JOURNAL OF MATERIALS RESEARCH AND TECHNOLOGY 2023;23:1285–1295

Available online at www.sciencedirect.com

jmr&t Journal of Materials Research and Technology

journal homepage: www.elsevier.com/locate/jmrt

Original Article

Improvement of rheological and transient response of magnetorheological grease with amalgamation of cobalt ferrite

S.M.A. Tarmizi [a], N.A. Nordin [a,*], S.A. Mazlan [a], U. Ubaidillah [b,], S.A.A. Aziz [c], N. Mohamad [d], N.M. Hapipi [e]**

[a] Engineering Materials and Structures (eMast) IKohza, Malaysia-Japan International Institute of Technology (MJIIT), Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100 Kuala Lumpur, Malaysia

[b] Department of Mechanical Engineering, Faculty of Engineering, Universitas Sebelas Maret, Surakarta 57126, Indonesia

[c] Faculty of Applied Sciences, Universiti Teknologi Mara (UiTM), Cawangan Pahang, Kampus Jengka, 26400 Bandar Tun Abdul Razak, Jengka, Pahang, Malaysia

[d] Faculty of Engineering, Universiti Malaysia Sabah, Jalan UMS, 88400, Kota Kinabalu, Sabah, Malaysia

[e] Physics Department, Center for Foundation Studies in Science, Universiti Malaya, 50603, Kuala Lumpur, Malaysia

ARTICLE INFO

Article history:
Received 13 October 2022
Accepted 24 December 2022
Available online 28 December 2022

Keywords:
Magnetorheological grease
Cobalt ferrite
Additive
Rheological properties
Transient response

ABSTRACT

The time responsiveness of magnetorheological grease (MRG) towards magnetic field stimulus is crucial in order to ensure the high performance of MR devices. However, due to the thixotropic properties of grease, MRG has been restricted in terms of responding rapidly towards these magnetic fields. Therefore, polygonal shapes made up of 1–3 μm of cobalt-ferrite ($CoFe_2O_4$) particles with different concentrations from 0 to 5 wt.% were introduced to enhance the responsiveness of the MRG. The results revealed that the linear viscoelastic (LVE) region of the modified MRG improved between 29% and 43% during the off-state and on-state conditions, respectively. The absolute MR effect of MRG increased by at least 60% due to the improvement in the particle's chain alignment with that of the applied magnetic fields. In terms of transient responses, particularly within the highly acceptable LVE region (0.05%), the MRG with $CoFe_2O_4$ performed about 5–6 s faster as compared to pure MRG, which was attributed to the improvement in the particle's mobility in the grease medium.

* Corresponding author.
** Corresponding author.
E-mail addresses: nurazmah.nordin@utm.my (N.A. Nordin), ubaidillah_ft@staff.uns.ac.id (U. Ubaidillah).
https://doi.org/10.1016/j.jmrt.2022.12.154

1. Introduction

Magnetorheological (MR) materials have received great attention due to their controllable properties when subjected to magnetic fields. This includes properties such as the stiffness, the damping factor, the viscosity and yield stress. Prior to these changeable properties against external magnetic fields, MR materials have been applied in many advanced engineering applications such as in semi-active dampers [1,2], clutches [3], brakes [4,5], and actuators [6,7]. Magnetorheological grease (MRG) was proposed by Rankin et al. [8] which basically consisted of dispersions of CIP in a grease medium to overcome the demerits and drawbacks of the MR fluid (MRF). It was found that with the thixotropic properties of the grease as a matrix medium in the MRG which, it compensated to improve the initial low viscosity of the MRF and prevented the magnetic particles from being suspended at the bottom when there was no force exerted on the material [9–11]. Despite these advantages, this smart grease (MRG) still possesses a slower transient response towards the stimuli as compared to MRF. Higher viscosity was exhibited by the fibrous structure of the grease, which significantly restricted the mobility of magnetic particles within the matrix phase [12,13]. The behavior then disrupted the reaction of magnetic particles towards the induced magnetic field during the on-state condition (presence of magnetic field), which finally resulted in a slower response of the rheological properties of the MRG towards the stimuli. At times, the MR devices would undergo lags and hiccups, which indirectly limit the performance of the device. Typically, the time response of the MRF was less than 1 ms, which indicated the high responsiveness of the MRF when subjected to the applied stimulus [14]. Numerous studies related to the transient responses of the MRF also found that this smart fluid tends to react with the stimulus within the range of 1–100 ms, depending on various factors, including the induced field strength, particle composition, as well as the type of tests utilized [15–18].

Generally, literature reviews related to the transient response of the MRG are noticeably limited. Today, the transient response of MRGs typically uses different shapes of CIP, which are plate-like and spherical shapes [19]. The results summarized that the MRG with plate-like CIPs had much better rapid responsiveness, and recorded a higher storage modulus of ~2.5 MPa when the magnetic field was applied to the MRG, as compared to the spherical-CIP MRGs. This finding was due to the larger surface contact area and sensitive edges possessed by the plate-like CIP, which strengthened the formation of the particle's chain alignment and simultaneously improved the magnetic responsiveness of the particles towards the magnetic field stimuli in the grease medium [20]. However, no specific value for the transient responses of both the MRGs were stated in the study. An alternate new discovery found that the MRG with plate-like CIPs performed at a slower transient response rate (6 s). The spherical-CIP MRGs and bi-dispersed MRGs, particularly ones mixed with CIPs made up of plate-like and spherical shapes, exhibited a notable but not much quicker rate which was 5 s when applied a strain value of 0.01% [21]. Nevertheless, as the applied strain increased to 0.5%, the transient responses for all the MRG samples were improved by 2 s. Literally, the improved transient responses were due to the destruction of the chain structures of the MRG, as the applied strain was beyond its linear viscoelastic (LVE) region, where the limitation region for the material to behave visco-elastically returned to the easing of the mobility of the particles in the grease medium. However, the improvement in the transient responses of the MRG by using different shapes of CIPs is still unsatisfactory compared to the responsiveness itself, which is considerably slower. In fact, the findings were not clear and did not state the use of either spherical, plate-like, or mixed shapes of CIPs, which would lead to the improvement in the transient responses of the MRGs.

The use of additives might be much more practical to improve the MRG's transient responses. Previous studies have shown that the use of additives could improve the stability dispersion, viscosity, shear stress, storage, and loss modulus of the MRG. For instance, through the introduction of chromium oxide (CrO_2) in the mixture of the MRF and grease, it was found that the CrO_2 acted as the stabilizing agent in the mix matrices by creating a barrier between the CIP in the medium [22]. There was also a study related to the use of graphite (Gr), with Gr as an additive in the MRG. The results showed that the storage modulus of the MRG was improved by about 126% when 0.8 T was applied to the MRG [12]. Molybdenum disulfide (MoS_2), which was used as an additive, was able to enhance the viscosity of the MRG as the applied temperature and magnetic fields increased [23]. The stated result was due to the agglomeration and monolayer formation of the MoS_2 which obstructed the built-up heat among the CIPs once temperature was induced to the MRG. Despite of the excellent properties of the MoS_2 in the MRG, