modes of MREs. F: Force and H: magnetic field strength.

General applications of MR elastomers can be found in the following fields: adaptively tuned vibration absorbers, vibration isolators, dampers, shock absorbers for vehicles, sandwich beams and force sensors. All MR devices either work in one of the above modes or the combination of them. When compared to MR fluid devices, MR elastomer devices work in a pre-yield region while MR fluid devices work in a post-yield region. Similarly, MR fluid devices only work in single degree-of-freedom (DOFs) while MR elastomer devices can operate in multiple DOFs. The following section will give the details of MREs applications.

2.6.1 Vibration Absorbers

Vibration absorbers are also known as tuned vibration absorbers (TVAs) and the main function of such absorbers is to attenuate the undesired vibration of a primary structure for example vibration from motors, pumps, and engines. There are two types of TVAs; passive and active or adaptive (ATVAs). The passive TVAs can only work for designed frequency whereas adaptive TVAs are known for adjusting the designed frequency accordingly. Being smart materials, MRE offers a high potential for the development of adaptive TVAs.

A suspension bushing, as illustrated in Figure 2.14, was developed by Watson [174] and Ginder [175] who pioneered the application of MREs. The smart behavior of MREs is practically applied to automotive suspension, where MRE works in a shear mode. The maximum stiffness denoted by the spring constant and the damping capacity can be increased by 25% and 40% in the axial direction respectively with a very short response time of 10 milliseconds. Further, a TVA was also developed with an MRE as a tunable

spring element. The TVA developed by Ginder et al [175], is a single DOF system. The TVA can shift the natural frequency by 22% by increasing to 610 Hz from 500 Hz at the maximum achievable magnetic field. An adaptive tunable vibration absorber (ATVA) was later developed by Deng et al [176] in 2006. ATVA also can tune the storage modulus as well as shift the natural frequency.

Figure 2.14: Automotive suspension bushing (flux path in red) [175].

The first ATVA can increase the storage modulus by 130% while natural frequency could be shifted by 45% at 0.9 Tesla magnetic field strength. Later, Zhang and Li [177] developed a modified ATVA. The modified vibration absorber can change its natural frequency to 90 Hz from 35 Hz at the maximum current of 3 A.

Figure 2.15: Tunable MRE spring developed by Kallio et al [93].

In 2007, Kallio et al [93] developed a tunable spring element, where MRE worked under a squeeze mode and it allowed compressive loading to be applied to it, as shown in Figure 2.15. The tunable spring element application was successfully demonstrated by adjusting compressive modulus via an externally applied magnetic field strength.

Figure 2.16: TVAs proposed by Lerner and Cunefare: (a) shear device, (b) longitudinal device and (c) squeeze device (arrow indicates excitation) [178].

In 2007, Lerner and Cunefare [178] also proposed different kinds of MREs based vibration absorbers as shown in Figure 2.16. Three different kinds of absorbers, i.e. a shear device, a longitudinal device, and a squeeze model device were proposed. A C-shaped magnetic circuit was designed for magnetic flux generation. Those devices working in different modes have different tuning capacities. The anisotropic MR elastomer with 35% volume iron particles was tested. The maximum of 507% increase in natural frequency was observed with a squeeze mode device at the magnetic field strength of 183 kAm^{-1} [178].

Similarly, a squeeze mode MRE-based ATVA has been developed by Sun et al [179] in 2014, as given in Figure 2.17. When a current up to 2.5 A was applied to the ATVA, the natural frequency shifted to 67 Hz from 37 Hz. In 2015, the same group [180] proposed a new type of ATVA featuring layered MRE sheets that are laminated to steel sheets as shown in Figure 2.18. One of the interesting features introduced in such layered MRE-based ATVA is that it can shift the transmissibility curve in both directions (Figure 2.18(b)). When the current was changed from 0 to (-4) A, the natural frequency of ATVA shifted left to 3.2 from 13.5 Hz, while the natural frequency shifted right to 19 from 13.5 Hz when the current was changed from 0 to 4 A. They used so-called a hybrid magnetic system to apply a magnetic field by utilizing both electromagnet and permanent magnets. Later, the same group of researchers also designed and verified a hybrid non-linear MRE-based vibration absorber [181, 182]. Those recent studies are devoted to addressing the problems of narrow bandwidth and adaptability of tunable devices. The detailed structure of such non-linear MRE-based absorber can be found in the article [181]. Here, the vibration attenuation performance of that non-linear system is given in Figure 2.19(a).

Figure 2.17: ATVA developed by Sun et al [179]. (a) Squeeze mode MRE absorber and (b) magnitude transmissibility of MRE-based ATVA versus frequency at various amounts of current to the electromagnet.

Figure 2.18: (a) The structure laminated MRE absorber and (b) the transmissibility of the laminated MRE absorber versus frequency under different currents [180].

The same group, Sun et al [183] has also developed another type of ATVA which possess double natural frequencies. The similar multilayer laminated structured MRE-based isolator was used as given in Figure 2.18, but an eccentric mass was attached on top of the multilayered MRE isolator. Therefore, the ATVA has two natural frequencies, one in the torsional direction and the other in the translational direction as shown in Figure 2.19(b). Two peaks on the transmissibility curve can be seen. The first peak of the transmissibility curve denotes the torsional natural frequency while the second denotes the translational natural frequency. Both natural frequencies increased with increasing current and the relative maximum change of 126% was observed for the torsional natural frequency.

Figure 2.19: (a) Vibration attenuation behavior of non-linear MRE ATVA as developed by Sun et al [181]. (b) Transmissibility of double natural frequencies ATVA [183].

Similarly, the development of ATVAs has been performed by various researchers [184, 185]. Self-sensing property of an MRE-based ATVAs has also been proposed by Komatsuzaki et al. Another graphite based MRE has been utilized as a sensing element embedded within a host MRE of ATVA. The natural frequency can be tuned automatically according to the self-detected signal of graphite based MRE without using an external displacement sensor. Sun et al. [186] have also developed the self-sensing MRE ATVA by modifying the laminated MRE ATVA, and they also did not utilize the external sensor. A self-sensing coil was used to automatically tune the vibration absorption capability of the ATVA.

Even though MRE devices work in different modes or combination of them, the core components in the vibration absorber are the base mass, absorber mass and MR elastomer, and the magnetic field (either generated by permanent magnets or electromagnets). In recent years, the improvement of MRE-based system can be seen in self-sensing and non-linear systems.

2.6.2 Vibration Isolators

MRE-based vibration isolators are installed between the vibration source and the component to be isolated. They can be either a base isolator or a force isolator as shown in Figure 2.20. However, the majority of MRE-based isolators are base isolators. Vibration isolators can also be passive and active like TVAs. Again, MRE is used for the development of active or semi-active isolators. Such isolator can work in both lateral and vertical directions.

Figure 2. 20: Base and force vibration isolation systems.

Hitchcock et al [159] patented in 2006 a tunable isolation device, which is composed of an MRE sandwich structure with two magnetic activation layers. The proposed device was designed as either a curved or flat beam or in the form of a plate as shown in Figure 2.21. In the curved beam configuration, the MRE works in a compression-shear mode.

Figure 2.21: Tunable vibration isolation device proposed by Hitchcock et al [159], a figure showing MRE sandwich structure, details of device and configuration in use respectively from left to right.

Similarly, Liao et al [187] designed a tunable stiffness and damping isolator. Three coils were used to generate a magnetic flux density of four layers of MR elastomers. Kavlicoglu et al [188] proposed a similar design as shown in Figure 2.23(a). The MR elastomer was used in a squeeze mode as a spring element and electromagnet generates the magnetic field. The change of compression static stiffness of the MRE element can be up to 90%. A seat vibration isolator was also designed using MR elastomer by Li et al [101] and Du et al [98] as shown in Figure 2.22(b). An integrated seat suspension that includes both a quarter-car suspension and a seat suspension was successfully fabricated with MR elastomer as a tuning element.

Figure 2.22: (a) MR elastomer developed by Kavlicoglu et al [188] and (b) MRE based seat isolation system by Li et al [101].

There are a number of MRE-based isolators working either in a shear or squeeze mode only. For example, in 2014, Yang et al [189] developed an isolator where MRE works in a shear mode. Likewise, in 2015 Li et al [190] developed a squeeze mode base isolator. In 2017, Mikhailov et al [14] developed an active damper that can be used as an actuator of micro-or nanopositioning for a microisolator object. Similarly, Du et al [191] developed a semi-active integrated vibration isolator by integrating MRE in a shear mode and a spring element as shown in Figure 2.23. They used on-off control and fuzzy logic control laws for the semi-active regulation of the isolator. The resonance frequency of integrated isolator shifted to 51 Hz from 30 Hz when 2 A current was supplied. Similarly, control law can eliminate the resonance peak to achieve effect isolation over a wide range of frequency as shown in Figure 2.23(c). Among two control laws, fuzzy logic control is recommended by authors owing to its superior performance in terms of acceleration reduction and energy consumption.

Figure 2.23: The structure of the semi-active integrated isolator developed by Du et al [191]. (a) prototype and (b) a schematic diagram and (c) experimental result of on-off control.

Starting from 2015, developments of MRE-based isolators working in a shear-squeeze/compression mixed mode have also been investigated [192-194]. Fundamental of such mixed mode isolator is that one MRE sample operates in a squeeze mode and other MRE sample operates in a shear mode as shown in Figure 2.24. The first mixed mode MRE isolator shifted the natural frequency by 103% when 1.5 A current was applied. Till date, the mixed mode MRE isolators have a similar design but shear MRE can be placed in some angle as reported by Leng et al [194], see Figure 2.24(b).

Mixed mode MRE isolator developed by Yang et al [192] in 2015.

Mixed mode MRE isolator developed by Leng et al [194] in 2018.

Figure 2.24: Shear-squeeze mixed mode MRE-based isolators.

Previously, only MR elastomer was used to develop tunable devices. In 2015, Zhiwei et al [195] developed a novel type of active vibration isolator. The schematic is shown in Figure 2.25. The MR device consists of both MR fluid and MR elastomer as the smart isolator element. MR elastomer is working in a shear mode while MR fluid is in a squeeze mode as shown in Figure 2.25(b). The hybrid isolator was able to increase the stiffness by 129.3%.

Figure 2.25: (a) A hybrid magnetorheological elastomer-fluid (MRE-F) isolation mount developed by Zing et al [195]. and (b) Motion state diagram for proposed MR isolator.

Similarly, MRE-based isolators have attracted a considerable amount of attention in the field of civil engineering, for example, isolation of earthquake motion. In 2011, Jung et al [196] started to explore the application of MREs in the field of civil engineering via experimental study, where they placed an MR elastomer on the bottom of a small-scaled single floor structure to study the isolation behavior from the artificial earthquake of a sinusoidal excitation.

Figure 2.26: Study of the isolation behavior of scaled three-story building developed by Behrooz et al [197]. (a) Experimental setup and (b) maximum acceleration of the scaled building.

Behrooz et al [197, 198] also investigated the base isolation. When current up 4 A is supplied to the isolator device, the MR effect of 57% was observed. Later, using the same isolator, Behrooz and group [197] studied the isolation behavior for a scaled building system as shown in Figure 2.26. Semi-active seismic control of a three-story structure was studied using a Lyapunov-based control strategy. The main function of a controller is to have feedback to achieve the maximum reduction in acceleration and displacement of the scaled building. The maximum acceleration reduction of the top

floor was 35.8% with the isolator. The reduction of acceleration was enhanced by the Lyapunov control and was reported that maximum acceleration reduction of the top floor was 47.3%. The results are given in Figure 2.26(b).

Li et al [199] developed a laminated base isolator for the first time with 47 layers of MR elastomer sheets, where MRE sheets were placed between steel sheets. One of the interesting features of such a laminated isolator is the strain-stiffening behavior, which means that the stiffness was increased with increasing shear deformation in a single force-displacement loop. In 2014, the same group further studied a laminated MRE isolator by incorporating a hybrid magnetic system, which has the ability to shift the natural frequency in both directions, as the ATVA presented in Figure 2.18.

Similar to Behrooz et al [197], Sun et al [200] also developed an MRE-isolator for the protection of a multi-story building from the seismic events. The previously developed multi-laminated MRE isolator was used to study the isolation of a scaled building. The results are also similar to that obtained by Behrooz et al [197] but here the fuzzy logic control was implemented. It is reported that the natural frequency was shifted from 3.1 to 7.1 Hz when the current was changed from 0 to 2.5 A.

2.6.3. Sandwich Structures

Adaptive sandwich structures are another type of application where MRE is a potential candidate material. In 2003, Zhou et al [201-203] piloted the use of MRE in sandwich beams. The MRE is used as either only part of a core of the sandwich beam (Figure 2.27(a)) or the entire core (Figure 2.27(b)). Usually, MRE is sandwiched between aluminum or steel plates.

Figure 2.27: Sandwich beams featuring MRE as (a) a part of core [204] (b) an entire core [100].

Dyniewicz et al [205], studied the behavior of a sandwich beam by placing the MR core at the end of the beam. In other words, an MRE-based cantilever was developed.

Similarly, Aguib et al [206], also developed a cantilever MRE sandwich beam and the behavior was studied both theoretically and experimentally.

In 2016, a new type of MRE sandwich structure was developed, where MRE layer was constrained by carbon-fiber-reinforced plastic (CERP) laminates [207]. The used resin was in powder form and the curing process was carried out during pressing with MRE. The reduction in vibration amplitude MRE/CERP sandwich structure arises mainly from the MRE component not from CERP. Furthermore, theoretical and experimental studies devoted vibration absorption or reduction of the behavior of MRE featured sandwich structures can be found in the articles [208, 209].

2.6.4 Sensing and Other MRE-based Devices

For an anisotropic MRE, the spacing between magnetic particles in the chains can be changed by the application of an external force or a magnetic field, which would lead to changes of other several properties in addition to the change in elastic and damping properties. Other properties include a change in shape and changes in thermal and electrical properties. Thus, MRE is also a potential candidate material for the development of sensing devices.

Studies devoted to the electrical properties of MREs have concluded that the resistance change is dependent on the magnetic field and compression force [210-213]. Additionally, change in resistance is more pronounced for anisotropic MRE than that of isotropic MRE [210, 211]. Addition of graphene nanoparticles to MREs can also be found from the study of the electrical properties of MREs by Bica et al [212, 214]. The electrical conductivity of graphene loaded MRE was found to increase with an increasing magnetic field and compression force while it was constant with time. Similarly, investigations devoted to the electrical properties of MREs are available in other studies [215, 216].

Ge et al [217] coated porous MRE with CNT to develop a displacement sensor. The resistance of MRE was measured under different forces and strains. The resistance value was found to be stable with cyclic loading. In 2017, Wang et al [218] investigated the fatigue dependent electrical properties of MREs. Generally, the resistance of MREs was found to be decreased with an increasing number of loading cycles.

Even though a number of experimental studies devoted to the investigation of electrical properties of MREs are available in the literature, development of MRE-based sensing devices by exploiting electrical properties are limited. In 2009, Li et al designed the first MRE based sensing device, a force sensor [219] that can identify the normal force. In 2011, Bica et al [220] developed a magneto-resistor sensor. Similarly, in 2012, Du et al [221] also developed an MRE-based MEMS magnetometer. In 2018, Qi et al [222] also developed an MRE thin film based highly sensitive self-powered tribo-sensor for magnetic field measurement. The highest sensitivity was reached at 16 mV mT^{-1} and the response time was just 20 milliseconds while the rest time is 30 milliseconds. Such type of sensor offers advantages such as no power requirement, low cost, simple fabrication, rapid response, and high sensitivity.

A tactile sensor is another type of sensing device that has drawn attention in the field of MRE-based sensing devices. In 2018, Kawasetsu et al [223] developed a flexible tactile sensor using an MRE, which can detect an applied normal force and vertical deformation. The tactile sensor has a bipolar spatial response like a Mexican-hat as shown in Figure 2.28. The same group also developed the tri-axis tactile MRE-based sensor, which can measure tri-axis force based on the change in inductance [224].

Figure 2.28: (a) The basic structure and (b) sensor output of the proposed flexible tactile sensor developed by Kawasetsu et al [223].

Soft actuators have also been developed using MRE. Soft actuators can be developed by exploiting the magneto-deformation behavior of MREs. In 2012, Bose et al [225] developed MRE-based valve with visible actuation as shown in Figure 2.29. Similarly, Kashima et al [104] also developed a soft actuator where MRE deformed under a magnetic field. The actuator is contracted upon the application of a magnetic field and released to recover its original shape after removal of the magnetic field.

Figure 2.29: MRE-based valve developed by Bose et al [225]. A visible actuation of an MRE can in a valve with an inner air gap(right): top view onto the ring-shaped MRE body in the open (top) and closed state (bottom).

In 2014, Lee et al [158] developed MRE featured micro-cantilevers, where modulus was significantly changed by application of a magnetic field and thus the authors claimed that it opens a door for the development of modulus-tunable force sensors. Behrooz et al [226, 227] developed a flexible micro fluid transport system based on MRE, as given in Figure 2.30. They have successfully modeled the system and demonstrated the propulsion of fluid through a microchannel. It was demonstrated that the maximum volume flow rate can be increased by 16 folds with a larger channel diameter.

Figure 2.30: 2D representation of an MRE featured micro-fluid transport system. SMREM: soft MRE membrane [226].

Recently, in 2018, in another study by Tang et al [228] MRE has been used as a micro-incubator for the pumping and mixing of liquids in the versatile microfluidic platforms. Achieving high pumping or mixing performance in microchannels with excellent controllability have been successfully demonstrated by the group.

The use of MREs in new fields has been continuously being explored and some new kinds of MRE featured applications include roller friction control [229] and adaptive magnetoelastic metamaterials (MMs) [230]. The rolling friction coefficient was found

to be decreased with the application of a magnetic field but can be regulated by changing the strength of the magnetic field. Thus, in the presence of a magnetic field, the optimal friction coefficient to maximum use of friction can be achieved to save friction energy and control the slip rate of a tire is possible. The new concept of MMs opens a door for a new method to modulate MRE's properties by balancing the coupled influences of the internal void architecture of metamaterials, pre-strain, and applied magnetic fields.

As the literature shows, MRE is a very competitive candidate material in a number of applications such as vibration isolators, vibration absorbers, sandwich beams, sensing devices, and actuators. Yet MRE potential has not been fully explored as many new developments are on the way. Nevertheless, the opportunities provided by the MRE for the current applications need to be optimized to fully take advantage of MRE materials. Following conclusions can be drawn based on the current application of MRE materials.

- Shifting of fundamental frequency from the resonance zone is a common practice in MRE-based vibration isolators/absorbers
- Shear and squeeze modes are the most common mode of MRE operation but the development of MRE-based devices in a shear-squeeze mixed mode is very limited
- Squeeze working mode can bear large loading capacity. However, the range of field dependent properties and frequency shift range are smaller than those in a shear mode
- Active or semi-active systems have been developed by implementing feedback systems. However, the implementation of a feedback system is challenging because the MRE behavior is complex
- The working range of the MRE based devices is limited by a narrow frequency range, higher power requirement, and bulky configuration
- If soft matrix materials are used the MR effect for MRE based devices can be higher
- In recent years, new types of application have been steadily explored such as tactile sensors, actuators to pumping and mixing of fluid in microfluidic devices

2.7 Summary and Research Gap of Literature Review

Figure 2.31 provides a clear picture of the interrelations of the four areas of MREs, based on current research and development. The four areas of MREs are raw materials, synthesis processes, characterization, and MRE-based devices. They are interrelated in many ways. In every arena of MREs, improvement, and optimization are continuously growing. The general remarks of each area can be found in the preceding sections, and here, the research gap and scope of the thesis are summarized.

Figure 2.31: Areas of MREs.

The area of raw materials of MREs is found to be much matured compared to other areas. The silicone-based materials are used as matrix material, CIPs are the most common magnetic particles and the addition of additives can be found in order to enhance the performance and MR effect of MREs. The addition of additives also opens the door for new applications, for example, providing sensing capabilities to be used as a sensor. Most of MRE matrix materials are vulcanizable or cross-linkable at elevated temperature or room temperature. Usually, room temperature curable silicone requires at least 24 hours to be fully cured and thus prolongs the synthesis process.

On the other hand, locked magnetic particles in MR elastomers limit the movement of particles upon the application of a magnetic field and thus the MR effect. If magnetic particles can be mobile like in a MR fluid the MR effect would be enhanced. Therefore, investigation of a bridge or connecting materials between MR fluid and MR elastomer with mobile magnetic particles within the matrix materials is worthy of research. Such bridge materials could exhibit anisotropic MR effect as well as avoid the need of a magnetic field during the synthesis process. At the same time, such materials should exhibit a higher MR effect than that of conventional MR elastomer.

The arena of synthesis is found to be concentrated only on the traditional method called molding. One of the common and great requirements is the need for a magnetic field during the crosslinking process for the production of anisotropic MREs. The need for a magnetic field during the crosslinking process simply increases the time and power. Nevertheless, the accurate control and allocation of magnetic particles in the desired fashion or a unique configuration is not achievable by only applying a magnetic field. Therefore, exploring a new method to develop various configurations of magnetic particles within the matrix material without applying a magnetic field is worthy of investigation.

In the characterization arena, the type of MRE materials (matrix, CIP concentration, isotropic/anisotropic) and the testing conditions (mode, strain level, the way of application and strength of magnetic field) and even data analysis methods may differ in each investigation. Additionally, the absence of a standard method for reporting a magnetic field makes the largest difficulty to replicate previously published MRE test results. Therefore, there is a need for making standard protocols for testing of MRE properties both under static and dynamic conditions in the absence and presence of a magnetic field.

In the arena of development of MRE-based devices, use of MREs in the field of vibration absorbers and isolators has been explored extensively. At the same time use of MRE in various new areas has been continuously increasing. Nevertheless, a few common downsides of MRE-devices can be illustrated as follows: the working range of the MRE-based devices is limited by narrow frequency range, higher power requirement, and bulky configuration. Low power consumption may be achieved by reducing the size of a magnetic device or electromagnet leading to a compact system. Usually, MR effects reach a plateau when CIPs are magnetically saturated, which is achieved at higher magnetic flux densities (>700 mT). To supply such a higher magnetic field strength a higher power is needed. As MRE is a viscoelastic material the preloading should change the MRE properties. However, implementation of a preloading to design MRE based devices is not available in the literature.

Chapter 3: Materials and Methods

The main aim of the work is to investigate various kinds of hybrid MR elastomers that are developed via the conventional fabrication technique and a 3D printing technique for the first time. The 3D printing of MR elastomer is new in the field of MRE. Thus, printed MRE may differ from conventional MRE. The controlled volume of MR fluid material is printed and confined within the elastomer. Depending on the type of MR fluid to be printed, the final cured elastomer may contain an MR fluid or a solid MR elastomer within the matrix material. Different hybrid elastomers have been developed using both 3D printing and conventional methods. The magnetorheological and magneto-mechanical properties of hybrid elastomers are measured using rheology, cyclic compression loading and through a forced vibration technique without and with the application of a magnetic field.

3.1 Materials

Materials section can be divided into two subsections: matrix/carrier fluid materials and magnetic particles. The details of the materials used are described in the following subsections.

3.1.1 Matrix and Carrier Fluid

Most of the previously used matrix materials are heat curable either at room or elevated temperature. As mentioned in the literature, silicone rubber is the most widely used matrix material. Here, the focus is to introduce a 3D printing technique to fabricate MR elastomers. Thus, the selection of materials for 3D printing must be performed by considering the specification of the 3D printer including rheological properties and curing nature of the material. A UV curable silicone-based resin would be the best choice because of the fast curing nature that facilitates continuous printing. Therefore, UV curable silicone (SS-155 UV) is used for making an elastomer matrix. This resin has a low viscosity and post-cured hardness of 25 Shore A. Vulcanizing silicone (SS-3006T) and non-vulcanizing modified Lord MR fluids (MRF-132DG) were used as printing materials. Carbonyl iron powders were added to SS-3006T to make an MR suspension.

Similarly, silicone oil from Dow Corning Corporation and carbonyl iron powders were added to modify MRF-132DG for the printing.

For the conventional method, soft hybrid MR elastomer samples contain two main components – the elastomer matrix, and the MR core. A UV curable silicone (SS-155 UV) was utilized as an elastomer matrix. The MR core consists of magnetic particles and a magnetic particle carrier medium. Three different magnetic particles carrier medium – a low viscosity silicone oil (Silicone oil AP 1000), high viscosity silicone oil (Dow Corning Corporation 200® fluid, 60,000 cSt) and a room temperature vulcanizing (RTV) silicone rubber (SS-6B) were used.

Basic properties such as viscosity, specific gravity, and appearance, and name of the supplier of matrix and carrier fluid materials used in this study are summarized in Table 3.1.

Table 3.1: Properties of matrix and carrier fluid materials and suppliers, information provided are based on supplier.

Properties	Materials					
	SS-155 UV	SS-3006T	MRF-132DG	Silicone oil AP 1000	Dow Corning Corporation 200® fluid	SS-6B
Viscosity	2.5 Pa.s	40-80 Pa.s	0.112 ± 0.02 Pa.s	0.1 Pa.s	60000 cSt (Kinematic)	2 Pa.s
Specific gravity	1.02 g/cm ³	1.08 g/cm ³	2.95-3.15 g/cm ³	1.09 g/cm ³	0.97 g/cm ³	1.02 g/cm ³
Tensile Strength*	80 PSI	500 PSI	-	-	-	200 PSI
Elongation at break*	-	450%	-	-	-	250%
Hardness*	25 Shore A	20 Shore A	-	-	-	30 Shore A
Appearance	Clear	Translucent	Dark Gray Liquid	Clear	Clear	Clear
Supplier	Silicone Solutions, USA	Silicone Solutions, USA	LORD Corporation USA	Sigma-Aldrich, Singapore	Sigma-Aldrich, Singapore	Silicone Solutions, USA

* it should be noted that these properties are after curing

3.1.2 Magnetic Particles

Magnetic particles were purchased from Sigma-Aldrich, Singapore and they have an average size of 3–5 μm . The magnetic particles are soft carbonyl-iron powders (CIPs) with low magnesium and manganese compounds. Because of their high magnetic permeability and low remnant magnetization with the high magnetic saturation point, CIPs are considered as the best choice for MR elastomers.

Table 3.2: Properties of CIP and MNPs used, information provided are based on supplier.

Property	CIPs	MNPs
Particle size	3-5 μm	<50 nm
Iron content	>99.5 %	68.2 - 71.7 % (Titration by $\text{Na}_2\text{S}_2\text{O}_3$)
Molecular weight	55.85 g/mol	159.69 g/mol
Appearance (color)	Faint gray to very dark gray	Red brown to brown
Appearance (form)	Powder	Nanopowder
Relative Density	7.86 g/cm ³	5.15 g/cm ³

Figure 3.1: SEM images and XRD patterns CIPs and MNPs.

Metal trace analysis shows the high purity of CIPs as characterized by 99.5 % pure iron (Fe). Similarly, magnetic nanoparticles (MNPs) were also added to enhance the viscosity

and MR effect of the hybrid MR elastomer. The properties of CIPs and MNPs used in the study are presented in Table 3.2. The SEM images and XRD patterns of pure CIPs and pure MNPs are shown in Figure 3.1. CIPs used here are the soft magnetic particles and mostly spherical in shape, nanoparticles are clumped together because of high surface area as shown in Figure 3.1.

3.2 Fabrication Methods

Two types of fabrication methods have been adopted in this study. The first one is known as the convention method, where samples were fabricated by molding. Second is a new method, called additive manufacturing or 3D printing, which adopts the layer by layer fabrication technique.

3.2.1 Fabrication of Core-shell Samples

As shown in Figure 3.2, the core-shell hybrid MR elastomer samples were fabricated by impregnating the MR material within the core cavity of the solid elastomer shell. Firstly, a layer of the UV curable resin was dispensed into the mold and cured with UV light. In the second layer, a cavity was formed for the MR fluid. After that, the MR fluid was dispensed into the cavity. Finally, the MR fluid was fully covered with the resin and cured with UV light. The samples were removed from the mold after 24 hours. In the case of solid core samples, a suspension of RTV silicone and CIP was injected into the cavity in the place of the MR fluid. The external diameter of all samples is 28 mm and the thickness of a sample is 7 mm, where the MR core has 3 mm thickness and 20 mm in diameter. The MR core is of 25% volume of the entire sample. The dimensions and sample specification are shown in Figure 3.3.

Figure 3.2: Schematic illustration of the process of soft hybrid MR elastomer development with an MR core within an elastomeric shell.

The MR core has four different concentrations of magnetic particles (20%, 40% 60% and 80% w/w) for three different carrier media (low viscosity fluid core, high viscosity fluid core, and solid core). The solid core was developed by utilizing the RTV silicone rubber as a carrier medium. In order to prepare MR fluid/suspensions in the lab, CIPs and/or MNPs were mixed with silicone oil/RTV silicone thoroughly and stirred for 15 minutes then the mixture was put into a sonicate bath for 20 minutes.

Figure 3.3: (a) Schematic diagram (b) a photograph of the soft hybrid MR sample. (c) and (d) cross-sectional view of the soft hybrid MR sample.

3.2.2 3D Printing

A multi-material 3D printer was required for fabrication of hybrid MR elastomers. In this study, a BioFactory 3D printer made by RegenHU in Switzerland was employed.

The printer is capable of printing various materials including cells, based on different dispensing technologies such as extrusion, inkjet, valve-controlled, temperature controlled, etc. Due to the high viscosity of the MR fluid and the elastomeric resin, the extrusion-based time-pressure printheads were chosen for dispensing these materials. The printer was equipped with a UV/laser-curing unit (365 nm wavelength) that was used to cure the elastomeric resin. Photographs of the printer are shown in Figure 3.4.

The 3D printing process was conducted at room temperature. The various patterns were generated from the 3D software (BioCAD) and the MR fluids suspension was continuously deposited in the predesigned path. As it was the first time for 3D printing to be implemented to develop MR elastomer, problems related to the selection of suitable printing materials and successful printing were inevitable. The details of 3D printing and its various challenges for the fabrication of hybrid MR elastomers are described in **Chapter 5**.

Figure 3.4: Photograph of the 3D printer employed to fabricate hybrid MR elastomers in this study.

3.3 Characterization and Analysis

Firstly, the rheological properties of the fluids have been studied in both the absence and presence of a magnetic field. In order to study the MR effect of hybrid MR elastomer samples, characterization is performed by two methods: (1) cyclic compression and (2) forced vibration testing in both the absence and presence of a magnetic field.

3.3.1 Rheology

For the core-shell hybrid MR elastomer, the rheological properties of both low and high viscosity carrier fluids with four different concentrations of magnetic particles (20%, 40%, 60% and 80% w/w) were evaluated using the rotation rheometer (DHR, TA instrument, USA). A parallel plate (40 mm in diameter) and a measurement gap of 0.55 mm were used for all measurements. The lower plate of the rheometer was modified in order to apply a magnetic field during the experiment. The experimental instrument is shown in Figure 3.5(a). The relationship between magnetic flux density and current to the electromagnet is presented in Figure 3.5(b). The average magnetic flux density at a different amount of current was calculated and is presented in Figure 3.5(c). The experiment was conducted at four different currents to the electromagnet (0, 1, 2, 3 A). To examine the MR characteristic of samples, a steady-state flow test was conducted in the range of 0.1-100 s^{-1} shear rate.

Figure 3.5: (a) Photograph of the rheometer with an electromagnet as the bottom plate, (b) magnetic flux density against the radial distance of the electromagnet at three different current and (c) the average magnetic flux density at different current (0, 1, 2, and 3 A) to the electromagnet.

For 3D printed hybrid MR elastomer, the rheological properties of all printing materials were also evaluated using the same rotation rheometer (DHR, TA instrument, USA). In order to select the proper materials for printing, two types of rheological tests were conducted to evaluate the 3D printability of the materials: (1) steady-state flow test and (2) recovery test. Thereafter, a frequency sweep test over the angular frequency range of 0.01-100 rad/s at a constant strain of 2% was carried out to get the information on storage modulus and loss modulus of the selected printing materials. The detail of the steady-state flow and recovery test are discussed in **Chapter 5**.

3.3.2 Cyclic Compression

The cyclic compression test was conducted with a universal testing machine Instron 5569 with a 500 N load. In addition, an electromagnet was used with a closed magnetic circuit to observe the MR behavior of the MR elastomer samples under varying magnetic flux densities. The experimental photograph is shown in Figure 3.6. The electromagnetic-compression testing set-up has a 9 mm air gap in between the position of the magnetic circuit for loading the sample and the Instron's compression plate.

Figure 3.6: (a) Schematic illustration of the magnetic circuit and (b) a photograph of the experimental setup as connected to a load cell.

The electromagnet consists of 2800 turns of coils with an electric resistance of 10 Ω and, a DC current up to 3.0 A was supplied to the electromagnet. The relationship between

the current supplied to the electromagnet and magnetic flux density in the air gap without placing any samples is presented in Table 3.2. The cyclic compression was conducted at various strain rate from 4.2 mm/min to 42 mm/min and up to 10 % strain and at four different currents (0, 1, 2, and 3 A). The effect of strain rate and strain amplitude will be presented in subsequent chapters.

Table 3.3: Magnetic flux density at different currents applied to the electromagnet.

Current (A)	0	1	2	3
Magnetic flux density (mT)	0	130	280	390

The MRE materials are sensitive to stress softening, a well-known effect in rubber-like materials, which is also called the Mullins effect. The Mullins effect can be explained as the strain softening behavior of an elastomer, indicated by the descending stiffness over the cyclic loading. The Mullins effect has a great influence on large-strain experiments, but it was also observed in the case of low-strain (up to 10% strain) experiments. The influence of the Mullins effect on stress-strain results is demonstrated in Figure 3.6 in both the absence and presence of a magnetic field for core-shell soft hybrid MR elastomer sample. The first loading cycle has the highest stresses and is found to be decreased over increasing cycles and then become stable. Such behavior is observed because the sample experiences a remnant deformation after the first cycle. For hybrid MR elastomer samples, the remnant deformation was more pronounced when the magnetic field was applied, so that the Mullins effect is more pronounced when a magnetic field is applied.

Figure 3.7: Stress-strain results of a three-cycle compression test to 10% strain of core-shell hybrid MR elastomer sample with 80% CIPs. (a) without and (b) with the application of a magnetic field.

In order to compensate for the Mullin's effect [231, 232], all MR samples were conditioned before the actual measurements, to make sure that material characteristics are the same for all measurements. The results of the third cycle were consistently used to characterize the samples.

3.3.2.1 Definition of moduli, Energy Dissipation, and Magnetorheological Effect

To interpret the stress-strain results linear and secant moduli are used. Secant modulus is defined as the instantaneous ratio of stress and strain, while the linear modulus is the slope of the stress-strain curve. All data interpretation are carried out using Engineering stress-strain.

Whenever the materials behave differently under unloading from under loading, the materials are said to be viscoelastic and to exhibit the phenomenon of hysteresis. Such hysteresis is very common for MR elastomers and also found to be exhibited by the hybrid MR elastomers (both conventionally developed and 3D printed) in this study. Therefore, to investigate the hysteresis behavior in both the absence and presence of a magnetic field, the energy dissipated under the loading-unloading cycle is obtained.

Magnetorheological (MR) effects are characterized by comparing the properties without and with the application of a magnetic field. The effect of the magnetic field can be studied by obtaining the absolute and relative MR effect. The absolute MR effect is defined as the change in the modulus or other properties by the application of a magnetic field as:

$$MR_{abs} = E_B - E_{B_0} \quad (3.1)$$

where E_B and E_{B_0} are the moduli or other properties with and without the application of a magnetic field, respectively. Secondly, relative MR effect is defined as the relative change between properties as presented in Equation 3.2.

$$MR_{rel} = \left(\frac{E_B}{E_{B_0}} - 1 \right) \quad (3.2)$$

Relative MR effect can also be expressed as $\left(\frac{E_B}{E_{B_0}} - 1 \right) \times 100\%$ defined as a percentage value.

3.3.3 Forced Vibration Testing

One of the widely used methods to characterize MR elastomer is vibration analysis. From the vibrational analysis, several relationships such as transmissibility vs frequency, phase angle versus frequency, and dynamic stiffness and damping versus frequency at various magnetic flux densities can be studied. As most dominant working modes for MR elastomers are squeeze and shear modes, this study also explores the properties of hybrid MR elastomer in both squeeze and shear modes in both the absence and presence of a magnetic field. In squeeze mode, MR elastomer also vibrates parallelly with the acceleration signals from the shaker. In shear mode, MR elastomer vibrates normal to direction of the acceleration signals from the shakers. The experimental apparatus of forced vibration testing can be seen in Figure 3.8.

Figure 3.8: (a) Experimental setups for forced vibration with base excitation where soft MR elastomer is in (b) squeeze and (c) shear mode operations.

The base of the single degree of freedom system was excited by a shaker (PM Vibration Exciter Type 4808, Brüel & Kjær). Different amplitude and frequency of vibration signals could be generated by a controller and then transferred to the shaker. Two accelerometers (353B15, PCB Piezoelectronics, Inc.) were used to measure the vibration of the base and the absorber mass. The signals from accelerometer were amplified by the charge amplifier and then transferred to the computer. The magnetic field was applied by incorporating the magnetic device (fabricated in Lab) to the experimental setup. The magnetic device can generate various magnetic fields and could easily be integrated into the experimental setup as shown in Figure 3.8.

Figure 3.9: Magnetic device to generate various magnetic fields that can be integrated into a forced vibration testing setup with an MR elastomer and corresponding magnetic flux density produced by the device.

Two solid cylindrical permanent magnets (50 mm diameter x 20 mm thickness) are separated at a certain distance to make a magnetic device. The various magnetic flux densities could easily be generated by changing the distance between two permanent magnets as shown in Figure 3.9. Testing was conducted at four different magnetic flux densities as 0, 110, 300 and 500 mT.

A schematic representation of an ideal system with a single degree of freedom (DOF) is given in Figure 3.10. The essential component of a single DOF system includes effective mass (M) and its elastic properties (flexibility or stiffness (K)) and its energy-loss mechanism or damping (C). For the MRE based systems, stiffness and damping components are not constant and they are magnetic field-dependent.

Figure 3.10: The ideal dynamic system with a single degree of freedom, where MRE can be considered as a viscoelastic spring-damper element.

The natural frequency ($\omega_0 = 2\pi f_0$) of a single DOFs system can simply be calculated as:

$$\omega_0 = \sqrt{\frac{K}{M}} \quad (3.3)$$

where K and M is the stiffness and the mass of the single DOF as shown in Figure 3.10. When the damping element is presented the natural frequency becomes the damped natural frequency (ω_d) and generally damping element has very less fluctuation of the natural frequency as:

$$\omega_d = \omega_0 \sqrt{1 - \xi^2} \quad (3.4)$$

where ξ is the damping ratio and defined as: the ratio of actual damping (C) to critical damping (C_c) value of the material, it is defined as;

$$\xi = \frac{C}{C_c} = \frac{C}{2m\omega_0} \quad (3.5)$$

The magnitude and phase transmissibility of single DOF vibrational system can be expressed as:

$$T = \sqrt{\frac{1 + (2\beta\xi)^2}{(1 - \beta^2)^2 + (2\beta\xi)^2}} \quad (3.6)$$

$$\delta = \tan^{-1} \frac{-2\xi\beta^3}{1 - \beta^2 + (2\beta\xi)^2} \quad (3.7)$$

where ξ is the damping ratio $\beta = \frac{\omega}{\omega_0}$ or $\frac{f}{f_0}$ is the frequency ratio between excitation frequency and the natural frequency of the system.

With the forced vibration technique, the frequency-response curve of the samples can be constructed through a frequency sweep by measuring the excitation and response amplitude of vibration. Typical frequency response of a single degree of freedom is shown in Figure 3.11. In the figure, magnitude transmissibility is obtained as a ratio of the response signal and the excitation signal ($\ddot{y}/\ddot{x}$ in Figure 3.10) and $A_{\max}$ is the maximum displacement at the resonance frequency. As the transmissibility amplitude reaches to the maximum value the phase delay between the base and mass becomes 90° , under this condition the corresponding excitation frequency becomes the natural frequency. Therefore, the stiffness of the system can be obtained using Equation 3.3, similarly, the damping ratio can be obtained using Equation 3.8 as follows:

$$\zeta = \frac{\Delta\omega}{2\omega_0} = \frac{\Delta f}{2f_0} = \frac{f_2 - f_1}{2f_0} \quad (3.8)$$

where $\Delta\omega$ or Δf is the frequency difference when the response amplitude is equal to $A_{\max}/\sqrt{2}$ of the peak response amplitude as illustrated in Figure 3.11.

Figure 3.11: The frequency response curve of a moderately damped single degree of freedom system. For the definition of symbols refer to the text.

In the subsequent sections, first, the natural frequency is obtained based on frequency-response of a single DOF system through a forced vibration testing at various magnetic flux densities (0, 110, 300, and 500 mT). Thereafter, Equation 3.3 and Equation 3.8 are

used to obtain stiffness (K) and damping ratio (ζ) for the single DOF system and finally the MR effects are calculated using Equations 3.1 and 3.2.

For every test, both in cyclic compression and forced vibration test, three samples were considered. The MR effect obtained are expressed as the average value of the three samples.

Chapter 4: Soft Hybrid MREs Developed by Forming a Core-shell Structure

This chapter deals with the development and characterization of core-shell soft hybrid MR elastomers by impregnating the MR materials within the core of the elastomeric shell. Two types of magnetic particle carrier media have been considered namely fluid core and solid core with four different concentrations of CIPs. In the end, the addition of MNPs to enhance the viscosity and MR effects of the fluid core are presented.

4.1 Introduction

As discussed in Section 3.3, the soft hybrid MR elastomers were developed with two types of cores: the fluid core and solid core. The fluid core consists of low viscosity and a high viscosity carrier fluid (silicone oil) for magnetic particles while the solid core consists of RTV silicone rubber loaded with magnetic particles. The viscosity and shear stress of pure low and high viscosity fluid without the addition of any magnetic particles are presented in Figure 4.1. The viscosity of high viscosity fluid is almost 50 times higher than that of low viscosity fluid at the shear rate of the range $0.1\text{--}100\text{ s}^{-1}$. The cores were prepared with four different concentrations of magnetic particles (20%, 40%, 60% and 80% w/w). The image of samples and SEM images of the four different concentrations of CIPs and elastomeric shell are shown in Figure 4.2. SEM images of different concentrations of CIPs were taken with solid core samples.

Figure 4.1: Shear stress and viscosity versus shear rate for low and high viscosity carrier fluids without the addition of any magnetic particles at room temperature (25°C).

Figure 4.2: SEM images of (a) 20%, (b) 40%, (c) 60% and (d) 80% soft hybrid MR elastomers' core and (e) the outside elastomeric matrix. Solid core samples were used for SEM images.

4.2 Rheological Study

Initially, the rheological properties of the low and high viscosity carrier fluid with various concentrations of CIPs were studied without and with the application of a magnetic field. A controlled shear rate mode at different magnetic flux densities ranging from 0 to 63 mT was conducted to investigate the MR characteristic of the flow curve. Figure 4.3 shows the flow curves for different concentrations of magnetic particles for both low and high viscosity carrier fluids with shear stress as a function of shear rate on a log-log scale at various magnetic flux densities. The close and open symbols stand for low and high viscosity carrier fluid respectively, and the symbols below (Figures 4.4 and 4.5) represents the same carrier fluids as in Figure 4.3. In the absence of a magnetic field, the shear stress and shear rate of both types of fluids (regardless of the concentration) seemingly exhibited a linear relationship. However, high viscosity MR

fluid always possessed higher shear stress than that of low viscosity MR fluid regardless of the concentrations and magnetic field. It was also clear that the increased concentration of the magnetic particles increased the shear stress for both low and high viscosity fluids. Upon the application of a magnetic field, both low and high viscosity MR fluid showed a noticeable shear stress enhancement and maintained a stable shear stress value in the entire range of the shear rate ($0.1\text{-}100\text{ s}^{-1}$), which is typical behavior of the Bingham fluid. The stress enhancement is more significant for higher concentrations of magnetic particles for both types of MR fluids. It can be noted that the stress enhancement is more pronounced for low viscosity fluid as the magnetic particles need to overcome less viscous resistance to form a chain along magnetic flux direction as compared to the high viscous fluid. For example, shear stress of the 80% low viscosity MR fluid could reach as high as 80% high viscosity MR fluid at the highest magnetic flux density (63 mT). The similar result can be found when viscosity was plotted against the shear rate as shown in Figure 4.5. This signifies that the stress enhancement is similar for both type of the carrier fluid, however, MR effect is higher for low carrier fluid.

Figure 4.3: Shear stress versus shear rate for low viscosity (closed symbols) and high viscosity carrier fluids (x symbols) containing different concentrations of CIPs (a) 20%, (b) 40%, (c) 60% and 80% at different magnetic flux densities.

In order to describe the rheological properties (field-induced yield stress and viscosity) of MR fluids, one of the widely used constitutive models is a Bingham Plastic model. However, Herschel-Bulkley model and Casson Fluid model are also used in many studies. In Bingham Plastic model the materials flow behavior is assumed as elastic solid up to critical shear stress (yield stress), beyond which the materials start to flow and behave like Newtonian fluids. The constitutive equation for this model is as follows

$$\tau = \tau_y + \eta \dot{\gamma}, \text{ for } \tau > \tau_y \text{ and } \dot{\gamma} = 0 \text{ for } \tau \leq \tau_y \quad (4.1)$$

where $\dot{\gamma}$ is the shear rate, η plastic viscosity, τ_y is the dynamic yield stress and τ is shear stress which is related to the magnetic flux densities.

Table 4.1: Yield stress obtained via curve fitting for different types of MRFs and R^2 value.

MRF	Yield Stress (Pa)				R ² value			
	Current to the electromagnet (A)							
	0	1	2	3	0	1	2	3
20%-low	0.29	33.6	67.95	131.38	0.999	0.866	0.874	0.768
40%-low	0.6	81.34	202.5	431.56	0.999	0.877	0.746	0.577
60%-low	1.722	129.05	359.47	853.92	0.999	0.811	0.729	0.559
80%-low	19.95	315.03	994.98	2499.5	0.999	0.959	0.698	0.644
20%-high	33.92	96.88	212.97	392.68	0.997	0.997	0.995	0.976
40%-high	47.18	195.44	391.83	852.48	0.997	0.996	0.989	0.946
60%-high	92.2	309.35	669.35	1714.1	0.995	0.994	0.986	0.913
80%-high	817.32	1250.9	2158.6	3471.2	0.968	0.966	0.927	0.913

The dynamic yield stress (magnetic field dependent yield stress) for both low and high viscosity carrier fluid obtained from flow curves (Figure 4.3) via curve fitting using Equation 4.1 are plotted as a function of the magnetic flux density (B) and shown in Figure 4.4. The coefficient of determination of curve fitting, denoted by R^2 and values of the yield stress for all four concentrations of two different MRFs at the various amount of current to the electromagnet are given in Table 4.1. It was clearly observed that the high viscosity fluid has higher yield stress than that of low viscosity fluid regardless of the concentration and magnetic field. It can also be noted that the dynamic yield stress also increased with increasing concentration of magnetic particles. When the magnetic field was applied, the dynamic yield stress drastically increased for both types of MR

fluids. The increase in the dynamic yield stress of the low viscosity fluids was more pronounced compared to the high viscosity fluid, which can be visualized from the slope of the curves (Figure 4.4). Again, this can be attributed to the higher viscous resistance of the high viscosity fluid limiting the movement of the magnetic particles when the magnetic field is applied. Therefore, most of the commercial and well-developed MR fluids utilize the low carrier fluid for better MR effect. However, the low viscosity fluids largely suffer from the sedimentation and leakage problem. Thus, in this study, in order to develop soft hybrid MR elastomers using core-shell structure, the high viscosity fluid is used with the aim of minimizing sedimentation and leakage problem even though the MR characteristics are not as good as that of low viscosity fluid.

Figure 4.4: Dynamic yield stress versus magnetic flux density for low (open symbols) and high viscosity (filled symbols) carrier fluids for 20-80% w/w ratio of CIPs.

The shear viscosity for two types of carrier fluids with four different concentrations of CIPs as a function of shear rate at various magnetic flux densities is shown in Figure 4.5. When the magnetic field is not applied, the low viscosity MR fluid is always less viscous than that of the high viscosity MR fluid. However, the viscosity of the low viscosity MR fluid can reach as high as the viscosity of the high viscosity fluid when the magnetic field is applied. Similarly, the viscosity increased with increasing concentrations of magnetic particles regardless of the magnetic field. When the magnetic field is applied, the shear viscosity of both MR fluids shows an obvious increase with increasing magnetic flux densities and apparent shear thinning behavior can be observed easily at each magnetic flux density. Typically, most of the MR fluids show similar kinds of shear thinning behavior in the presence of a magnetic field. Yet again, the shear thinning is more pronounced for the low viscosity carrier fluid.

Figure 4.5: Viscosity versus shear rate of low viscosity (open) and high viscosity (closed) carrier fluid for different concentration of CIPs (a) 20%, (b) 40%, (c) 60% and 80% at different magnetic flux densities.

Sedimentation ratio is defined as

$$\text{Sedimentation ratio} = \frac{\text{Height of the sediment layer}}{\text{Height of total suspension}} \quad (4.2)$$

Sedimentation behavior for two MR fluids was also studied and found that the resistance of the high viscosity fluid to the sedimentation is much higher than that of low viscosity carrier fluid. Sedimentation ratio was obtained using Equation 4.2 and is presented in Figure 4.6. As shown in Figure 4.6, for low viscosity MR fluid, sedimentation was faster and in less than 5 days, the sedimentation ratio reached about 50%, which is undesirable. On the other hand, the high viscosity MR fluid exhibited extremely good resistance to sedimentation. Even after 15 days, the sedimentation ratio was above 90 %.

In conclusion, the magnetorheological study revealed that the high viscosity carrier fluids do not show as good MR effect as the low viscosity carrier fluid. However, the advantage of the high viscosity carrier fluid is that it overcomes the sedimentation problem even at low concentration of magnetic particles (60% w/w) because of the high initial viscosity. And is the key factor to be considered for developing core-shell soft hybrid MR elastomers.

Figure 4.6: Sedimentation ratio versus time for low and high viscosity fluids at 60% concentration of CIPs.

4.3 Cyclic Compression

In order to study the MR effect of the core-shell soft hybrid MR elastomers, a cyclic compression test was conducted under both conditions: in the absence and presence of a magnetic field. Figure 4.7. shows the experimental results of engineering stress-strain hysteresis loops for low viscosity core soft hybrid MR elastomer samples and with increasing magnetic flux density by increasing an applied current up to 3 A. Test were conducted at a constant frequency of 0.1 Hz, and strain amplitude of 10% (the effect of strain rate or frequency and amplitude for the soft hybrid MR elastomers in a cyclic compression will be presented in **Chapter 5**). Here, the goal is to understand the effect of viscosity of carrier fluid, the concentration of magnetic particles and the type of the core for core-shelled hybrid MREs. Thus, an experimental condition is fixed as mentioned above. A number of observations become evident in Figure 4.7. First, the shifting of the stress-strain curve is difficult to observe for 20% and 40% of the concentrations of magnetic particles with an increasing amount of current to the electromagnet. The movement of stress-strain curves is noticeable for 60% and 80% of particles concentrations when a magnetic field is applied. However, shifting of the curves is more pronounced with the 80% concentration of CIPs. The similar result was also obtained for high viscosity fluid core samples. On the other hand, the shifting of the stress-strain curves is negligible for the solid core samples (Figure 4.8(b)) regardless of the concentrations of magnetic particles, this is due to the locked magnetic particles inside the solid MR core. For low and high viscosity carrier fluid cores with 80%

particles concentrations, it is highly noticeable that the modulus and energy dissipation were increased with increasing current applied to the electromagnet. The increase in the modulus of the soft MR elastomer can be visualized from the increase in the slope of the stress-strain curves, while the increase in the energy dissipation can be visualized from the increase in the area enclosed by the stress-strain hysteresis loops when a magnetic field is applied.

Figure 4.7: Engineering stress-strain hysteresis loops for increasing magnetic field with applied current 0, 1, 2 and 3 A at an excitation frequency of 0.1 Hz and a strain amplitude of 10 % for soft hybrid MR elastomer with low viscosity carrier fluid core samples with (a) 20%, (b) 40%, (c) 60% and (d) 80% concentration of CIPs.

Figure 4.8: Compressive stress-strain hysteresis loops for core-shell soft hybrid MR elastomer (a) high viscosity fluid core, and (b) solid core for 80% w/w CIPs concentration.

4.1 Linear Moduli and Energy Dissipation

Firstly, the stress-strain responses are analyzed by obtaining the linear moduli. The linear moduli of soft MR elastomers were obtained as a slope of the stress-strain curve for the loading condition. Energy dissipation per unit volume of the sample was obtained by obtaining the area under the loading and unloading curves. Figure 4.9 shows the linear moduli of the soft hybrid MR elastomers with three different types of the core (i.e. low viscosity, high viscosity and solid) with four different concentrations of CIPs (20%-80% w/w). A number of observations can be visualized in Figure 4.9. In the absence of a magnetic field, the modulus of soft hybrid MR elastomer slightly increased with increasing concentration of magnetic particles. The zero-field modulus for all three cores (low viscosity, high viscosity and solid) are found to be similar for concentrations up to 60%. On the other hand, the modulus was slightly higher for 80% CIPs; the modulus of the solid core sample was found to be the highest followed by high viscosity core and low viscosity core samples. The effect of the magnetic field was hard to be observed for solid core samples regardless of the concentrations of magnetic particles. For fluid cores, the modulus was also found to be increased with increasing the current to the electromagnet. The increase in the modulus was more pronounced for 80% particles concentration.

Figure 4.9: Linear moduli at various amounts of current supplied to the electromagnetic device for all three types of cores (low viscosity fluid, high viscosity fluid and solid) and at four different concentrations of CIPs.

Energy dissipation under loading and unloading curves were obtained in both the absence and presence of a magnetic field for all three types of samples and is given in

Figure 4.10. In the absence of a magnetic field, the energy dissipation under loading and unloading cycle for fluid core soft MR elastomers was found to be slightly decreased or almost constant with increasing concentration of CIPs for both low and high viscosity fluids. That signifies that the hysteric damping is decreased with increasing particle concentration for fluid core samples. In the presence of a magnetic field, the energy dissipation was also found to be increased with an increasing magnetic field for all concentrations of CIPs and is more pronounced with a higher concentration of CIPs. The highest effect was again observed for samples with 80% CIPs. It was also noted that the increase in energy dissipation was less significant after 2 A current. This is attributed to encapsulated MR fluid. For solid core, the energy dissipation for the solid core was found to be increased with increasing concentration. That signifies that the zero-field hysteric damping is increased with increasing particle concentration within the solid core samples. However, the effect of the magnetic field was virtually negligible for solid core samples. Even though the amount of energy dissipation can be found from the cyclic compression testing, these test results are not used for obtaining storage and loss modulus as the cyclic compression only have triangular loading not the sinusoidal. These results are only used to analyze the MR effect by obtaining the secant modulus. Elastic and damping properties will be achieved via forced vibration analysis and will be presented in **Chapter 5**.

Figure 4.10: Energy dissipation under loading and unloading curves at various amounts of current supplied to the electromagnet for all three types of MR cores (low viscosity fluid, high viscosity fluid and solid) and at four different concentrations of CIPs.

4.2 Secant Modulus and Magnetorheological Effect

The linear modulus is unable to describe the non-linearity of the stress-strain response of viscoelastic materials as it is just obtained as a slope of the curve. Therefore, other moduli such as secant and tangent moduli are used. The tangent modulus is the slope of the stress-strain response at any point. Secant modulus is stress divided by strain at any given value of stress or strain. It also is called the stress-strain ratio. Small non-linearity in stress-strain response can provide a bigger variation in tangent modulus as it is a slope at a point, so tangent modulus becomes highly biased. The unbiased modulus is secant modulus as it is simply a ratio of stress and strain. Thus, in this study, in order to investigate the MR effects, the stress-strain diagrams are analyzed by obtaining secant modulus. Typical secant moduli obtained from the experimental stress-strain response of core-shell hybrid MRE samples is given in Figure 4.11. In the absence of a magnetic field, fluid core samples with all concentration showed almost constant secant moduli with increasing strain. The constant secant modulus indicates a linear stress-strain response. For solid core samples, the low concentration of CIPs up to 60% also showed the linearity, however, 80% CIPs sample showed the highly non-linear response of stress-strain. Upon the application of a magnetic field, for fluid core, the samples up to 60% concentration of CIPs still exhibited linear behavior, however, samples with 80% CIPs showed highly non-linear behavior with increasing strain in the presence of a magnetic field. On the other hand, the magnetic field did not significantly alter the response of solid core samples.

Figure 4.11: Secant moduli versus engineering strain of hybrid MR elastomers with different concentration of CIPs without and with the application of a magnetic field. (a) fluid MR core with high viscosity and (b) solid MR core.

As described in a previous section, compression test with four different amounts of current to the electromagnet was performed to determine the increase of modulus due to the exposure to magnetic flux. The stress-strain hysteresis curves are influenced by the application of a magnetic field as illustrated in Figure 4.7 and 4.8. To study the influence of magnetic field both absolute and relative MR effect (Equation 3.1 and 3.2) are plotted against engineering strain. Figure 4.12 shows the absolute and relative MR effect for four different concentration of CIPs at 3 different amounts of currents to the electromagnet plotted versus engineering strain for high viscosity MR core samples. The MR effects increase with increasing concentration of CIPs and increasing current to the electromagnet, with the 80% CIPs samples achieving the highest relative MR effect. The increasing strain does not affect the MR effects of low concentration samples. On the other hand, for 80% samples, the relative MR effect is found to be increased with increasing strain and become almost constant when strain value reached 8% or higher. Some negative values of MR effects were also observed, this is because of remnant deformation of magnetic particle chains or even the elastomeric case up on the cyclic compression hence causing some amount of plastic strain. Nevertheless, the MR effect increased and to a positive value and became stable within 10% strain value. The maximal values of the relative MR effect are determined within 10% strain. The maximum values of absolute and relative MR effects are summarized in Table 4.2.

Figure 4.12: Absolute and relative MR effect, calculated with secant modulus, of all high viscosity MR core samples of different concentrations of CIPs achieved by applying 3 different amounts of current (1, 2 and 3 A) to electromagnet versus compressive strain.

Solid core samples have less than 10 kPa absolute MR effects because of the locked magnetic particles. The absolute MR effect is slightly increased with increasing

concentrations of CIPs but the relative increased in MR effect remained less than 5%, which is even smaller than that was achieved with 20% CIPs of fluid cores. For fluid cores, the MR effects are found to increase with increasing concentration of CIPs and amounts of current to the electromagnet.

Table 4.2: Maximum absolute and relative MR effects achieved with three different amounts of current (1, 2 and 3 A) to the electromagnet and calculated with secant modulus are listed for all three types of MR cores (low and high viscosity fluid core and solid core) with 20-80% concentrations of CIPs.

MRE samples	CIPs (%)	Absolute MR effect (kPa)			Relative MR effect (-)		
		Current to the electromagnet (A)					
		1	2	3	1	2	3
Low Viscosity Fluid Core	20	3.82	4.24	4.42	0.03	0.03	0.03
	40	21.59	26.16	29.62	0.19	0.22	0.24
	60	38.27	60.88	74.12	0.30	0.47	0.57
	80	122.58	221.32	280.21	0.42	0.99	1.25
High Viscosity Fluid Core	20	12.56	20.83	30.96	0.13	0.19	0.23
	40	25.74	32.50	35.22	0.27	0.32	0.37
	60	48.44	71.73	78.38	0.47	0.69	0.75
	80	141.40	263.06	315.16	0.59	1.05	1.32
Solid Core	20	1.62	1.61	1.96	0.01	0.01	0.01
	40	3.45	6.09	8.63	0.02	0.03	0.05
	60	1.76	3.86	5.39	0.01	0.03	0.04
	80	1.68	2.53	6.18	0.01	0.01	0.01

It was noted that MR effects of low viscosity MR fluid core are slightly lagging compared to that of high viscosity core for a given concentration of CIPs. This behavior must be attributed to the sedimentation phenomenon. For low viscosity MR fluid, magnetic particles need to overcome low viscous resistance compared to high viscosity carrier fluid, thus, they sediment easily for low viscosity fluid core. The sedimented particles would form clumps at the bottom. When the magnetic field is applied, it is possible that magnetic particles would form non-uniform chains along the magnetic flux direction. Because of clumped particles, the non-uniform chains might be stronger on the bottom and weaker on the top. Thus, the MR effects might have been lower for low viscosity fluid core. However, the difference is not very significant at a higher concentration of magnetic particles. Typically for low concentrations of magnetic particles, the results suggested that the low viscosity carrier fluid is not as good as a high viscosity carrier fluid for the core-shell soft hybrid MR elastomers. The relative MR

effect is very competitive for a higher concentration of CIPs (80%). The absolute MR effect can reach as high as 300 kPa and relative MR effect can reach up to 1.3. This signifies that the core-shell structure MR elastomer with MR fluid as a core, MR effect is slightly better for high viscosity carrier fluid.

With respect to the higher MR effects shown by high viscosity carrier fluid cores, relative MR effects are studied versus the magnetic field strength and also versus particle weight concentration in Figure 4.13. To summarize, the MR effect increases with increasing particle concentration and magnetic flux density. The effect of 80% soft hybrid MR elastomers is the highest.

The experimental investigation found that the MR effect was very similar regardless of the viscosity of the core fluid. The free magnetic particles within the MR fluid core could easily be moved and aligned to the direction of magnetic flux lines, and therefore, exhibiting a good MR effect. On the other hand, because of the locked magnetic particles, the solid core soft elastomer was incapable to exhibit the MR effect as good as the fluid core.

Figure 4.13: Maximum relative MR effects of high viscosity fluid core soft hybrid MR elastomers are illustrated (a) versus the concentration of CIPs and (b) versus the magnetic flux density with 3 different amounts of current to the electromagnet.

4.3 Comparison with Previous Investigations

Here, the MR effects achieved in this study are compared with the results of published studies. Even though the direct comparison of the MR effects of this study with the published investigations is difficult task as the MR effects are highly dependent on concentration of CIPs, type of matrix, structure of MREs (isotropic or anisotropic), and strength of an applied magnetic field, an attempt has been given in view of the maximum

reported MR effect. To compare the MR effect achieved in this study with experiments found in the literature, the absolute and relative MR effect related to 100 mT magnetic flux density are obtained for all studies and illustrated in Figure 4.14.

Figure 4.14 also includes the two types of hybrid MREs developed in this study, core-shelled and 3D printed MREs (**Chapter 5**). The data analysis and all method adopted to obtain the MR effects and modulus are the same for every investigation including this study. Only results obtained via the compression testing method are considered for the comparison. When the MR effects are scaled down to 100 mT, the highest relative MR effect is about 95% and was achieved by Abramchuk et al [162], however, the absolute MR effect is much lower, 16.91 kPa. On the other hand, Schubert et al [15] have the highest absolute MR effect, 811.11 kPa, but lower relative MR effect, 24.67 %. The literature results strongly suggest that achieving higher values for both absolute and relative MR effect is not convergent. In other words, absolute MR effect and relative MR effect are inversely related. One common phenomenon observed in these studies is strain level, the highest MR effect for all studies is observed below 10% strain.

Firstly, the MR effects achieved for core-shell hybrid elastomers and the comparison with published investigations are discussed. Both the absolute and relative MR effects achieved in this study for 80% CIPs and added 2% MNPs (**Section 4.5**) core-shell hybrid MR elastomers are stable. In other words, both absolute and relative MR effects of core-shell hybrid MR elastomers are higher than that of previous investigations. The highest absolute MR effect is 275 kPa and relative MR effect is 68% for 80% CIPs core-shell hybrid MRE. The absolute MR effect of core-shell hybrid MRE of this study is neither as low as that result in the highest relative MR effect nor as high as that result in the lowest MR effect. Moreover, addition of MNPs increased the both absolute and relative MR effect of core-shell hybrid MR elastomers.

Similar trend of the absolute and relative MR effect can be seen for 3D printed hybrid MR elastomers developed in this study. The 3D printed MR elastomers, both line and dot patterned (**Chapter 5**), showed stable absolute and relative MR effect than most of previous investigations.

Figure 4.14: (a) Absolute and (b) relative MR effect related to 100 mT magnetic flux density is compared for each other of the experiments published in the literature and the MR effect achieved in this study. The seven blue bars on the left side represent the MR effect of published articles and the four pink bars on the right represent the MR effect achieved in this study. The results are compared only for compression test. The reference of above instigations can be found in Section 2.5.

4.4 Forced Vibration Testing

In this section, forced vibration testing results are presented for 80% particles concentration for all three types of the core for soft hybrid MR elastomers. Samples are tested in both squeeze and shear mode of operation.

4.4.1 Squeeze Mode

Soft hybrid MR elastomer samples were characterized by a frequency sweep in the range of 50-200 Hz at a constant acceleration amplitude of 0.5 g (effect of frequency and amplitude of vibration for hybrid MR elastomer using forced vibration testing will also be presented in **Chapter 5**). Figure 4.15 depicts the magnitude transmissibility for all

three types of cored soft hybrid MR elastomers at various magnetic flux densities against the excitation frequency. The direction of the magnetic field can be found in Section 3.2. Several features can be noted in Figure 4.15. The peak of the magnitude transmissibility curve represents the natural frequency of the single degree of freedom system. All types of samples exhibited a single peak, which indicates the system is a single degree of freedom. When a magnetic field is not applied, the low viscosity fluid core has the lowest natural frequency and the solid core has the highest. Upon the application of the magnetic field, magnitude transmissibility curves are moving toward the right, which indicates the change in stiffness of the soft hybrid MR elastomers.

Figure 4.15: Magnitude transmissibility versus excitation frequency of the soft hybrid MR elastomers in a squeeze mode at various magnetic flux densities.

The peak of the magnitude transmissibility curve indicates the natural frequency. Using Equations 3.3 and 3.8, the stiffness and damping ratio were obtained as described in **Section 3.3**. Stiffness and damping ratio versus magnetic flux density are presented in Figure 4.16. In the absence of magnetic field, the solid core has the highest stiffness, followed by high viscosity fluid core and low viscosity fluid core samples. It can be noted that the stiffness values for the fluid cored elastomers are very close. When the magnetic field was applied, again the solid cored elastomer showed a very small change in the stiffness while the fluid cored elastomers showed a clear change in the stiffness with increasing magnetic flux density. In case of the fluid core sample, it was also found that the increase in the stiffness is less pronounced as the magnetic flux density exceeded 300 mT, which is true because the field strength around 0.3 Tesla is found to be optimal for MR fluid. This can also be visualized from the magnitude transmissibility curves;

the shifting of the transmissibility curves is less pronounced above 300 mT of magnetic flux density.

Figure 4.16: Compressive stiffness and damping ratio of soft hybrid MR elastomers at various magnetic flux densities.

For the damping ratio, in the absence of the magnetic field, the high viscosity fluid core elastomer has a higher damping ratio than the other two types. This suggests that the high viscosity fluid has a higher ability to absorb the shock and vibrations because the viscous damping coefficient depends on the viscosity of the damping material. Because of the high viscosity of the fluid core, samples with high viscosity core have higher damping ratio than that of low viscosity core and solid core. When the magnetic field is applied, the damping coefficient was either almost constant or slightly decreased with increasing magnetic field for all three types of soft hybrid MR elastomer samples, which is also typical behavior of the conventional MREs [11, 12].

The relative MR effect for natural frequency and stiffness were also obtained using Equation 3.2 at 0 mT and 500 mT of magnetic flux density. The relative MR effect for solid core samples was found to be very small (<10%). The relative MR effect for fluid core samples was found to be 96.5% and 40.2% for stiffness and natural frequency respectively for low viscosity fluid core. Again, the samples with high viscosity core have a slightly higher increment in stiffness compared to that of low viscosity core samples as obtained in the cyclic compression test. Here, the force vibration results also suggest that fluid core-based soft hybrid MR elastomers exhibit a good MR effect even at the moderate magnetic flux density. Moreover, the high viscosity fluid core-based samples also showed as good or even better MR effect as the low viscosity fluid core-based samples.

4.4.2 Shear Mode

With respect to the better MR effect compared to others, 80% high viscosity fluid core samples have been characterized in a shear mode of operation. In the shear mode, in order to study the anisotropic MR effect of the soft hybrid MR elastomers, the magnetic field was applied in parallel (H_0) and normal (H_{90}) as shown in Figure 4.17.

Figure 4.17: Representation of the application of a magnetic field in a shear mode.

The results for the effect of the orientation of the magnetic field is shown in Figure 4.18. As the magnetic particles are free to move within the core of the carrier fluid, they can form chains along the direction of magnetic flux. Thus, a clear anisotropic effect can be observed in Figure 4.18 when a magnetic field is applied in a different direction.

Figure 4.18: Magnitude transmissibility as a function of excitation frequency for the fluid core soft (80%) hybrid MR elastomer in a shear mode in two different orientations and four at different magnetic flux densities.

The shifting of the magnitude transmissibility curve is more pronounced when the magnetic field was normal to the thickness of the samples. This can be explained as an effect of the length of the chains of the magnetic particles in such conditions. When a magnetic field is parallel to the thickness of the sample, longer chains of magnetic

particles were formed. On the other hand, shorter chains of magnetic field particles were formed when the magnetic field was normal to the thickness of the samples. The longer chains are obviously weaker compared to the shorter chains that are formed by the equally strong magnetic field because the longer chains are prone to buckling. Secondly, the total cross-section area of the shorter chains is higher than the longer chains within the core of the samples. Thus, soft hybrid MR elastomer exhibited a higher MR effect when the magnetic field was applied normally to the thickness of the samples. The anisotropic behaviors of the soft hybrid MR elastomer can also be visualized from Figure 4.19 where stiffness and damping ratio versus magnetic flux density are given. The shear stiffness was found to be increased with increasing magnetic flux density but was more pronounced when the magnetic field was applied normal to the thickness of the samples. On the other hand, the damping ratio was decreased with increasing magnetic field and again showed anisotropy with respect to the direction of the applied magnetic field. This finding provides the evidence that the soft hybrid MR elastomer exhibits the anisotropic MR effect as exhibited by conventional anisotropic MR elastomers. However, these hybrid MR elastomers do not need a magnetic field during the fabrication process as that of conventional anisotropic MREs.

Figure 4.19: Shear stiffness for the soft hybrid MR elastomer (80% particles concentration) at different orientations and strengths of the magnetic field.

4.5 Effect of Addition of MNPs

MNPs are one of the best choices of additives to enhance the stability and performance of MR fluid. Similarly, such MNPs are also used in conventional MREs as one of the effective additives to increase the MR effect. In this study, MNPs are added with two aims, (1) to enhance the MR effect and (2) to increase the viscosity of low viscosity MR

core to overcome the sedimentation problem. The effect of the addition of MNPs to 80% low viscosity carrier fluid has been studied by adding MNPs up to 2% w/w. To prepare the suspensions, the MNPs are added to a low viscosity with 80% CIPs and thoroughly stirred about 20 mins, thereafter, the mixture was put on the sonicate bath for another 20 mins. The SEM images of the CIPs and MNPs mixture of fully cured sample is given in Figure 4.20.

Figure 4.20: SEM images of samples loaded with CIPs and MNPs.

Figure 4.21: Shear stress versus shear rate for the addition MNPs (closed) on 80% CIPs and compared with low viscosity with 80% CIPs (open) at different magnetic flux densities. (a) 1% MNPs and (b) 2% MNPs.

The magnetorheological results for the addition of MNPs and compared to 80% CIPs are given in Figure 4.21. Both shear stress and viscosity were found to be increased with increasing MNPs. Upon the application of the magnetic field, the enhancement of shear stress and viscosity are higher than that of 80% CIPs. The MNPs can fill the voids between micro CIPs making the chains stronger in the presence of a magnetic field. The shear thinning was also enhanced with the addition of MNPs. The rheological result showed the significance of the addition of MNPs to enhance the viscosity and MR effect of suspension. The effect of the addition of MNPs for the core-shelled hybrid MR

elastomers will be presented later in this section. In addition, resistance to sedimentation was also significantly enhanced by the addition of MNPs. The MR suspension with added MNPs are found to be very stable and sedimentation was not observed over 30 days (Figure 4.22).

Figure 4.22: Sedimentation behavior of MR suspension with added MNPs (2% w/w) to CIPs (80% w/w).

Figure 4.23: Absolute and relative MR effect, calculated with secant modulus, of fluid MR core samples with addition of MNPs up to 2% on 80% CIPs and compared with low viscosity with 80% CIPs achieved by applying 3 different amounts of current (1, 2 and 3 A) to electromagnet versus compressive strain.

The absolute and relative MR effect achieved from the cyclic compression testing for the addition of 1% and 2% MNPs to 80% CIPs versus engineering strain and compared with 80% CIPs are given in Figure 4.23. As achieved in the magnetorheological study, the effect of the addition of MNPs is clear that the MR effects are increased compared to that of pure CIPs. In the cyclic compression, the maximum MR effect at 3 A current was found to be 1.51 and 1.55 for samples with 1% and 2% MNPs respectively while it was 1.25 for the sample with pure CIPs only. The rest of behaviors such as an increase in MR effect with strain and becoming stable and increase of MR effect with increased current are similar even with the addition of MNP as that was achieved for other samples as discussed in **Section 4.3**.

Similarly, the forced vibration testing was also performed to study the MR effect of added MNPs. The magnitude transmissibility curves obtained from forced vibration testing in a squeeze mode at various magnetic flux densities is given in Figure 4.24. The zero-field stiffness is slightly increased with the addition of MNPs. On the other hand, the peak of transmissibility curves slightly decreased with increased concentration of MNPs. The decrease in the peak of the transmissibility curve signifies that the damping ratio of the MRE system is increased, this is a result of increased viscosity with the addition of MNPs. When the magnetic field is applied, the shifting of the transmissibility curve is found to be significantly enhanced by the addition of MNPs. Still, the shifting of the transmissibility curve is found to be gentler when magnetic flux density is higher than 300 mT as that was achieved for other fluid core samples. The natural frequency of the system was increased to 134.7 Hz from 72.35 Hz for the sample with 1% MNPs, to 176.7 Hz from 86.05 Hz for the sample with 2% MNPs, whereas natural frequency only increased to 94.92 Hz from 67.7 Hz for the sample with only 80% CIPs. The relative increase in natural frequency of the system with hybrid MRE samples is 141%, 105% and 40% for samples with 2% MNPs, 1% MNPs and 80% CIPs respectively. On the other hand, increase in the stiffness of the elastomer would be even higher than that was achieved for natural frequency as the increase in stiffness depends on both initial and the increase in the natural frequency. The relative increase in stiffness was found to be 322%, 246% and 96% for samples with 2% MNPs, 1% MNPs and 80% CIPs respectively. All the relative MR effect are obtained using Equation 3.2. The stiffness and damping ratio versus magnetic flux density as obtained from the transmissibility curves for all three types of samples are summarized in Figure 4.25.

In summary, the addition of MNPs is proven to be noteworthy. Firstly, it enhanced the rheological properties of the suspension in both the absence and presence of a magnetic field, so the sedimentation behavior of the MR fluid is improved. Secondly, a significant improvement on the MR effect of the core-shell hybrid MR elastomers was clearly observed in both cyclic compression and forced vibration testing.

Figure 4.24: Magnitude transmissibility versus excitation frequency of the soft hybrid MR elastomers with MNPs added to 80% CIPs and compared with the sample of pure CIPs in a squeeze mode at various magnetic flux densities.

Figure 4.25: (a) Compressive stiffness and (b) damping ratio of soft hybrid MR elastomers with MNPs added to 80% CIPs and compared with the sample of pure CIPs at various magnetic flux densities.

4.5 Summary of the Chapter

This chapter described the development and characterization of core-shelled soft hybrid MR elastomers. The soft hybrid MR elastomers with two types of fluid cores and a solid core with four different concentrations of magnetic particles have been fabricated and their magneto-mechanical properties have been investigated. It was found that the solid core samples showed relatively smaller or no MR effect, whereas the MR effect was increased with increasing concentration of magnetic particles for fluid core samples. The highest MR effect was observed for 80% w/w concentration of magnetic particles. But the MR effect can be enhanced by the addition of MNPs. The addition of the MNPs was proven to be significant in terms of both MR effect as well as the stability of the core fluid. The experimental results showed a clear increment of modulus, energy dissipation,

absolute and relative MR effects of the soft hybrid MR elastomers with increasing magnetic flux density. Forced vibration testing results also showed a clear increment in the stiffness with increasing magnetic flux density while the damping coefficient was found to be not significantly altered by the magnetic field.

From the magnetorheological study, it was found that the high viscosity carrier fluid is not as good as a low viscosity carrier fluid for the shear stress and viscosity enhancement. However, for the core-shell soft hybrid MR elastomer with the fluid core, the finding suggested that the MR effect exhibited by high viscosity carrier fluid core is as good as the low viscosity carrier fluid core or even higher (for low concentration of CIPs). For core-shell structure, it was also found that the MR effect was relatively lower for low concentrations (20% & 40% w/w) of magnetic particles, however, was not the case when the only MR fluid property was studied via magneto rheology. The high viscosity carrier fluid is also superior to low viscosity carrier fluid when the sedimentation and leakage are concerned. The high viscosity fluid core soft hybrid MR elastomers are free from the sedimentation issue of MR fluids of the core. It was also demonstrated that such hybrid MREs also exhibit anisotropic MR effect as conventional anisotropic MREs when the direction of the applied magnetic field is changed as the mobile particle within the core can easily move in the direction of magnetic flux. Moreover, the addition of MNPs is also proven to be significant as they enhanced sedimentation behavior for low viscosity carrier fluid and also improved the MR effect of core-shell hybrid MR elastomers. The soft hybrid MR elastomer with fluid core with either high viscosity carrier fluid or enhanced viscosity of low viscosity carrier fluid with MNPs are proven to be potential materials to bridge the gap between MR fluid and MR elastomer. Such fluid core hybrid MR elastomers provide good resistance to flow and sedimentation of magnetic particles within the core, as well as they, exhibit anisotropic MR effect. The core-shell soft hybrid MR elastomer is capable of changing its stiffness even in a moderately strong magnetic field thus such elastomers also offer the lower working range of the magnetic field.

Chapter 5: Development of Hybrid MREs by 3D Printing

This chapter presents the details and challenges to develop hybrid MR elastomers by a 3D printing method. In 3D printing, various MR fluid filaments are precisely and accurately configured and encapsulated in the elastomeric matrix. Thereafter, the MR effects shown by various patterned 3D printed MR elastomers are investigated. Lastly, the chapter presents the dynamic behavior of 3D printed MR elastomer under uniaxial deformation in the absence and presence of a magnetic field through a forced vibration technique.

5.1 Introduction

Magnetic particles are homogeneously distributed in an isotropic MRE, whereas in anisotropic MRE, magnetic particles could be distributed in a unique arrangement which might offer higher MR effect. However, it is very challenging to uniquely distribute the magnetic particles in the desired fashion without applying a magnetic field. One of the possible ways to configure magnetic particles inside an elastomer is to use the 3D printing method for the fabrication of MRE, where, magnetic particles can be configured accurately and precisely within the matrix material in the desired fashion without applying a magnetic field. This possibly would result in anisotropic MREs or other types. Nonetheless, 3D printing has not been studied in detail in the field of MR materials except a single work presented by Hannes et al [233] in 2014, however, the printing was unsuccessful. Their dry powder printing had a very poor flowability, resulting in several problems such as discontinuous printing lines, weak bonding between a silicone layer and dry powder, and scattering of the printed patterns. Yet, the study is a good reference. Afterward, no further studies dealing with the development of MREs via 3D printing can be found in the literature by the same group or by others. It is expected that 3D printing would allow configuring the magnetic particles with completely arbitrary distributions or configuration, therefore, 3D printing would be more appropriate to develop anisotropic MR elastomer. 3D printing or additive manufacturing enables fabrication of intricate geometric structures with the flexibility of co-printing various materials in a layer by layer fashion. Various materials such as

alloys, polymers, ceramics, and even composite have already been 3D printed. The materials that could be printed include powder, liquid or suspension forms, and different types of dispensing mechanisms and equipment are used to print them into 3D constructs.

5.2 Deduction of the Shear Rate During Printing

The fundamental idea of the hybrid MR elastomer printing is to extrude the MR fluid to print various patterns within the elastomer matrix. In an extrusion-based 3D printing process, shear forces may break the internal network of the fluid suspension ink, and as a result, the viscosity will be reduced. Therefore, the extrusion-based printing method is also applicable to print highly viscous materials. An extrudable material should have a thixotropic nature, which means that the viscosity of the given material is low when a shear force is applied but viscosity can be recovered quickly after the shear force is removed. Thus, the rheological properties of MR fluids play a vital role in extrusion-based printing. In order to study the thixotropic properties of MR fluid, a recovery test has to be carried out. In the recovery test, the shear rate experienced by the MR fluid during the printing process must be known. Hence, prior to the recovery test, the shear rate experienced by the MR fluids during the printing process was deduced as follows by exploiting the basic fluid mechanics and the rheological properties of MR fluids.

Figure 5.1: (a) Schematic illustration of the MR fluid printing system consisting of a piston-cylinder unit and a printing nozzle. (b) Printing cartridges with MR fluid (black) and elastomer matrix (clear).

The printing system consists of a cylinder-piston unit and a dispensing nozzle as shown in Figure 5.1. The cylinder and the nozzle have different internal diameters. Considering the laminar flow inside both the cylinder and the nozzle, the shear rate experienced by

the MR fluid during the printing could be deduced. Henceforth, the deduced shear rate is useful in the recovery test on the rheometer.

Figure 5.2: Stress and velocity profiles for a non-Newtonian fluid in the circular pipe.

Let us consider a laminar and steady flow of an incompressible fluid in a circular pipe with diameter $2R$. The schematic illustration is shown in Figure 5.2. If the pressure drop along the pipe length is ΔP , shear stress (τ) at any point inside the pipe can be obtained as:

$$\tau = \frac{-\Delta P}{2L} r \quad (5.1)$$

where r is the radius ($0 < r < R$). The shear stress can be obtained as a function of shear rate ($\dot{\gamma}$) as given by the power-law for fluids.

$$\tau = m\dot{\gamma}^n \quad (5.2)$$

where m and n are the power law consistency coefficient and index respectively. Similarly, the viscosity of any fluid can be written as

$$\eta = m\dot{\gamma}^{n-1} \quad (5.3)$$

Now, combining equations 5.1, 5.2 and 5.3, the maximum shear rate in the cylinder wall (i.e. $r = R$) is

$$\dot{\gamma} = \frac{-\Delta P}{2\eta L} R \quad (5.4)$$

Equation 5.4 is the maximum shear rate experienced by the MR fluid inside the cylinder during the printing. However, the actual pressure difference (ΔP) is difficult to obtain from the printing process. Similarly, for any shear thinning fluids, the viscosity (η) is shear rate-dependent. Thus, to obtain the shear rate of the flowing fluid inside a cylinder, we need to get rid of these two terms (pressure difference and viscosity) from Equation 5.4.

For a non-Newtonian fluid, the volumetric flow rate (Q) as related to the pressure drop (ΔP) along the length L with a uniform flow rate (V) can be written as [234]:

$$Q = \pi R^2 V = \pi \left(\frac{n}{3n+1} \right) \left(\frac{-\Delta P}{2mL} \right)^{\frac{1}{n}} R^{\left(\frac{3n+1}{n} \right)} \quad (5.5)$$

Now, the pressure difference can be obtained as

$$\Delta P = -2mL \left[\frac{VR^2}{\left(\frac{n}{3n+1} \right) R^{\left(\frac{3n+1}{n} \right)}} \right]^n \quad (5.6)$$

Thus, by combining Equations 5.3, 5.4 & 5.6, the shear rate inside of the circular hose can be expressed as

$$\dot{\gamma} = \frac{VR^{(2+n)}}{\left(\frac{n}{3n+1} \right) R^{\left(\frac{3n+1}{n} \right)}} \quad (5.7)$$

Again, the shear rate is depended on the power law index (n) which is unknown. Thus, the rheological study of the MR fluid is essential prior to obtain the actual shear rate inside the cylinder and the printing nozzle. Based on the power-law model and experimental data, the power law indices (m and n) could be obtained via curve fitting. In this fashion, the shear rate experienced by the MR fluid during the printing can be deduced. For a given system, cylinder radius (R_2) and nozzle radius (R_1) are known. The information for the flow rate (V) of the MR fluid can be obtained from the printing process. Thus, the shear rate experienced by the MR fluid inside the nozzle during the printing can be obtained from the following equation (Equation 5.8).

$$\dot{\gamma} = \frac{V_2 R_2^{(2+n)}}{\left(\frac{n}{3n+1} \right) R_2^{\left(\frac{3n+1}{n} \right)}} \quad (5.8)$$

3.3 Rheological Study

This study aims to explore the 3D printability of MR materials to develop two types of hybrid magnetorheological elastomers (H-MRE) via 3D printing. The first type of MR material must solidify after being printed, whereas in the second type of MR materials the MR fluid encapsulated should remain fluid even after printing. The H-MREs are to

be developed by encapsulating the MR suspensions/fluids inside the elastomer matrix via 3D printing. The detailed fabrication process is presented in **Section 5.4**. Hereafter, the hybrid MR elastomer developed via 3D printing with solid MR structures inside the elastomer matrix is regarded as 3DP-MRE1 and the hybrid MR elastomer with MR fluid structures inside the elastomer matrix is regarded as 3DP-MRE2. For 3DP-MRE1, vulcanizing materials are desired while non-vulcanizing materials are required for 3DP-MRE2. Thus, selecting the printable MR materials for both types of 3DP-MREs was a great challenge because there are no previous studies focusing on 3D printing of MR materials.

One of the efficient ways to select printing materials for the extrusion-based printer is to study the rheological properties of the materials. The ideal printable materials should be highly shear thinning and thixotropic. Therefore, to select the best 3D printable materials, the rheological study had to be conducted. For 3DP-MRE1, total six types of vulcanizing silicones were considered for the rheological study and the silicone with the good shear thinning and thixotropic properties was further modified for the final printing, whereas, the commercial Lord MR fluid (MRF-132DG) was modified to make printable to develop 3DP-MRE2.

5.3.1 Materials Selection for 3DP-MRE1 Printing

Six vulcanizing silicones were considered for the rheological study as summarized in Table 5.1. Four different types of carrier materials for magnetic particles were chosen: namely, UV curable silicone (SS-UV 465 and SS-155), room temperature vulcanizing silicone (SS-B6), elevated temperature vulcanizing silicone (SS-3003 and SS-3303) and room temperature vulcanizing with platinum catalyzed additional cure silicone (SS-3006T). The similar types of vulcanizing silicones have also been extensively used for the development of conventional MREs [11, 12]. Here, the aim is to select the vulcanizing silicone: it has to be printable as well as highly stable throughout the printing period.

Table 5.1: Vulcanizing silicones considered as a carrier fluid for magnetic particles to develop H-MRE1 via 3D printing.

S.N.	Name	Property	Supplier
1	SS-UV 465	Silicone solution, UV curable	AB Technology Group, USA
2	SS-155	Silicone solution; UV curable	Silicone solution, Cuyahoga Falls, USA
3	SS-3003	Silicone solution; curable at 150 °C for 1-10 minutes	same as above
4	SS-3303	Silicone solution; curable at 150 °C for 1-10 minutes	same as above
5	SS-B6	Silicone solution; curable at room temperature for 24 hours	same as above
6	SS-3006T	Silicone solution (platinum catalyzed, addition cure system); curable at room temperature for 8 hours	same as above

Figure 5.3: (a) Viscosity as a function of shear rate and (b) Viscosity recovery ability for different vulcanizing silicones at room temperature.

When the viscosity of a fluid decreases with increasing shear rate, the material is regarded as a shear thinning fluid. In other words, for a shear thinning fluid, power law index (n) should be smaller than 1 [234]. Figure 5.3(a) depicts the shear thinning

properties of all 6 types of vulcanizing silicones. In the steady-state flow test, it was found that the UV curable silicones (SS-UV 465 and SS-155) had low viscosity and weak shear thinning (n near to 1). The room temperature vulcanizing (RTV) silicone (SS-B6) showed a significant shear thinning ($n = 0.25$) with moderate viscosity. The elevated temperature vulcanizing silicones (SS-3003 and SS-3303) had more shear thinning than the UV curable silicones but weaker in shear thinning than the RTV silicone with moderate viscosity. Last of all, the SS-3006T showed the remarkable shear thinning ($n = 0.19$) and the highest viscosity among these vulcanizing silicones.

Table 5.2: Power law indices m and n obtained via curve fitting and the maximum shear rate inside the nozzle for all six types of vulcanizing silicones.

Silicone Matrix	m	n	Shear Rate ($\dot{\gamma}$)
SS-UV465	3.89	0.98	483.68
SS-155	2.22	0.86	534.50
SS-3003	29.74	0.63	148.62
SS-3303	24.86	0.66	133.64
SS-B6	11.52	0.25	313.60
SS-3006T	374.3	0.19	112.37
SS-3006T MR	651.6	0.22	90.54

Thereafter, recovery tests were conducted to mimic the printing process. The test consists of three stages. At stage I, the material inside the nozzle before printing was mimicked by applying a very low shear rate of 0.1 s^{-1} for 60 seconds. At stage II, the material extrusion during the printing was mimicked by applying the shear rate experienced by the fluid during the printing (as described in **Section 5.2**) for 10 seconds. Finally, at stage III, the material after printing was again mimicked by applying a very low shear rate of 0.1 s^{-1} for 60 seconds. In rheometer, experiments could not be conducted with zero shear rate, therefore, a very low shear rate of 0.1 s^{-1} was chosen to mimic the before and after printing condition. Whereas, the shear rate experienced by the MR fluid during printing can be obtained using Equation 5.8 as described in **Section 5.2**. Firstly, the power law indices (m and n) were obtained for each sample via curve fitting based on the power-law and the experimental data. The material extrusion velocity (v) could be obtained from the printing process. Thus, using the extrusion velocity (v), power law index (n) and the printing nozzle size (R_2), the shear rate experienced by the fluid during the printing can be obtained using the deduced equation (Equation 5.8). The power law indices (m and n) and the maximum shear rate inside the nozzle for all six vulcanizing silicones are given in Table 5.2. Several features can be obtained from Table 5.2. For example, the lower the value of n , the more the shear

thinning and vice versa. The higher the viscosity of the fluid, the higher the values of m and vice versa. On the other hand, the low viscosity fluid experiences the higher shear rate inside the nozzle. It was found that the different value of the shear rate was observed for different types of silicones. It is not a good practice to apply different shear rates for different silicones in the recovery test because it might result in incomparable experimental outcomes. Therefore, a rational shear rate value had to be chosen. Here, as the best shear thinning materials (SS-3006T) experienced the maximum shear rate of around 100 s^{-1} , this value was used for all materials during the recovery tests.

Figure 5.3(b) shows the viscosity recovery ability of the different types of vulcanizing silicones. Because the UV curable silicones (SS-UV465 & SS-155) did not show a significant shear thinning behavior, viscosity did not change much when the higher shear rate was applied. Whereas, the viscosity of the RTV silicone (SS-B6) was significantly decreased when the high shear rate was applied, and the viscosity was also highly recovered (up to 70% within 10 seconds) after the shear force was removed. On the other hand, the high-temperature vulcanizing silicones (SS-3003 & SS-3303) had a poor recovery ability (viscosity recovered less than 50%). Yet again, the highly shear thinning silicone (SS-3006T) showed a remarkable viscosity recovery ability by recovering its viscosity up to 75% within 10 seconds after the shear force was removed. The viscosity could be recovered up to 85% if longer recovery time was given. However, the viscosity could not be further recovered after 30 seconds.

The 3D printability is determined by the shear thinning and the thixotropic property of the material. To this end, vulcanizing silicones, SS-B6 and SS-3006T, showed a good shear thinning and viscosity recovery. However, the SS-3006T has the lowest value of power law index $n = 0.19$ (i.e. highest shear thinning rate) and highest recovery ability (75 % viscosity was recovered within 10 seconds). On the other hand, SS-3006T is very stable and does not cure at room temperature even when it is left over a few days, whereas, rest silicones start to solidify within a short time even though they take about 24 hours to be fully cured. Such printing material is desired, which is 3D printable as well as stable at least during the printing period. Therefore, with respect to the noteworthy rheological properties and the stability, SS-3006T was selected as a 3D printing material for the fabrication of 3DP-MRE1.

Afterward, iron particles were added to SS-3006T silicone to make MR suspension and it was named as SS-3006T MR. The shear thinning, and recovery ability of the SS-3006T-MR are also presented in Figure 5.3 (b). The concentration of magnetic particles is about 80% w/w. The addition of iron particles increased the viscosity so that the shear thinning behavior was not altered significantly. Likewise, the viscosity recovery ability remained similar, but the curve moved upward (red curve in Figure 5.3) because of the increased viscosity.

In conclusion, with such desirable properties (shear thinning, thixotropic and stability), the MR suspension with SS-3006T silicone was chosen as the printing material for the fabrication of 3DP-MRE1 via 3D printing.

5.3.2 Materials Selection for 3DP-MRE2 Printing

The 3DP-MRE2 is a type of hybrid MR elastomer that has MR fluid filaments inside the elastomer matrix. Here, the commercial Lord MR fluid (MRF-132DG) was modified to be printable. As mentioned earlier, printable materials should have low power law index (n) and high viscosity recovery ability.

For the printing, the viscosity of the Lord MRF was significantly low, thus to increase the viscosity of Lord MRF, firstly, only CIPs were added. The addition of iron particles increased the viscosity to some extent, but shear thinning and recovery ability were slightly decreased. Even with 90% w/w iron particles, the viscosity was still low for the printing. Therefore, the addition of iron particles remained unhelpful and an alternative was needed.

Another type of MRF with high viscosity was prepared using silicone oil from Dow Corning Corporation and carbonyl iron powders. The same silicone that has used to developed core-shell hybrid MR elastomers as presented in **Chapter 4**. This MRF had a very poor shear thinning behavior ($n = 0.81$) and a low viscosity recovery ability, as shown in Figure 5.4. This implies that the Dow Corning silicone MRF has a poor printability even it has a high viscosity. Thus, we mixed the low viscosity Lord MRF with the high viscosity Dow Corning MRF. When the volume ratio of these two MRFs was 1:1, the mixed MRF interestingly showed a good shear thinning property as well as a high viscosity recovery ability. The power law index value was 0.39 and viscosity was

recovered about 75% within 10 seconds. Nonetheless, the viscosity was still low for the printing and the printed patterns collapsed quickly. Yet again, iron particles were added to enhance the viscosity of the mixed MRF. The red curves (Figure 5.4) show the results of the modified mixed MRF. After the addition of the iron particles, the viscosity was increased, and which was similar to the previous printable material (SS-3006T MR). Simultaneously, the shear thinning was slightly enhanced i.e. power index value was decreased ($n = 0.32$). Yet, the recovery ability remained identical. In this way, commercially available Lord MRF was modified to make printable via mixing with highly viscous fluid and iron particles. The modified mixed MRF had a power law index of 0.32 and viscosity was recovered by 75% within 10 seconds. This modified MRF was used as a printing material for 3DP-MRE2 development.

Figure 5.4: (a) Viscosity as a function of shear rate and (b) recovery ability of Lord MRF and modified Lord MRF with the addition of Dow Corning high viscosity fluid and CIPs.

Thereafter, a frequency sweep test over the angular frequency range of 0.01-100 rad/s at a constant strain of 2% was carried out to get the storage (G') and loss modulus (G'') of

selected materials (SS-3006T, modified Lord MRF & SS-155). The results are presented in Figure 5.5. The frequency sweep test measures dynamic viscoelastic properties. All three materials were viscoelastic liquids at room temperature. The two printable materials (modified Lord MRF and SS-3006T MR) had similar moduli that slightly depended on frequency. However, the elastomer resins had lower moduli that were increasing with frequency.

Figure 5.5: (a) Dependence of storage modulus G' and loss modulus G'' and (b) dependence of $\tan \delta$ on angular frequency for printing materials (SS-3006T MR & modified Lord MRF) and elastomer matrix (SS-155).

Based on the rheological analysis, two materials, SS-3006T MR and modified Lord MRF, were selected for the 3DP-MRE printing. The SS-3006T MR and modified Lord MRF were used to print various patterns of MR fluids inside the elastomer matrix. Both printing materials exhibited a very good shear thinning behavior and were also highly thixotropic. In addition, the printing materials had similar viscosity and strength before printing.

Figure 5.6: Shear stress and viscosity versus shear rate for two different printing materials, SS-3006T MRF (closed) and modified Lord MRF (open) at different magnetic flux densities.

Lastly, the magnetorheological test was performed to study the rheological behavior of the printing materials under the magnetic field. The same testing method and the apparatus that were presented in **Chapter 4** are adopted. Figure 5.6 shows the shear stress and viscosity versus the shear rate at various magnetic flux densities for both types of printing materials. For both printing materials, a clear shear stress enhancement can be observed with increasing magnetic flux density. In the presence of a magnetic field, the modified lord MRF seemingly exhibited higher shear stress and viscosity at given shear rate than that of SS-3006T MRF, this can be attributed to two factors, firstly, the concentration of two MRF might not be exactly the same and it shows that modified Lord MRF has a slightly higher concentration of CIPs. This can be true because the original Lord MRF has CIPs concentration about 81% w/w and the same concentration of CIPs was maintained when Lord MRF was modified. While the SS-3006T MRF has a concentration of 80% w/w. Secondly, the viscous resistance needs to overcome by CIPs cannot be the same for different types of fluid, it is likely that SS-3006T MRF has higher viscous resistance for CIPs to overcome than that of modified Lord MRF. Nonetheless, the influence of the magnetic field for shear stress enhancement and shear thinning behavior can clearly be observed. The shear thinning is more pronounced for SS-3006T MRF even in the presence of a magnetic field.

5.4 3D Printing of Hybrid MREs

In this study, 3D printing was carried out by accurately configuring the viscous MR fluid filaments inside the elastomer matrix. Therefore, a multi-material printing technique was desired: that is, one head to print the viscous MR fluid and another to print the elastomer matrix. The overall process for 3D printing of the hybrid MR elastomer is illustrated in Figure 5.7. The hybrid MR elastomer was printed layer-by-layer. Each layer required a series of steps. Firstly, two piston-cylinder units, one filled with MR fluid and another with elastomer matrix resin were installed in the printer. To form a bottom layer, the elastomer matrix resin was dispensed from the nozzle and it was cured with the UV light as shown in Figure 5.7. Thereafter, on top of the bottom layer, various structures of MR fluid filaments were printed from another nozzle. After that, those printed patterns were completely covered by the elastomer matrix resin from the previous nozzle and again it was fully cured with the UV light. In this way, the first layer of the hybrid MR elastomer

was completed. The process was repeated until the desired thickness of the sample was achieved.

The viscosity recovery ability and shape fidelity of the elastomer matrix resin was not as crucial as of the MR materials because the main function of the elastomer matrix was to cover the printed patterns, on the other hand, the printability of the MR suspension played a vital role in quality printing. The elastomer matrix resin could be dispensed from a certain location to cover the printed patterns; thus, a low viscosity and a fast UV curable and self-leveling silicone (SS-155) was used as an elastomer matrix resin. This UV curable silicone facilitated the continuous printing process because of its fast curing nature. On the other hand, choosing materials for the preparation of a 3D printable MR fluid was a great challenge. The rheological study was conducted to screen and select the materials as discussed in Section 5.3.

Figure 5.7: Schematic illustration for the printing of hybrid MRE via extrusion printing.

The extrusion pressure, feed rate, and initial height are regarded as printing parameters and their combinations played an important role in quality printing. The pressure applied to the piston-cylinder unit is regarded as the extrusion pressure. The extrusion pressure should be able to extrude the printing material smoothly and steadily out of the nozzle. The nozzle diameter and rheological properties of the printing materials (i.e. MR fluids) determined the optimum extrusion pressure. Lower viscosity materials require lower pressure to be extruded and vice versa. For the given viscosity, a smaller nozzle diameter requires a higher extrusion pressure and vice versa. Similarly, it was also found that the shear thinning material could easily produce continuous fluid filaments during extrusion.

Figure 5.8: (a) Cross-section of 3D printed hybrid MRE. (b) Illustration of printing parameters: extrusion pressure, initial height and feed rate for the extrusion-based printing of MR elastomer.

The optimized pressure allowed printing of the continuous fluid filaments out of the nozzle, while the rest two parameters also play a significant role in the quality of printed patterns. The initial height is the distance between the printing nozzle tip and printing platform while the feed rate is the movement rate of the printing platform in the XY plane as illustrated in Figure 5.8.

Figure 5.9 shows the printed filaments with two different nozzle sizes (800 μm and 500 μm of nozzle diameter) and the effects of the combinations of initial height and feed rate on the printing quality. The optimum extrusion pressures of the 800 μm and 500 μm nozzle sizes were 2.2 bars and 3.2 bars respectively. A smaller nozzle diameter requires

a higher extrusion pressure for the continuous extrusion. Several features can be observed in Figure 5.9.

(a) Nozzle size: 800 μm		Optimum pressure: 2.2 bars	
Initial height (mm) $\rightarrow$ Feed rate (mm/min) $\downarrow$	0.6	1	1.5
200
400
300
Optimum		0.8	
250

(b) Nozzle size: 500 μm		Optimum pressure: 3.2 bars	
Initial height (mm) $\rightarrow$ Feed rate (mm/min) $\downarrow$	0.4	0.6	0.8
150
250
200
Optimum		0.5	
200

Figure 5.9: Quality of printed patterns of filaments for the SS-3006T MR suspension and with different initial heights, feed rates, and nozzle diameters: (a) 800 μm and (b) 500 μm .

At low feed rate and low initial height (less than the nozzle diameter), flat filaments were printed, the flatness of the printed filament further increased as the feed rate and the initial printing height decreased. At a low initial height, the higher feed rate could produce circular filaments but after a certain speed, discontinuous filaments were

formed. At a higher initial height (for both low and high feed rates), the extruded filaments were either discontinuous or materials could not be separated from the nozzle tip. It was found that the best filaments were printed when the initial height was equal to the printing nozzle internal diameter. Finally, the optimum feed rate and initial height for the 800 μm diameter nozzle were obtained to be 250 mm/min and 0.8 mm respectively. Similarly, for the 500 μm diameter nozzle, the optimum feed rate and initial height were 200 mm/min and 0.5 mm respectively. These optimized printing parameters were utilized to print the various patterns inside the elastomer matrix.

Moreover, the printing parameters could be played to control the shape, width, and height of the printing patterns. As shown in Figure 5.10, dot patterns of diameter 1 mm to 2 mm and lines patterns of height 0.5 mm to 3 mm were printed. As can be seen in Figure 5.10(a), different sizes of dots could be printed with the same nozzle. The size of the dots was controlled by increasing the extrusion time without changing any other parameters. Similarly, multiple layers were printed continuously on the top of the previous layer to increase the height of the printed filament as shown in Figure 5.10(b). It was found that the printed filament could withstand its self-weight and remain stable till the sixth layer. In this way, the printing parameters could be utilized to control the dimensions of the printed patterns.

Figure 5.10: Controlling the shape, width and height of the printed patterns (with SS-3006T suspension); (a) dots patterns with different dot sizes and (b) line patterns with different heights.

Based on the CAD drawings, various patterns could easily be printed such as discrete (dots patterns) and continuous patterns. Various patterns of MR suspension were printed inside the elastomer matrix as shown in Figure 5.11. Discrete patterns are usually of dots

while the continuous patterns could be of any designs such as linear, circular and etc. The optimization of printing parameters has to be performed prior to printing the actual samples. The different patterns might need a different set of printing parameters. The feed rate and initial values would be different for printing discrete and continuous patterns. The feed rate would not be crucial for printing discrete patterns as the printing head dispenses materials on the distinct locations, so a high feed rate could be chosen to reduce the overhead time. However, various problems had to be resolved to achieve successful printing. A few common problems that were encountered during the printing process are illustrated in Figure 5.12. The problems include a discontinuous filament, unwanted flow, poor control over the shape and size of the filament and unstable layer.

Figure 5.11: Various 3D-printed hybrid MR elastomers with SS-3006T suspension patterns and elastomer matrix SS-155. (a) Dot pattern. (b) Line pattern. (c) Line pattern with mesh. (d) Asterisk shaped pattern. (e) Circular pattern.

The several optimizations have to be achieved by controlling printing parameters for the successful printing. The printing temperature, humidity, and other environmental conditions were kept the same for all the printing experiments. The printing temperature and the relative humidity were 25⁰ C and 40 % respectively. A low feed rate or low initial height or combination of them led to flat filaments with no patterns at all as illustrated in Figure 5.12(a) & (f). In this case, the initial height must be increased, while the feed rate must be decreased. Such problems can also occur due to the high extrusion pressure and low feed rate. So, the printing results as illustrated in Figure 5.12(f) were obtained because of the high pressure and low initial height at which the material was extruded while the nozzle was moving to another position. Secondly, the presence of the air bubble would result in a discontinuous filament with a shot of MR fluid in some locations as presented in Figure 5.12(b). Therefore, the printing cartridge must be air bubble free. A high feed rate and an initial height that exceeded the nozzle diameter led to discontinuous filaments with undesired printing as illustrated in Figure 5.12(c) & (e).

These kinds of problems can also occur at low extrusion pressures and higher feed rates. The reduction of feed rate and the initial height improved the printing quality. Moreover, the problem illustrated in Figure 5.12(e) is most likely to happen when the smaller nozzle is used it is because that the smaller filaments have the smaller surface area and are prone to weak bonding with the elastomer matrix. Lastly, it is possible that the CIPs presented in MR fluid patterns can be diffused to the elastomer matrix if the filament is subjected to some kind of compressive load before the elastomer matrix was not fully cured. This problem usually occurs after the printing if the matrix materials are not fully cured.

Figure 5.12: Examples of various problems occurred during the printing process and their resolution. (a) flattened filaments due to very low initial height; (b) discontinuous filament as air bubble was trapped within the printing material; (c) non-uniform filaments rather than continuous straight filaments because of the higher feed rate; (d) diffusion of magnetic particles between the adjacent filaments; (e) discontinuous filaments and weak bonding with the elastomer matrix leading to unwanted distribution of filaments; (f) jointed dots patterns rather than distinct dots.

The problems summarized in Figure 5.12 might occur during the printing of the first layer or in the mid-way. Therefore, it is not guaranteed that the flawless samples would be produced in a single shot. In fact, these problems occurred because of the improper combinations of printing parameters or conditions. Thus, careful optimization of the printing parameters is a very crucial phase of successful printing.

In summary, it has been verified that the shear thinning and thixotropic materials are required for extrusion-based 3D printing. The vulcanizing silicone (SS-3006T) and modified Lord MR fluid were used as printing materials for the development of hybrid MR elastomer (3DP-MREs) via 3D printing. Various patterns such as continuous filaments and discrete patterns were printed under the optimized printing condition. In

addition to rheological properties of the printing materials, a combination of the three-printing parameters including extrusion pressure, feed rate, and initial height, has a significant role in quality printing. In the subsequent sections: development and MR effect characterization of two different patterns namely, line and dot patterns 3DP-MREs will be presented in detail.

5.5 Line-patterned 3DP-MREs

Five different line patterns have been considered namely line, grid_45, grid_90, circle, and circle1 as presented in Figure 5.13. The MR fluid filaments were encapsulated layer by layer within the elastomer matrix. Internal configurations of patterns are shown in Figure 5.14. The 800 μm nozzle was used for the printing. After printing, the width of the printed filaments is found to be slightly expended laterally. The total volume of the printing filament was calculated, and all five patterns were printed with the same amount of the MR suspension. Thus, these five patterns have the same volume ratio of magnetic particles to the elastomer matrix. Both 3DP-MRE1 and 3DP-MRE2 samples were printed out.

Figure 5.13: Various line patterned filaments, namely (a) line, (b) grid_45, (c) grid_90, (d) circle and (e) circle1. Width of the printed filament (f).

The line patterns samples have three MR layers. The cross-sectional view of MR layers is given in Figure 5.14. The MR filaments in the MR layers for line, circle, and circle1 patterns were printed without offsetting the layers as given in Figure 5.14(a), whereas

the printed layers were offset by 45° and 90° for grid_45 and grid_90 patterns samples, respectively as given in Figure 5.14(b). Photograph of the 3DP-MRE samples ready for testing is given in Figure 5.15. All 3DP-MREs have 7 mm thickness and 28 mm diameter and consist of 3 MR layers.

Figure 5.14: Cross section views of the 3D printed samples. (a) line pattern (b) grid pattern (c) illustration of the size of the printed filament after the printing and SEM images.

Figure 5.15: Photograph of line patterned samples namely (a) line, (a) line, (b) grid_45, (c) grid_90, (d) circle and (e) circle1.

5.5.1 Cyclic Compression

The magnetic field was applied in the normal plane to printed layers in the cyclic compression test as shown in Figure 5.16. Like that is presented in **Chapter 4**, four

different currents (0, 1, 2 and 3 A) are applied to the electromagnet to investigate the magneto-mechanical properties of 3DP-MREs in cyclic compressive loading.

Figure 5.16: Illustration of the application of a magnetic field for 3DP-MREs in the cyclic compression testing.

Figure 5.17 shows the experimental results of cyclic compressive stress-strain hysteresis loops at 0.1 Hz and with the increasing magnetic field by increasing an applied current up to 3 A. A number of features become obedient from Figure 5.17. Firstly, when the magnetic field is not applied, 3DP-MRE1 samples exhibit higher stresses than 3DP-MRE2 samples, similarly, the area enclosed under the loading and unloading stress-strain curve is higher for 3DP-MRE1 than that of 3DP-MRE2. 3DP-MRE1 samples also possess a higher degree of non-linearity compared that to 3DP-MRE2 samples. When the magnetic field is applied, an MR effect can clearly be observed from Figure 5.17. The stress-strain curve moves upward, which signifies that the elastomer becomes stiffer. The movement of the stress-strain curve is more pronounced for 3DP-MRE2 samples, this is attributed to the free magnetic particles within the encapsulated fluid filaments of 3DP-MRE2 samples. Similarly, the area enclosed by the hysteresis curve is also found to be increased in the case of 3DP-MRE2 samples while the area enclosed by the stress-strain curves of 3DP-MRE1 samples was not significantly altered by the application of a magnetic field.

Figure 5.17: Compressive engineering stress versus strain for 3DP-MRE1 (left) and 3DP-MRE2 (right). The results presented are obtained with line patterned samples.

The MR effect for all five types of samples of both categories has to be studied in order to understand the effect of the patterning. That can be studied by obtaining the absolute and relative MR effect using the same method described in **Chapter 4**. Prior to that, the influences of strain rate and strain amplitude for both 3DP-MRE1 and 3DP-MRE2 samples are presented.

Figure 5.18: Strain amplitude effect without and with the application of a magnetic field for both 3DP-MRE1 (top figures) and 3DP-MRE2 (bottom figures).

The influence of strain amplitude on the response of the 3D printed MR elastomers at a constant frequency (0.1 Hz) and under two different magnetic flux densities (at 0 and 1 A current to the electromagnet) was studied. In the off-state (0.0 A current), 3DP-MRE2 samples acted like a normal spring-damper system as shown in the bottom left of Figure 5.18, where overlying hysteresis loops at various strain amplitudes can be observed. On the other hand, non-linear behavior can be observed for 3DP-MRE1 samples. The 3DP-MREs were less sensitive to the increasing strain amplitude in compressive loading when the magnetic field was not applied. However, in the on-state (2.0 A current), the hysteresis loops were enlarged and shifted slightly downward with increasing strain amplitude. As discussed earlier, like the effect of the increased magnetic field, the effect of strain amplitude is also more noticeable for 3DP-MRE2 samples than that of 3DP-MRE1 samples. The linear modulus or stiffness (slope of the curve) was found to

decrease with increasing strain amplitude. This is due to the breaking and restructuring of the free magnetic particle chains formed upon application of a magnetic field at higher strain amplitudes for 3DP-MRE2 samples. The particle chains started to yield, and chains might have buckled, thus increasing the distance between the freely suspended magnetic particles. This would be able to decrease the stiffness of MR elastomer. This strain amplitude dependent behavior suggests that the 3DP-MRE1 samples have the higher degree of non-linearity than that of 3DP-MRE2 samples in the absence of a magnetic field, while 3DP-MRE2 also behaves as a non-linear viscoelastic material under the application of a magnetic field.

Figure 5.19: Strain rate effect for 3DP-HMRE1 and 3DP-MRE2 samples without and with the application of a magnetic field.

The effect of strain rate from 4.2 to 42 mm/min in both off-state and on-state for both types of samples (3DP-MRE1 & 3DP-MRE2) is given in Figure 5.19. Strain rates 4.2, 8.4, 21 and 42 mm/min corresponds to a cyclic frequency of 0.05, 0.1, 0.5 and 1 Hz with a strain amplitude of 10%. As shown in Figure 5.19, increasing strain rate does not significantly alter the response of 3DP-MREs in both the absence and presence of a magnetic field. However, the stress-strain curve slightly moved downward with increasing strain rate, this might be explained as the field-induced formation of magnetic particle chains could not respond quickly to the higher strain rate. The effect of both strain amplitude and strain rate for core-shell hybrid MREs (**Chapter 4**) are similar to 3DP-MREs as core-shell samples also developed by encapsulating MR fluids.

For 3DP-MREs, in all further studies, the results are presented with a strain rate of 8.4 mm/min (0.1 Hz) at strain amplitude of 10 % (0.7 mm). The comparison of 5 types of samples (line, grid_45, grid_90, circle, and circle1) are carried out by obtaining the

absolute and relative MR effects. MR effects for both types of samples (3DP-MRE1 and 3DP-MRE2) are obtained using Equations 3.1 and 3.2 and are plotted against engineering strain as given in Figure 5.20. As the secant modulus is unbiased compared to linear modulus and tangent modulus, all MR effects are again obtained based on secant modulus from stress-strain curves, the same method was used in **Chapter 4**.

Figure 5.20: Absolute and relative MR effect of five different line-patterned 3DP-MREs versus strain at different amounts of current to the electromagnet.

The absolute and relative MR effects are plotted against the engineering strain and are given in Figure 5.20 for all 5 types of samples of two categories (3DP-MRE1 and 3DP-MRE2). MR effect is clearly observed with increasing current to the electromagnet. In addition, a number of features can be observed in Figure 5.20. Firstly, 3DP-MRE1 samples showed much lower MR effect compared to 3DP-MRE2 samples, again this can be attributed to free magnetic particles within the printed filaments of 3DP-MRE2 samples. Secondly, 5 different types of samples do not have a significant difference in

MR effects. Thirdly, MR effect is increased with increasing engineering strain and then become stable within 10% strain amplitude as that was obtained for core-shell hybrid MR elastomers (**Chapter 4**). Some negative MR effects were also observed, it is again attributed to the remnant deformation of the samples after the few cycles and hence causing some amount of plastic strain. However, MR effect is increased to positive value with increasing strain and become stable within 10% strain amplitude. The maximum values of the relative MR effect at three different currents to the electromagnet are determined using secant modulus within 10% strain for all five patterns. These values are summarized in Figure 5.21 for all five patterns of both 3DP-MREs.

Figure 5.21: Maximum values of relative MR effect for all five patterns at three different currents (1, 2 and 3 A) to the electromagnet. (a) 3DP-MRE1 (b) 3DP-MRE2 samples.

As expected, 3DP-MRE2 samples show higher relative MR effect than that of 3DP-MRE1 samples because of the free magnetic particles within the printed filaments. The maximum relative MR effect of 3DP-MRE1 samples is less than 0.1, whereas, relative MR effect can reach up to 0.6 for 3DP-MRE2 samples. It was found that the relative MR effect was not significantly affected by patterning printed filaments in the cyclic compression tests. This can be explained as the effect of the direction of an applied magnetic field, as the magnetic field was applied through the thickness of the samples and not along the plane of printed layers (Figure 5.16). Thus, the influence of patterning might have not observed. If the magnetic field is applied along the plane of the printed layers, the influence of patterning could be observed, which will be studied using a forced vibration technique.

5.5.2 Forced Vibration Results

The effect of line patterning can be studied deeply through forced vibration testing as the magnetic field can be applied in the various direction as shown in Figure 5.22. Three patterns, line, grid_45, and grid_90 could be a representation of anisotropic MREs while remaining two patterns (circle and circle1) is that of the isotropic MREs. The MR effect of the patterns (line, grid_45, grid_90) is expected to be different along the different direction of the magnetic field with respect to the printed filaments. Thus, they can be regarded as anisotropic MREs. On the other hand, the MR effect of the circularly patterned samples should not be influenced by the direction of the magnetic field.

Figure 5.22: Illustration of the application of a magnetic field with respect to the orientation of the printed filaments in the squeeze mode of analysis by forced vibration technique.

Figure 5.23: Magnitude transmissibility versus excitation frequency of the 3DP-MREs in a squeeze mode at various magnetic flux densities at three different directions for line patterned sample. 3DP-MRE1 (left) and 3DP-MRE2 (right).

The behavior of 3DP-MREs is expected to be different along with the different directions of the magnetic field with respect to the configuration of printed filaments. The magnetic field was applied in three different directions with respect to the orientation of printed filaments, (1) parallel (H_0), (2) oblique (H_{45}) and (3) perpendicular (H_{90}). Line patterned sample subjects to all three directions of the magnetic field,

grid_45 sample subjects to H_0 and H_{90} , grid_90 sample subjects to H_0 and H_{45} . Whereas circle and circle1 patterned samples subject to only H_0 . The application of the magnetic field is illustrated in Figure 5.22.

Figure 5.23 indicates the magnitude transmissibility for both 3DP-MRE1 and 3DP-MRE2 at various magnetic flux densities against the excitation frequency. Several features can be noted in Figure 5.23. The magnitude transmissibility curves shifted toward the right when the magnetic field was increased, which indicates a change in the natural frequency of the MRE system. The increase in the natural frequency of the MR elastomer system signifies the increase in stiffness of the 3DP-MREs. For 3DP-MRE1, the transmissibility curve shifted gradually but constantly toward higher frequency with increasing magnetic field. But, for 3DP-MRE2, shifting of the curve is gentler when a magnetic field is higher than 0.3 Tesla, similar results were achieved for core-shell hybrid MREs with fluid core (**Chapter 4**). Which is because the field strength around 0.3 Tesla is found to be optimal for MR fluid. On the other hand, 3DP-MRE1 has a higher natural frequency than that of 3DP-MRE2, which implies that the 3DP-MRE1 samples are stiffer. Which is true, as 3DP-MRE2 samples contain the viscous fluid filaments while 3D printed filaments are cured for 3DP-MRE1 samples. For both 3DP-MREs, the peak response amplitude also slightly increased with increasing magnetic flux density. The increase in the peak amplitude signifies a decreased damping ratio of the 3DP-MREs. Likewise, 3DP-MRE1 has higher transmissibility peaks compared to 3DP-MRE2 at resonance, meaning that 3DP-MRE1 has a smaller damping ratio than that of 3DP-MRE2. However, the shifting in the transmissibility curve was more pronounced for 3DP-MRE2 when the magnetic field was applied. This is again attributed to the free magnetic particles suspended in the fluid filaments. Moreover, the magnitude transmissibility values of both 3DP-MREs were significantly reduced near the resonance zone when a magnetic field was applied. This indicates that the 3DP MR elastomers also have the capability to attenuate the vibration amplitude. When the direction and amplitude of the magnetic field are varied, a significant difference is difficult to be observed from the transmissibility curves alone. Therefore, in order to study the effect of variation of magnetic field direction and patterning of the printed filament, elastic and damping properties are obtained using the method described in **Chapter 3**.

Figure 5.24 shows the stiffness and damping ratio for all 5 patterned samples of both categories (3DP-MRE1 and 3DP-MRE2) when the magnetic field is applied in various directions. For both types of 3DP-MREs, stiffness increased when the magnetic field was increased. The 3DP-MRE2 samples could be as stiff as the 3DP-MRE1 samples at the magnetic flux density of 500 mT. On the other hand, the damping ratio of 3DP-MRE1 is always smaller than the 3DP-MRE2 samples. Because the viscous MR fluid filament provides a higher capability to absorb the vibration amplitude for 3DP-MRE2 samples. For 3DP-MREs, the damping ratio was found to be almost constant or decreased with increasing magnetic flux density. This behavior is also exhibited by conventional MREs in previous studies.

Figure 5.24: Compressive stiffness and vibrational damping ratio for 3DP-MRE1 (top figures) and 3DP-MRE2 (bottom figures) of different patterns at the various orientation of magnetic flux direction plotted against magnetic flux density.

Relative MR effect as a function of magnetic flux density for all five types of the sample of both categories at various directions of magnetic flux density is given in Figure 5.25. The relative MR effect increased with increasing magnetic flux density and is higher for 3DP-MRE2 samples as compared to that of 3DP-MRE1 samples. The MR effect was also influenced when the direction of the application of the magnetic field was changed with respect to the orientation of the printed filaments. The MR effect was higher when

the applied magnetic field was parallel (H_0) to the orientation of the printed filaments. In this case, the magnetic particles would form a long chain (for 3DP-MRE2) or would interact (for 3DP-MRE1) along the whole length of the printed filament. While the magnetic particles would form shorter chains/interact only which are as big as the width of the printed filament when the applied magnetic field was normal to the orientation of the printed filaments. The longer the chains of the magnetic particles, the higher the magnetic interaction, thus resulting in a higher MR effect. This finding strongly suggests that the 3D printed MR elastomers have the capability to exhibit anisotropic behavior. In low magnetic flux region, such anisotropic behavior of 3DP-MREs also depends on printed patterns, line pattern samples showed the higher MR effect at H_0 direction than other patterns. This is because grid_45 or grid_90 patterns have one offset layer which is either subjected H_{45} or H_{90} when H_0 is applied. This is more visible in the case of 3DP-MRE2 samples as they have free magnetic particles in the printed filament. For 3DP-MRE2 samples, the MR effect was found not to be affected as much as it was in low magnetic flux region by the patterning at higher magnetic flux density (500 mT) because the particle chains might have become equally strong at higher magnetic flux density regardless of the patterns. On the other hand, for 3DP-MRE1 samples, the MR effect of grid_45 and grid_90 patterned samples is found to be slightly higher than other patterns and weakly dependent on the direction of magnetic flux density. This can be explained as the effect of the higher surface area along the plane of magnetic flux direction as can be visualized from Figure 5.22. The samples with grid_45 and grid_90 patterns have a higher surface area that is covered by printed filaments in the horizontal plane compared to other patterns.

The influence of the change in direction of the magnetic field with respect to the orientation of the printed filaments was studied in the squeeze mode. Subsequently, the MR effect of 3DP-MREs is also studied in a shear mode. Here, the main aim of the shear mode test is to study the behavior of the 3D printed hybrid MR elastomers when the direction of the magnetic field is varied with respect to the orientation of the printed layers. The magnetic field was applied laterally (0-degree) and normally (90-degree) to the printed layers, the detail of shear testing can be found in **Chapter 3**.

Figure 5.25: The effect of the different orientation of printed filament and, changes of relative MR effect at the various direction of the magnetic field as a function of the magnetic flux density.

The magnitude transmissibility curves in a shear mode for both 3DP-MRE1 and 3DP-MRE2 are presented in Figure 5.26. Again, the transmissibility curves shift toward the right when the magnetic field is applied, but the shifting of the curves is very gentler. The shifting of the transmissibility curve is more pronounced when the applied magnetic field is normal to the printed layers. Again, the 3DP-MRE1 system has a higher natural frequency and higher peak amplitude than that of the 3DP-MRE2 system.

Figure 5.26: Magnitude transmissibility versus excitation frequency for the 3DP-MREs in a shear mode at various magnetic flux densities. 3DP-MRE1 (left) and 3DP-MRE2 (right).

The stiffness and damping ratio of 3DP-MREs in the shear mode of operation were also obtained. They are presented in Figure 5.27. 3DP-MRE1 samples have a higher stiffness than 3DP-MRE2 samples and the stiffness of both types of samples is increased when a magnetic field is increased. Yet again, the 3DP-MRE2 samples showed a larger change in stiffness than samples of 3DP-MRE1. In contrast, 3DP-MRE2 has a higher damping coefficient than 3DP-MRE1. In shear mode, the damping ratio was also found to be slightly influenced by the application of a magnetic field for both types of 3DP-MREs.

The influence of the change in direction of the magnetic field can also be realized in Figure 5.27. When the applied magnetic field was normal to the printed layers, the magnetic particles had the opportunity to interact with the neighboring particles within the same layer as well as adjacent layers. Therefore, the MR effect was higher when a magnetic field was normal to the printed layers. This result also provides one more evidence that the 3D printed MR elastomers have an anisotropic feature. Put simply, the 3D printed MR elastomers are also influenced by the direction of the application of a magnetic field in both squeeze and shear modes.

Figure 5.27: Shear stiffness (left) and damping ratio (right) for 3DP-MRE1 and 3DP-MRE2 at different orientations and magnetic flux densities.

5.6 Dot-patterned 3DP-MREs

The printing of distinct dots is very similar to the ink-jet printing system. So, the printing can also be referred as the discontinuous printing process. Figure 5.28 illustrates discontinuous printing process to develop dot-patterned 3DP-MREs. This section explores the capability of the 3D printing to control the size, location, and distribution of MR fluid dots to produce various hybrid MR elastomers such as isotropic, anisotropic, BCC and FCC structures.

Figure 5.28: Schematic illustration of dot-patterned MRE fabrication via 3D printing method.

Firstly, a fixed volume of the MR fluid was divided into 5 different volumes, as 1, 1/4, 1/8, 1/16 and 1/32, henceforth, the 3D printed MR elastomers are named as Dot_1,

Dots_4, Dots_8, Dots_16, and Dots_32, respectively. All five samples have the same amount of the MR fluid printed however configured in a different way with disjoint dots as shown in Figure 5.29(a). The Dot 1 sample was produced by combining the four dots. The size and the location of MR fluid dots were controlled by controlling the printing pressure, initial height, feed rate and time. The crucial printing parameter for controlling the size of dots is the dispensing time. All other parameters (extrusion pressure, initial height, and feed rate) were kept constant and only dispensing time was controlled to control the size of the dot at the given location. Printing condition of five different samples is presented in Table 5.3. As can be seen from Table 5.3, the total time to print the MR fluid layer is the same, while the dispensing time can be seen in the geometric series for different samples (i.e. dispensing time per dot = $a \cdot r^{n-1}$, where $a=3$, $r=2$ and $n=1, 2, 3$, and 4 for Dots_32, Dots_16, Dots_8, and Dots_4 respectively). The SEM image of the cross-section of the 3D printed samples is presented in Figure 5.29(b). Similar to that of line patterns, here, the MR fluid dots are sandwich between the elastomer layers.

Figure 5.29: (a) Dot patterned 3DP-MREs with different sized MR fluid dots, Dots_32, Dots_16, Dots_8, Dots_4, Dot_1 and pure elastomer from right to left respectively and (b) Crossectional micrograph of 3DP-MRE under SEM.

Table 5.3: Summary of the printing parameters for Dots_32, Dots_16, Dots_8, Dots_4, and Dot_1 dot-patterned 3DP-MRE samples.

Sample	Extrusion Pressure (Bars)	Initial height (mm)	Feed rate (mm/min)	Dispensing time per dot (s)	Total time per MR fluid layer (s)
Dot_1	2.1	1.2	150	96	96
Dots_4	2.1	1.2	150	24	96
Dots_8	2.1	1.2	150	12	96
Dots_16	2.1	1.2	150	6	96
Dots_32	2.1	1.2	150	3	96

Figure 5.30: Variation of the size of the MR dots. (a) diameter and (b) height for Dots_32, Dots_16, Dots_8, Dots_4 samples.

As presented in Figure 5.30, the base of the printed dots is very close to a perfect circle and the size of dots is increased with increasing printing time. The size of the printed dots is analyzed based on two dimensions: diameter and peak height of the dots. The average diameter and average peak height of the dots are presented in Table 5.4. Both diameter and peak height are increased with increasing printing time. The curve fitting shows that both dimensions (diameter and height) increase almost exponentially with increasing time.

Table 5.4: Summary of the variation of the size of the printing dots: diameter and height.

Sample	Size of the dots	
	Diameter (mm)	Height (mm)
Dots_4	5.1 ± 0.2	1.8 ± 0.1
Dots_8	4.2 ± 0.15	1.6 ± 0.1
Dots_16	3.6 ± 0.1	1.4 ± 0.1
Dots_32	2.6 ± 0.1	1.2 ± 0.1

Figure 5.31: Morphology of the BCC and FCC structured dot-patterned 3DP-MREs.

In the second phase, to demonstrate the potential of 3D printing, the MR fluid dots were printed in such a way that the dots form basic crystal structures of the elements such as BCC and FCC. The morphology of 3D printed hybrid MREs with BCC and FCC structure is shown in Figure 5.31. The total volume of the printed MR fluid has been kept the same for both BCC and FCC structure. The MR fluid volume was again controlled by controlling the dispensing time, the information of the printing process is summarized in Table 5.5. The number of dots is higher in FCC structure and size of the dots are higher for BCC structure. 3 layers of MR fluid are printed.

Table 5.5: Summary of the printing condition for BCC and BCC structured dot-patterned 3DP-MREs.

Sample	Extrusion Pressure (Bars)	Initial height (mm)	Feed rate (mm/min)	Dispensing time per dot (s)	Total number of dots	Total time (s)
BCC	2.1	1.2	150	6	41	246
FCC	2.1	1.2	150	4	62	248

5.6.1 Cyclic Compression

As described in Section 5.5 for line-patterned 3DP-MREs, the same testing condition, and the method of analysis have been adopted to study the MR effect of dot-patterned 3DP-MREs. A typical stress-strain loop under cyclic compression is presented in Figure 5.32. In the absence of a magnetic field, 3DP-MRE1 has a higher modulus than that of 3DP-MRE2. Samples with different MR dots have a similar modulus for 3DP-MRE1,

while, for 3DP-MRE2 samples, it was also noted that the Dot_1 sample has a slightly lower modulus than others. Upon the application of a magnetic field, 3DP-MRE1 samples showed very small changes while the 3DP-MRE2 sample showed noticeable changes. Yet again, this is attributed to the free magnetic particles within the printed fluid dots of 3DP-MRE2 samples as they can freely move and make chains along the magnetic field.

Figure 5.32: Stress-strain loops for Dots_4 samples of two categories 3DP-MRE1 (left) and 3DP-MRE2 (right) at 3 different amounts of current to the electromagnet.

As expected, again, 3DP-MRE2 samples showed higher MR effect than that of 3DP-MRE1 samples. The MR effect is increased with increasing current to the electromagnet, with Dot_1 sample achieving the highest MR effects. This is attributed to the bulk volume of the MR fluid at one location. The maximal relative MR effect is given in Figure 5.33 for all five dot-patterned samples of both 3DP-MRE1 and 3DP-MRE2. For 3DP-MRE1 samples, the relative MR effect is smaller and is highest for Dot_1 sample and is decreased with an increasing number of dots. Similarly, for the 3DP-MRE2 category, the MR effect is also decreased as the size of the MR dots decreased and the MR effect becomes almost similar for Dots_8, Dots_16, and Dots_32 samples. The Dots_4 sample showed almost similar MR effect as that of Dot_1 sample, and the MR effect is considerably decreased (almost by 50%) for Dots_8, Dots_16, and Dots_32 samples. The experimental results suggested that the division of the given amount of MR fluid into smaller dots could uphold a reasonable MR effect. Such division is also beneficial when leakage and particle settling are concerned. The finding suggested that dividing the given volume of MR fluid into 4 dots can maintain a similar MR effect as that of a single MR dot. On the other hand, increasing the number of dots beyond 4 is not recommended if the MR effect is the main concern, however, can be considered if the leakage and sedimentation are the main concern.

Figure 5.33: Relative MR effect achieved with 1 A, 2 A and 3 A current and obtained using Equation 3.2 are listed for all five types of samples (Dot_1 to Dots_32) of two categories (a) 3DP_MRE1 and (b) 3DP-MRE2 samples.

Figure 5.34: (a) Illustration of isotropic and anisotropic samples and the application of a magnetic field. (b) Secant moduli for an isotropic and anisotropic sample of both category.

Thereafter, isotropic and anisotropic dot-patterned samples were also developed, by off-setting the specified printing location in the successive layers, picture and illustration are presented in Figure 5.34(a). The secant moduli obtained from the cyclic compressive testing for the isotropic and anisotropic samples are shown in Figure 5.34(b). For 3DP-MRE2, in the absence of the magnetic field, moduli of isotropic and anisotropic samples are similar, however, in the presence of a magnetic field, isotropic sample significantly lags the increase in modulus compared to the anisotropic sample. This result

demonstrates that the 3D printing method can also be used to develop isotropic and anisotropic MR elastomers. The maximum MR effect for the isotropic sample was found to be 0.23, where anisotropic sample showed the MR effect of 0.75, these values for 3DP-MRE2 samples.

Similarly, moduli for BCC and FCC structured samples of both categories in the absence and presence of a magnetic field are shown in Figure 5.35. The main aim of developing BCC and FCC structure is to study the MR effect of the same amount of the MR materials but configured differently inspired by basic crystal structures. Both BCC and FCC structure have the same amount of MR materials. Cyclic compression result revealed that BCC structure showed slightly higher MR effect than that of FCC structured 3DP-MREs. This is believed that the effect of size of the MR dots, the BCC patterns have bigger dots and, therefore, showed higher MR effect.

Figure 5.35: Results of the BCC and FCC structure 3D printed MR elastomer of both categories, (a) 3DP-MRE1 and (b) 3DP-MRE2

4.2.2 Forced Vibration Testing

MR effects of dot-patterned 3DP-MREs were also studied through forced vibration testing similar to that of line patterned samples. A typical result of the force vibration testing of dot-patterned 3DP-MRE is presented in Figure 5.36(b). The magnetic field was applied as illustrated in Figure 5.36(a). Here, the magnetic field is parallel to the plane of the printed layers. A number of features can be noted in Figure 5.36(b). Firstly, when the magnetic field is not applied, the transmissibility curves of the two types of samples (3DP-MRE1 and 3DP-MRE2) are easily distinguishable. Similar to that of line patterned samples, 3DP-MRE1 samples have higher natural frequency and higher peaks at the natural frequency. Higher natural frequency signifies that 3DP-MRE1 samples are

stiffer than that of 3DP-MRE2 samples and higher peak of transmissibility curve indicates that the damping ratio of the 3DP-MRE1 system is smaller than that of the 3DP-MRE2 system. Upon the application of a magnetic field, the transmissibility curves are moving towards the right. Shifting of the curves towards right indicate that stiffness of the 3DP-MRE is increased. Again, the shifting of the curve is more pronounced for 3DP-MRE2 samples. Yet again, like that of 3DP-MRE2 line patterned samples, this must be attributed to the free magnetic particles that are suspended within the dots of 3DP-MRE2 samples while magnetic particles are locked within the elastomer of 3DP-MRE1 samples. On the other hand, the damping ratio of the samples are almost constant or slightly decreased with the increasing magnetic field as the peak of the transmissibility curve is almost same or increased upon the application of a magnetic field.

Figure 5.36: (a) Illustration of the direction of the application of the magnetic field with respect to the printed layers. (b) Magnitude transmissibility versus excitation frequency of the dot-patterned 3DP-MREs (Dots_4 samples) at various magnetic flux densities, 3DP-MRE1 (left) and 3DP-MRE2 (right).

The effect of changing the size of the dots are discussed by obtaining the stiffness and damping ratio as given in Figure 5.37. When the magnetic field is not applied, it was found that the 3DP-MRE1 samples are stiffer than that of 3DP-MRE2 samples. Similarly, the 3DP-MRE1 samples have a low damping ratio than that of 3DP-MRE2 samples. Dots_1 sample was found to be least stiff compared to other samples for 3DP-MRE2 samples and the stiffness was found to be increased as the size of MR dots decreased. This can be attributed to the size of the dots; as the smaller fluid dots could

behave like viscoelastic solid dots. On the other hand, for 3DP-MRE1 samples, the stiffness was found to increase up to 8 dots and start to decrease with an increasing number of dots. This can be attributed to the soft elastomeric casing where the presence of smaller dots could not significantly alter the stiffness of the elastomer matrix. Upon the application of a magnetic field, MR effect exhibited by 3DP-MRE1 samples was found to be much smaller compared to 3DP-MRE2 samples and the effect of size of the dots are not significant. Whereas for 3DP-MRE2, MR effect was found to be decreased with an increasing number of dots. The dot_1 sample has relative MR effect of 77% while dots_32 sample only has 28% at 500 mT. However, the difference between dots_4 and dot_1 sample is not very significant, dots_4 has 70% MR effect at 500 mT. When the same amount of volume of MR fluid is divided into the different amount of volume the MR effect gets affected. Nonetheless, it is worthy that if dots are divided into four parts MR effect is not significantly decreased. This is also one of the reasons that the printing of four dots are considered to develop isotropic and anisotropic samples.

Figure 5.37: Stiffness and damping ratio versus magnetic flux density five types of dot sample of both 3DP-MRE1 and 3DP-MRE2.

In the cyclic compression, the magnetic field was applied normal to printed layer as given in Figure 5.34(a). Whereas, in the forced vibration testing, the magnetic field is

applied as given in Figure 5.38(a), which is to observe the difference of isotropic and anisotropic configuration in MR effect when magnetic field is applied parallelly to the printed layers. The stiffness achieved via frequency sweep with isotropic and anisotropic samples of both 3DP-MRE1 and 3DP-MRE2 is given in Figure 5.38(b). The MR effect exhibited by the isotropic and anisotropic 3DP-MREs is found to be similar when the magnetic field was applied parallel to the printed layers. Yet again, the MR effect is higher for 3DP-MRE2 samples. This result provides the evidence that the anisotropic nature chiefly depends upon the direction of the applied magnetic field and 3D printing has the capability to configure the MR fluid or magnetic particles in a unique fashion.

Figure 5.38: (a) Illustration of the direction of the applied magnetic field. (b) Stiffness versus magnetic flux density isotropic and anisotropic samples of both 3DP-MRE1 (left) and 3DP-MRE2 (right).

Lastly, as achieved in cyclic compression testing, the significant difference of BCC and FCC structure was not observed through a vibration testing even when the magnetic field was applied parallel to the printed layers. However, BCC structures seemingly showed slightly higher MR effect than that of FCC, which is again attributed to the higher size of MR dots as described previously. Even though the difference in MR effect was not observed by printing the basic crystal structures such developments yet again provide the capability of a 3D printing method, which is difficult to be performed by other fabrication techniques.

5.7 Dynamic Properties of 3DP-MREs Under Uniaxial Deformation

The goal of this section is to investigate the dynamic properties of hybrid MRE samples developed in this study. The 3DP-MREs samples with line pattern were chosen. The 3DP-MRE1 samples also represent the core-shell hybrid MREs with a solid core and 3DP-MRE2 samples represent the core-shell hybrid MREs with a fluid core that is presented in **Chapter 4**.

The dynamic properties of line patterned 3DP-MREs were obtained by means of forced vibration testing in a squeeze mode, and the magnetic field was applied parallel to the printed layers which is also parallel to the printed filaments as given in Figure 5.39.

Figure 5.39: Illustration of the direction of the application of a magnetic field (left) and model of 3DP-MRE (right) in a single DOF system.

An absorber mass is attached to the 3DP-MRE sample and the base is mounted to the shaker. The motion is unidirectional in the z -direction (normal to the printed layer). The dynamic properties of 3DP-MREs were analyzed using the frequency sweep by obtaining the magnitude and phase transmissibility. The testing is conducted in the range of frequency from 50 to 500 Hz. Samples are tested with three different input acceleration and four different magnetic flux densities. The accelerations are 0.25g, 0.5g and 0.75g and magnetic flux densities are 0 mT, 110 mT, 300 mT and 500 mT.

5.7.1 Analysis Method

The shaker control software allows the user to view the results as the test is running. Separate files containing the information on the input and output data can be saved. These files are then used to achieve the storage modulus and loss factor of the 3DP-MRE test samples.

The following equation of motion, stress-strain relation, and force-stiffness relation are used for the analysis [235].

$$\ddot{y} + \frac{k_e^*}{m}(y - x) = 0 \quad (5.9)$$

$$\sigma = G^* \gamma \quad (5.10)$$

$$F = k_e^* (y - x) \quad (5.11)$$

The star (*) indicates that the variable is complex. $\ddot{y}$ is the acceleration and y is the displacement of the absorber mass (m), k_e^* denotes the stiffness of 3DP-MRE, x is the displacement of the shaker base, G is the elastic modulus of the 3DP-MRE, σ represent the compressive stress, γ represent compressive strain and F is the force to the 3DP-MRE sample.

As the stiffness of an elastomer can be changed with excitation frequency, the natural frequency of the system also changes. For the frequency sweep, the above equations can be deployed to find the natural frequency of the system, the storage modulus and the loss factor of the 3DP-MREs as follows:

$$\delta = -\frac{\tan(\varphi)}{\frac{T}{\cos(\varphi)} - 1} \text{ for } \varphi > 0 \quad (5.12)$$

$$f_n = f \sqrt{\frac{\tan(\varphi) - \delta}{(1 + \delta^2)\tan(\varphi)}} \quad (5.13)$$

$$G' = \frac{mH(2\pi f_n)^2}{A} \quad (5.14)$$

where δ is loss factor, T is the magnitude of transmissibility, φ phase angle of system, f is excitation frequency, f_n is the natural frequency, G' is the storage modulus, H is thickness and A is the area of the MRE sample.

Using Equation 5.12, 5.13 and 5.14 the storage modulus and the loss factor are obtained from the experimental data.

5.7.2 Results and Discussion

Figure 5.40 shows a typical transmissibility plot. The specific areas of interest are circled in the plot. The first area of the interest is the natural frequency (transmissibility peak) of the system. It varies for each sample (3DP-MRE1 and 3DP-MRE2) and depends on

the magnetic field. The second reason of interest occurs in the zone of 150-200 Hz. This is caused by the noise in the system. The final area of interest occurs above 400 Hz. This abnormality even occurs from 300 Hz for 3DP-MRE1 sample. The irregularities in the region can be attributed to the noise in the system or non-linearity of the sample. “The irregularities in the different regions have a direct effect on the storage modulus and loss factor of the 3DP-MRE. As can be seen in 5.41, the transmissibility peak makes the sudden dip on the storage modulus in both the absence and presence of a magnetic field. Also, the loss factor is also affected in a similar manner. The noise from the shaker also has a direct effect on the storage modulus trend. Lastly, above 300 Hz, the non-linearity of the samples provides the jumping of both storage and loss factor trends.

Figure 5.40: The transmissibility curves of 3DP-MRE2 sample. The areas are circled to show where the system is affected by noise.

The abovementioned irregularities have a direct effect on the loss factor and storage modulus. The first region (transmissibility peak), affects both the storage modulus and loss factor. In that region, there is a slight dip in the storage modulus, while the loss factor trend either has a dip or sudden rise. The noises in the system also cause sudden dip or rise in both storage modulus and loss factor trends. Jumping of loss factor can be seen when the frequency is above 400 Hz. Nonetheless, the general trend can be studied with the help of these transmissibility curves.”

5.7.2.1 Effect of Magnetic Field on the Dynamic Properties of 3DP-MREs

Figure 5.41 shows testing results for both 3DP-MRE1 and 3DP-MRE2 samples at various magnetic flux densities. In the absence of a magnetic field, 3DP-MRE1 sample possess higher storage modulus than that of 3DP-MRE2 sample. While the loss factor is higher for the 3DP-MRE2 sample. This is because of the presence of the viscous fluid filament in the 3DP-MRE2 sample. As can be seen in Figure 5.41, both the storage modulus and loss factor are affected when the magnetic field is applied. The storage modulus increased with increasing magnetic flux density for both types of samples. It can be noted that the effect of the magnetic field is more pronounced for 3DP-MRE2 sample and is true because of the presence of MR fluid filaments.

Figure 5.41: Storage modulus and loss factor for 3DP-MREs at various magnetic flux densities. (a) storage modulus and (b) loss factor for 3DP-MRE1. (c) storage modulus and (d) loss factor for 3DP-MRE2.

The storage modulus of the 3DP-MRE2 sample can be as high as that of the 3DP-MRE1 sample in the presence of a magnetic field. The effect of the magnetic field is like a step function for 3DP-MRE2 samples, which means that the storage modulus immediately jumps to higher values when a magnetic field is applied and keep increasing slowly with a further increment of a magnetic flux density. On the other hand, the effect of the

magnetic field is like a ramp function for 3DP-MRE1 sample, which means that the storage modulus keeps steadily increasing with increasing magnetic flux density.

For the 3DP-MRE2 sample, the loss factor is found to be decreased when the magnetic field is applied as encapsulated MR fluid filaments become a semi-solid state in the presence of a magnetic field. For 3DP-MRE1 sample, loss factor is found not to be altered by the application of a magnetic field. Lastly, the 3DP-MRE2 sample is more affected by the noise from the system, which can be visualized by the disturbance on the trend of the storage modulus and loss factor. Again, this must be attributed to the encapsulated MR fluid filaments of 3DP-MRE2 sample.

5.7.2.2 Effect of Frequency on the Dynamic Properties of 3DP-MREs

For the dynamic effect of frequency on 3DP-MREs, please refer to Figure 5.41 again. The general trend is that both the storage modulus and loss factor increase as the frequency increases. This trend can be explained with the help of two reasons. First, MREs are viscoelastic materials, indicating that the materials' properties of 3DP-MREs are dependent on the strain rate. The strain rate of the system is directly proportional to the excitation frequency for a constant strain amplitude. Thus, the viscoelastic properties of 3DP-MREs are dependent on the frequency. When the frequency is increased material becomes stiffer, which result in an increase in the storage modulus. This is a well-understood phenomenon of the viscoelastic materials.

The second reason for the effect of frequency on the properties of 3DP-MREs is related to the change in strain amplitude with a change in frequency. For viscoelastic materials, the storage modulus is inversely proportional to the strain amplitude. This is also called the Payne effect. The strain amplitude can be achieved using Equations 5.26.

$$\gamma = \frac{|Y_{input} - Y_{output}|}{H} \quad (5.26)$$

$$Y_{input} = \frac{\ddot{Y}_{input}}{(2\pi f)^2} \quad (5.27)$$

$$Y_{output} = Y_{input} T \quad (5.28)$$

Where γ is strain amplitude, Y_{input} is the position of sample, Y_{output} position of the mass, H is the thickness of the sample, $\ddot{Y}_{input}$ is the input acceleration, f is the

excitation frequency and T is the magnitude transmissibility obtained as a ratio of output to input acceleration.

Figure 5.42: The effect of frequency on the strain amplitude in dynamic testing. The graphs represent the 3DP-MRE2 sample in both the absence and presence of a magnetic field.

Figure 5.42 shows a decrease in the strain amplitude for frequencies higher than the natural frequency of the system. According to the Payne effect, the decrease in the strain amplitude causes an increase in storage modulus. Put together, the storage modulus increases because the viscoelastic properties of 3DP-MREs are dependent on the excitation frequency and because of the strain amplitude changes with excitation frequency.

5.7.2.3 Effect of Acceleration on the Dynamic Properties of 3DP-MREs

To study the effect of acceleration, experiments are conducted with three different amplitude of acceleration as 0.25g, 0.5g, and 0.75g. The input acceleration amplitude does affect the modulus of the 3DP-MREs in both the absence and presence of a magnetic field. Figure 5.43 shows the effect of acceleration on the storage modulus of 3DP-MREs. The storage modulus decreases as the acceleration amplitude increases. This must be attributed to the strain amplitude effect. Figure 5.43 shows how the strain amplitude is related to the acceleration. The strain amplitude increases as the acceleration increases. Therefore, the storage modulus is decreased with increased acceleration.

Figure 5.43: The storage modulus of 3DP-MREs at different acceleration level (0.25g, 0.5g, and 0.75g) in the absence and presence of a magnetic field. Storage modulus of 3DP-MRE1 (a) 0 mT and (b) 500 mT. Storage modulus of 3DP-MRE2 (c) 0 mT and (d) 500 mT.

3DP-MRE1 is equally affected by the increasing acceleration in both the absence and presence of a magnetic field. On the other hand, 3DP-MRE2 is less affected by the acceleration in the absence of the magnetic field, but the effect is almost similar to that of 3DP-MRE1 in the presence of a magnetic field. This is because of the MR fluid filament in the 3DP-MRE2 sample change to a semi-solid state in the presence of a magnetic field. So, the Payne effect is more visible. Lastly, as the frequency increases the strain amplitude becomes almost similar for a different level of acceleration (Figure 5.44), thus, the Payne effect is less pronounced. In other words, storage modulus comparatively less affected by the increasing acceleration level at higher frequencies (above 200 Hz).

Figure 5.44: The effect of acceleration on the strain amplitude in dynamic testing. The graph represents the 3DP-MRE2 sample in both the absence and presence of a magnetic field at three different accelerations (0.25g, 0.5g, and 0.75g).

5.8 Summary of the Chapter

The feasibility of a 3D printing technology for the fabrication of MR elastomers has been studied in detail. An extrusion-based multi-material printing process was implemented to develop two types of hybrid MR elastomers. The rheological properties of the printing materials played a vital role in extrusion-based printing. It was found that the shear thinning, and thixotropic behavior are highly desired for printing materials. The successful printing also strongly depends on the combined effect of the key printing parameters including extrusion pressure, feed rate, and initial height. The most crucial printing parameter was the initial height and it was found that the initial height should be equal or slightly higher than that of the internal diameter of the printing nozzle. On the other hand, the printing size of dot-patterns can also be controlled by controlling the printing time.

Using a multi-head printer, two patterns of MR fluid filaments, namely, line-patterned and dot-patterned samples were developed with a different configuration of MR fluid within the elastomer matrix. Development of line patterns samples can be referred to continuous printing while the dot-pattern printing is a discontinuous printing. In other words, for line patterns, the nozzle continuously dispenses the materials in the printing path, while, for dot patterns, nozzle only dispenses materials in the specified points. Developments of unique structures such as anisotropic line patterns, grid patterns, various dot patterns and basic crystal structures such as BCC and FCC provide the capability of a 3D printing method, which is difficult to be performed by other

fabrication techniques by solely applying a magnetic field during the fabrication process.

It was demonstrated that the 3DP-MREs can change elastic and damping properties when exposed to the external magnetic field. The stiffness of the 3DP-MREs was found to be increased with an increasing magnetic field. The change in the properties was more pronounced for 3DP-MRE2 than for 3DP-MRE1, which is because of the presence of the MR fluid filaments. Furthermore, 3DP-MREs exhibited an anisotropic behavior when the direction of the magnetic field was varied with respect to the orientation of the printed filaments or printed dots and printed layers. Put simply, the 3DP-MRE2, which consist of MR fluid filaments within the elastomeric matrix is a bridge material between MR fluids and MR elastomers.

Lastly, dynamic testing showed that both the storage modulus and loss factor of the 3DP-MREs are depended on the strength of the magnetic field, excitation frequency, and strain amplitude. Effect of the magnetic field is again more pronounced for 3DP-MRE2 while the effects of frequency and strain amplitude are similar for both 3DP-MRE1 and 3DP-MRE2. Storage modulus was found to be increased as the strength of the magnetic field or frequency is increased, while decreased with increasing strain amplitude. Thus, the hybrid MREs developed in this study are also viscoelastic materials as that of conventional MREs.

Chapter 6: A New Type of Vibration Isolator Based on MRE

This chapter presents the development of a new type of MRE-based vibration isolation. The isolator has been developed by implementing the preload and magnetic field simultaneously.

6.1 Introduction

Base isolation is one of the efficient ways to reduce unwanted vibration to prevent structural damage. Active and semi-active isolators have been developed by incorporating the magnetic field-responsive materials such as magnetorheological (MR) fluids and MR elastomers into the structures or systems as described in the literature section (**Chapter 2**). These smart materials, MR fluids and MR elastomers help to mitigate the unwanted vibration by changing their elastic and damping properties under the external magnetic field. Thus, the reduced amplitudes of acceleration or displacement are transmitted to the structure because of the integrated MR materials. However, MR fluids have limitations due to leakage and sedimentation problems. Thus, in recent decades, MR elastomer-based isolation or absorption systems have gained considerable attention owing to its advantages over MR fluid such as no leakage and sedimentation.

MR elastomer-based isolators shift the natural frequency or resonance zone of the system upon the application of a magnetic field. Such isolators also have the capability to attenuate the transmitted vibration amplitude by changing the stiffness and damping capability of MR elastomers under the magnetic field. A number of studies dealing with the development of the MRE-based isolation systems are available in the literature (**Chapter 2**). Researchers have developed various kinds of active or semi-active MRE-based isolators/absorbers by coupling the MR elastomer either a single mode (shear or squeeze) or a shear–squeeze mixed mode. The mixed mode is found to be a more effective way to change the natural frequency of the MRE based isolators as compared to a single mode of operation. However, most of the MRE- based devices include a narrow working frequency range, high power consumption, and a bulky configuration.

Thus, there is a need to find an efficient method to enhance the performance of the MRE-based isolator that is free from the inherent problems of current isolators.

For the single DOF system, there are two ways to shift the natural frequency of the MRE-based isolator; (1) apply higher magnetic fields so that MR elastomer would change its stiffness until the magnetic saturation of the magnetic particles is reached and (2) increase or decrease the amount of payload. They are illustrated in Figure 6.1. The first one needs a strong magnetic field that usually makes a bulky system and requires higher power if an electromagnet is used, while the second one needs a dynamic system in order to have a variable payload.

Figure 6.1: Illustration of a single degree of freedom MRE-based isolation system and for the shifting of the transmissibility curve with increasing magnetic field or payload, where each shifting direction is indicated by the arrow.

When the magnetic field is applied, the magnetic interaction among the magnetic particles increases the stiffness of the MR elastomer, thus the natural frequency is increased and therefore the resonance zone will shift toward the higher frequency. On the other hand, the increased payload always shifts the natural frequency toward lower frequency range and vice versa. At the same time, the increased payload provides a preloading effect to the MR elastomer and thus MR elastomer would become stiffer and again the resonance zone shifts to the higher frequency range. Therefore, the combined effect might be seemingly neutral or significant. In the previous studies, such a combined effect of the magnetic field and preloading effect have not been considered in developing MRE-based isolators or absorbers.

It should be noted that the increasing or decreasing the amount of payload to control the preloading might be a troublesome act in the actual system. Thus, there is a need for an efficient way to change the amount of payload. The magnetic attraction force can be very powerful if the ferromagnetic materials are brought near to the strong magnets. Thus, the basic idea here is to use the magnetic attraction force between the strong magnet and ferromagnetic material to increase or decrease the amount of payload in the MRE-based isolator. Therefore, the amount of preloading can be controlled easily. At the same time, the MR elastomer has to be exposed to the magnetic field.

In this chapter, the approach is to simultaneously apply a magnetic field and preloading to study the vibration isolation behavior of an MRE-based isolator. A simple MRE-based isolator has been developed, where various magnetic flux densities can be generated, and at the same time, the amount of payload can be changed to produce different levels of preloading. The magnetic attraction force has been used as a source to change the amount of payload. Therefore, when a different magnetic field is applied the preload will be changed accordingly. Prior to studying the behavior of the isolator, the preloading effect at the various magnetic field strengths for the MR elastomer is presented under a cyclic compression loading. Thereafter, the combined effect of the preloading and magnetic field in the MRE-based isolator is studied in detail in the frequency range of 50-800 Hz.

6.2 Experimental Apparatus and Method

The experimental apparatus connected to the vibratory shaker is shown in Figure 6.2. The MRE-based isolator consists of the magnetic field generator, MR elastomer, and absorber mass. The absorber mass has a ferromagnetic base in order to generate the magnetic attraction force when a magnetic field is applied to control the amount of preloading. The base of the isolator could be connected to the shaker. The shaker provides the sinusoidal excitation from the base. Two accelerometers were used to measure the excitation and response signals of the isolator.

Prior to studying the behavior of the MRE-based isolation system, the preloading effect for MR elastomer was investigated under cyclic compressive loading. Four different preloading levels (0, 0.2, 0.4 and 0.6 mm) to the MR elastomer without and with the

application of a magnetic field are considered. Tests with 0 mm level represent the condition without preload.

Figure 6.2: Ensemble configuration of the MRE-based isolator and schematic representation of the single degree of freedom MRE-based isolator.

Figure 6.3: Graphical representation of the frequency sweep (50-800 Hz) at a constant acceleration amplitude of 0.5 g.

Thereafter, the behavior of the MR elastomer-based isolator was studied via a frequency sweep test. The excitation frequency has a constant peak acceleration amplitude of 0.5g over 50-800 Hz frequency as shown in Figure 6.3. The combined effect of the magnetic field and preload was studied at four different magnetic flux densities (0, 190, 320, and 520 mT). These different magnetic fields would provide different preloads to the MR elastomer system. The relationship of the magnetic flux density and preload is presented in Table 6.1. The preload values presented here exclude the weight of the absorber mass. The weight of the absorber mass is 290 grams.

Table 6.1: Preloads at the various magnetic flux densities.

No.	Magnetic flux density (mT)	Preload (N)
1	0	0
2	190	10.5
3	320	31
4	520	53.9

6.3 Results and Discussion

Figure 6.4: Engineering stress as a function of engineering strain under different preloads (0, 0.2, 0.4 and 0.6 mm) at cyclic compression at 0.1 Hz frequency. Various magnetic flux densities applied are: (a) 0 mT, (b) 190 mT, (c) 320 mT and (d) 520 mT.

First, the preloading effect for the MR elastomer was studied under the cyclic compression loading without and with the application of a magnetic field. Figure 6.4 depicts the preloading effect (four different level 0, 0.2, 0.4 and 0.6 mm) on the response of the MR elastomer at different magnetic flux densities and at a constant peak to peak displacement of 0.5 mm and a frequency of 0.1 Hz. A few observations from Figure 6.4 become evident. First, increasing preload made the stress-strain curves separated. This result signifies that the preloading has a direct effect on the MR elastomer's behavior. Similarly, the curves become steeper and the area enclosed by the hysteresis loops slightly increased with increasing preloading. Thus, it can be claimed that both the stiffness and damping capability of the MR elastomer increased with increasing preload because the slope and area under the force-displacement curve represent stiffness and

damping capability, respectively. Second, in the absence of a magnetic field, the shift of preload curves seems to be relatively greater than that in the presence of a magnetic field (190, 320 or 520 mT) when a preload was applied. This indicates that the influence of the preload was more prominent when a magnetic field was not applied. This can also be realized from Figure 6.5 when the maximum stresses are compared at various magnetic field strengths and preloads. Finally, the curves without a preload (0 mm curves) are continually softer while the curves with a 0.6 mm preload are continually stiffer.

Figure 6.5: Comparison of the stress of the MR elastomer to compress by 7% strain at various preloads and different magnetic fields.

Figure 6.5 shows the influence of preloads on the amount of stress developed to when the MR elastomer is compressed by 7% strain under the various magnetic flux densities. The experimental results revealed that the preload has a significant effect on the response of the MR elastomer under both conditions, without and with the presence of a magnetic field: the peak stress was significantly increased. The maximum increment at 0.6 mm level of preloading and 520 mT magnetic field was observed to be 4.2 times higher than the load required at no preload and no magnetic field to compress the MR elastomer sample for the same strain level (i.e. 7 %). When the strength of the magnetic field was increased, the influence of preload was found to be rather lesser compared to its influence without the application of a magnetic field. This could be explained as the field-induced particle-particle interaction among the magnetic particles would have already made the elastomer stiffer in the presence of the magnetic field. Thus, the higher amount of load is required to deform the MR elastomer to the same extent as it does in

the absence of a magnetic field. The cyclic compression results strongly showed that the preloading has a significant effect on the MR elastomer performance.

Figure 6.6: Magnitude and phase transmissibility are of the MRE-based isolator as a function of excitation frequency under the various magnetic flux densities.

Now, the vibration isolation behavior of the MRE-based isolator is presented. Figure 6.6 shows the magnitude transmissibility and phase transmissibility curves for the MRE-based isolator at various magnetic flux densities in the frequency range of 50-800 Hz without and with the preloading effect. At the peak amplitude of the magnitude transmissibility curve, the phase angle between response signal (y) and excitation signal (x) is $\pi/2$ and the corresponding excitation frequency represents the natural frequency (ω) of the single DOF system. Here, magnitude transmissibility is obtained as the ratio of acceleration signals of response mass and exciter and phase difference is simply the difference of phase angle between response and excitation signals.

Several features can be noted in Figure 6.6. The system has one maximum peak on the magnitude transmissibility curve. This signifies that the MRE-based isolator is a single

DOF system. Both frequency and phase transmissibility curves are moving toward higher frequency range when magnetic flux density is increased. But the shifting of the transmissibility curve is significantly enhanced when preloading and magnetic field both are simultaneously applied. Similarly, the peak amplitude of the phase transmissibility curve increased when the magnetic flux density and payload were increased. The increase in the peak amplitude signifies that the damping ratio of the isolator is decreased. However, for the curves of 320 mT seems to be influenced by the noises from the environment or the shaker itself and not followed the trend. Nonetheless, the peak is higher than when compared to no preload and no magnetic field curve. Moreover, the amplitude of the vibration has been significantly reduced in the resonance zone when only the magnetic field is applied or preload and magnetic field both are simultaneously applied. Again, the influence of the combined preload and the magnetic field is dominant.

When only the magnetic field is applied, the MRE-based isolator changed the natural frequency due to an increase of the stiffness of the MR elastomer only via particle-particle interaction of the magnetic particles under the magnetic field. In the case when both the magnetic field and preload were applied simultaneously, there is not only the magnetic interactions but also a preloading effect provided by the magnetic attraction force to the MR elastomer, making the MR elastomer even stiffer. Therefore, the natural frequency of the system is boosted by the preloading as well. There is no doubt that the magnetic interaction between the magnetic particles would always increase the natural frequency. However, the magnetic interaction would be even higher at higher preloads as the distance between the magnetic particles decreases when the preload is applied as illustrated in Figure 6.7. Thus, as a result of a combined effect, the speedy shifting of the transmissibility curve is observed.

Figure 6.7: Illustration of the reduction of the particle-particle distance under preloading.

The peak of the magnitude transmissibility curves represents the natural frequency of MRE-based isolator while the stiffness of the MR elastomer can be obtained using Equation 3.3. They are presented in Figure 6.8. Both the natural frequency and stiffness are increased with increasing magnetic field as well as the boosting of preloading can be seen. The natural frequency was shifted to 123 Hz (1.56 times) from 68 Hz when an only magnetic field (520 mT) was applied. However, the natural frequency was found to be increased to 390 (4.97 times) when both preload (53.9 N) and magnetic field (520 mT) are simultaneously applied. The maximum increase in the natural frequency was found to be 4.97 times higher than that of its original natural frequency in the combined loading condition which is 3.4 times higher than the one achieved with the application of a magnetic field only. On the other hand, the relative increment of the stiffness was significantly higher as high as 550 times of its zero-field stiffness when both magnetic field and preload are simultaneously applied while the change was only 3.63 times when only the magnetic field was applied. Therefore, the relationship between stiffness and the magnetic flux density is expressed in the semi-log graph (Figure 6.8 (b)) in order to have a clear picture of the significant increment of the stiffness. This can be explained with the help of Equation 6.2. If the zero-field natural frequency of the MRE based system is high, the smaller change in the natural frequency would make a significant change in the stiffness of the MR elastomer. Thus, a relatively small change (4.97 times) in the natural frequency resulted in the very high (550 times) change in the stiffness of an MR elastomer when the magnetic field was applied.

Figure 6.8: (a) The natural frequency of the MRE-based isolator and (b) stiffness of the MR elastomer under the combined application of both magnetic field and preload.

$$\Delta f_n \approx \frac{1}{2} \frac{\Delta K}{f_n} \quad (6.2)$$

where f_n is the natural frequency at zero field, Δf_n is the change in the natural frequency of the MR elastomer system and ΔK is the change in the stiffness of the MR elastomer when a magnetic field is applied.

Furthermore, a substantial influence of the magnetic field can be seen in the resonance zone. In other words, the acceleration amplitude is significantly attenuated in the resonance zone when the magnetic field is applied. Figure 6.9 shows the reduction of the acceleration amplitude in the resonance zone when a magnetic field is applied. The vibration amplitude is reduced by more than 60% when the applied magnetic field was just 190 mT. This signifies that the combined loading condition can also significantly attenuate the vibration amplitude under resonance even in the moderately strong magnetic field. Such dynamic responsiveness of the new type of isolator demonstrates the ability to be used for the highly adaptive vibration control system.

Figure 6.9: Reduction of the acceleration amplitude at the zero-field resonance frequency (68 Hz) of the MRE isolator when a magnetic field is applied.

Based on the findings, Figure 6.10 shows a potential method to develop active vibration system based on MREs. As illustrated in Figure 6.10, if the transmissibility curve (black curve in Figure 6.10) without magnetic field is combined with the curve (purple curve in Figure 6.10) with simultaneous application of magnetic field and preload, an efficient isolator could be developed. The isolator has to be developed in such a way that it should be subjected to both preloading and the magnetic field below a certain frequency (called critical frequency, as marked by a black dotted line in Figure 6.10). After the critical frequency, isolator should work without the application of a magnetic field. The resulted output of the isolator can be described by the red shaded section in Figure 6.10. The adoption of the method would result in an active vibration isolation system with

minimum transmissibility coefficient even in the resonance zone with a lower strength of the magnetic field.

Figure 6.10: The proposed method for the best isolation by combining zero field and magnetic field plus preload curve. The red shaded portion shows the efficient isolation zone.

In this study, the shifting of the transmissibility curve is experimentally boosted using permanent magnets as a semi-active isolator system. Additionally, it has been successfully demonstrated that the higher magnetic field is not necessarily required for developing efficient isolation system. Therefore, even the usage of the electromagnet to generate a magnetic field requires lesser power at the same time the system becomes compact. It should be noted that in order to develop an active isolation system, the magnetic field from the electromagnet are the best because the control system can easily be integrated into the isolator to control the current and thus the magnetic field.

6.4 Summary of the Chapter

In this chapter, a simple setup to study the behavior of an MRE-based isolator system where both the magnetic field and preload could be applied simultaneously has been presented. It was demonstrated that the MRE-based isolator can quickly shift the natural frequency even at the moderately strong magnetic field. The combined effects of the magnetic field and preloading are attributed to providing such a substantial shifting of the transmissibility curve. The result suggests that the method adopted in this work opens a new door to develop highly tunable MRE-based isolators which work in the low magnetic field range.

Chapter 7: Conclusions and Future Works

This chapter summarizes the work presented in this thesis, draws conclusions and provides recommendations for future works.

7.1 Conclusions

This study concentrated on the behavior of hybrid MR elastomers developed by utilizing the MR fluid or MR elastomer and change in properties when the magnetic field was applied. The main aims of the thesis are to provide a potential solution to bridge the gap between MR fluid and MR elastomer and lower the strength of the magnetic field required for current MRE-based devices. The works presented in this thesis can be divided into three sections: first, development of hybrid MR elastomers using a core-shell structure, second, introduction of 3D printing method for the development of hybrid MR elastomers and lastly, development of MRE based vibration isolator by simultaneous utilization of preload and magnetic field.

Core-shell hybrid MREs were developed by a conventional method called molding. In the conventional method, a cavity of an elastomer matrix was formed by molding and MR materials were deposited into the cavity to form an MR core within the elastomer matrix. Three types of MR core were considered, low viscosity fluid core, high viscosity fluid core, and solid core. On the other hand, in the 3D printing method, hybrid MREs were developed by a layer-by-layer technique, MR fluid filaments were encapsulated layer by layer within the elastomeric matrix using a multi-head 3D printer. This study presented a 3D printing method for the development of hybrid MR elastomer for the first time. The main challenges were materials selection, selection of suitable printing method and finally the successful printing. The fluid filament of MR materials was to be printed; therefore, extrusion-based printing remained the ideal choice. Taking the advantage of rheology, the printing materials were selected, printing materials must have highly shear thinning and thixotropic nature. The printing parameters such as initial height, feed rate, extrusion pressure and time played a vital role for successful printing, optimization of these printing parameters is necessary for different types of patterns and structure to be 3D printed. 3DP-MRE1 and 3DP-MRE2 are two different categories of 3D printed hybrid MR elastomers, 3DP-MRE1 consists of solid MR filaments and 3DP-

MRE2 consists of fluid MR filaments. Two types of unique patterned were considered in this study, namely, line-patterned and dot-patterned 3DP-MREs. The line-patterned samples were formed by continuous printing while the dot-patterned samples were formed by discontinuous printing.

The hybrid MREs developed by both conventional and 3D printing methods were characterized using a cyclic compression and through a force vibration testing in both absence and presence of a magnetic field. Lastly, the dynamic properties of hybrid MREs under uniaxial deformation were also investigated.

Chapter 4 described the development and characterization of core-shelled soft hybrid MR elastomers. The soft hybrid MR elastomers with two types of fluid cores and a solid core with four different concentrations of magnetic particles have been fabricated and their magneto-mechanical properties have been investigated. It was found that the solid core samples showed relatively smaller or no MR effect, whereas the MR effect was increased with increasing concentration of magnetic particles for fluid core samples. The highest MR effect was observed for 80 wt.% concentration of magnetic particles. But the MR effect can be enhanced by the addition of MNPs. The addition of the MNPs was proven to be significant in terms of both MR effect as well as the stability of the core fluid. The experimental results showed a clear increment of modulus, energy dissipation, absolute and relative MR effects of the soft hybrid MR elastomers with increasing magnetic flux density. Forced vibration testing results also showed a clear increment in the stiffness with increasing magnetic flux density while the damping coefficient was found to be not significantly altered by the magnetic field.

From the magnetorheological study, it was found that the high viscosity carrier fluid is not as good as a low viscosity carrier fluid for the shear stress and viscosity enhancement. However, for the core-shell soft hybrid MR elastomer with the fluid core, the finding suggested that the MR effect exhibited by high viscosity carrier fluid core is as good as the low viscosity carrier fluid core or even higher (for low concentration of CIPs). For core-shell structure, it was also found that the MR effect was relatively lower for low concentrations (20 & 40 wt.%) of magnetic particles, however, was not the case when the only MR fluid property was studied via magneto rheology. The high viscosity carrier fluid is also superior to low viscosity carrier fluid when the sedimentation and

leakage are concerned. The high viscosity fluid core soft hybrid MR elastomers are free from the sedimentation issue of MR fluids of the core. It was also demonstrated that such hybrid MREs also exhibit anisotropic MR effect as conventional anisotropic MREs when the direction of the applied magnetic field is changed as the mobile particle within the core can easily move in the direction of magnetic flux. Moreover, the addition of MNPs is also proven to be significant as they enhanced sedimentation behavior for low viscosity carrier fluid and also improved the MR effect of core-shell hybrid MR elastomers. The soft hybrid MR elastomer with fluid core with either high viscosity carrier fluid or enhanced viscosity of low viscosity carrier fluid with MNPs are proven to be potential materials to bridge the gap between MR fluid and MR elastomer. Such fluid core hybrid MR elastomers provide good resistance to flow and sedimentation of magnetic particles within the core, as well as they, exhibit anisotropic MR effect. The core-shell soft hybrid MR elastomer is capable of changing its stiffness even in a moderately strong magnetic field thus such elastomers also offer the lower working range of the magnetic field.

For core-shell hybrid MREs, the following specific conclusions can be drawn from the experimental investigation:

- The fluid core samples show higher MR effect than that of solid core samples, which is because of the presence of the free CIPs within the MR core.
- The MR effects of core-shell hybrid MR elastomers increase as the concentration of CIPs increases.
- Even though pure magnetorheological studies reveal that the high viscosity fluid is not as good as that of low viscosity fluid for shear stress and viscosity enhancement, high viscosity fluid core exhibits competitive or even better MR effects (for low concentration) for core-shell hybrid MREs. In other words, the MR effect of core-shell MREs is not significantly depended on the viscosity of the carrier fluid of the MR core.
- The stiffness of the core-shell MR elastomers increases while damping ratio decreases or remains the same as the strength of magnetic flux increases.
- Core-shell hybrid MRE with high viscosity fluid core avoid the sedimentation issue of the current MR fluid and show higher MR effect than that of current MREs and also exhibits anisotropic MR effect.

- Put simply, core-shell hybrid MRE with high viscosity fluid MR core can be considered as a bridge material between MR elastomer and MR fluid.
- The MR effect of core-shell hybrid MREs can be enhanced by adding the MNPs.
- Addition of MNPs increases the rheological properties, stability of MR fluid and MR effect of core-shell hybrid MREs

Chapter 5 provided the feasibility of a 3D printing technology for the fabrication of MR elastomers in detail. An extrusion-based multi-material printing process was implemented to develop two types of hybrid MR elastomers. The rheological properties of the printing materials played a vital role in extrusion-based printing. It was found that the shear thinning, and thixotropic behavior are highly desired for printing materials. The successful printing also strongly depends on the combined effect of the key printing parameters including extrusion pressure, feed rate, and initial height. The most crucial printing parameter was the initial height and it was found that the initial height should be equal or slightly higher than that of the internal diameter of the printing nozzle. On the other hand, the printing size of dot-patterns can also be controlled by controlling the printing time.

Using a multi-head printer, two patterns of MR fluid filaments, namely, line-patterned and dot-patterned samples were developed with a different configuration of MR fluid within the elastomer matrix. Development of line patterns samples can be referred to continuous printing while the dot-pattern printing is a discontinuous printing. In other words, for line patterns, the nozzle continuously dispenses the materials in the printing path, while, for dot patterns, nozzle only dispenses materials in the specified points. Developments of unique structures such as anisotropic line patterns, grid patterns, various dot patterns and basic crystal structures such as BCC and FCC provide the capability of a 3D printing method, which is difficult to be performed by other fabrication techniques by solely applying a magnetic field during the fabrication process.

Lastly, dynamic testing showed that both the storage modulus and loss factor of the 3DP-MREs are depended on the strength of the magnetic field, excitation frequency, and strain amplitude. Effect of the magnetic field is again more pronounced for 3DP-MRE2 while the effects of frequency and strain amplitude are similar for both 3DP-MRE1 and

3DP-MRE2. Storage modulus was found to be increased as the strength of the magnetic field or frequency is increased, while decreased with increasing strain amplitude. Thus, the hybrid MREs developed in this study are also viscoelastic materials as that of conventional MREs.

Following specific conclusions can be drawn for 3DP-MREs:

- The 3DP-MRE2 samples always show a higher MR effect than that of 3DP-MRE1 samples, this is again attributed to free CIPs within MR fluid filaments.
- Again, the stiffness increases with increasing magnetic flux and damping ratio slightly decreased or remain the same with increasing magnetic flux for both 3DP-MRE1 and 3DP-MRE2 samples.
- 3D printing method has the potential to develop unique and anisotropic patterns such as line patterns, grid patterns, dot patterns, BCC and FCC structures.
- 3DP-MREs shows anisotropic nature when the direction of the applied magnetic field is changed with respect to the printed filaments or plane of the printed layers.
- Line pattern MR elastomer exhibits anisotropic MR effect when the direction of the applied magnetic field is parallel to the plane of printed filaments.
- Dot-patterns MR elastomers exhibit anisotropic MR effect when the direction of the applied magnetic field is normal to the plane of printed dots.
- Dot-patterned 3DP-MREs are an advanced form of core-shell hybrid MR elastomers, the leakage or contamination of one of the MR dots does not affect the other MR dots.
- The sedimentation and settling of CIPs are highly unlikely within the small MR fluid filaments and 3D printing method can develop a different configuration of MR fluid or CIPs without applying a magnetic field. Putting together, 3DP-MREs are also the bridge materials between MR fluid and MR elastomers.
- Lastly, core-shell hybrid MR elastomer with fluid core or 3DP-MRE2 also offer the low working range of magnetic field, the increase in the stiffness is much gentler as the applied magnetic flux density exceeds 300 mT.

Lastly, Chapter 6 presented a new method for the development of the MRE based vibration isolator which works in the low magnetic field range. The simultaneous implementation of preload and magnetic field resulted in the boost of performance of

the isolator. MRE being the viscoelastic material, preloading significantly affects the MRE performance, so the performance of MRE-based vibration isolator was enhanced even in low magnetic field strength. A simple setup to study the behavior of an MRE-based isolator system where both the magnetic field and preload could be applied simultaneously has been presented. It was demonstrated that the MRE-based isolator can quickly shift the natural frequency even at the moderately strong magnetic field. The combined effects of the magnetic field and preloading are attributed to providing such a substantial shifting of the transmissibility curve. The result suggests that the method adopted in this work opens a new door to develop highly tunable MRE-based isolators which work in the low magnetic field range.

7.1 Future Works

Based on the current work, when characterizing the MRE materials, and when designing applications using MRE in the future, a number of recommendations can be given:

- While characterizing the MRE materials in compressive or tensile loading, the Mullins effect plays a vital role, therefore, always consider preconditioning the sample before the actual test.
- The MR effect is higher in the low strain level, therefore, when characterizing in different operation modes such as squeeze, tension, and shear modes and when designing applications using MRE materials, low strain level ($< 10\%$) can be considered.
- The force vibration testing method used in this study can be implemented to characterize the MRE materials as this testing method does not deform the samples.
- The hybrid MR elastomers developed in this study are to be tested under fatigue loading condition in both the absence and presence of a magnetic field.
- The novel approach of the development of MRE materials using 3D printing in this study can further be explored, other potentials of 3D printing technique such as shape memory effect can be studied in the future.
- 3D printing method can be implemented to develop lattice structures such as Kagome and MRE-based sandwich structures.

- The simulation of the 3D printed samples can be performed to optimize the filament size, orientation and finally the MR effect.
- The morphing structures is another possibility which can be developed using 3D printing of MREs.
- Obtaining the dynamic properties such as storage modulus and loss factor of magnetorheological elastomers using the forced vibration method is highly recommended as this method do not deform the sample and can be performed in a wide frequency range (10-1000 Hz).
- The novel approach of boosting the performance of MRE-based isolator by the simultaneous application of preload and magnetic field can be recommended for the development of MRE-based devices, additionally, it is also recommended to combine this technique with other control logic such as on-off and fuzzy logic for the development of MRE-based devices.
- Boosting effect of the isolator at constant magnetic field and varying preload can be studied.
- Future MRE based isolator can be developed as per the recommendation provided in Figure 6.10 in Chapter 6.

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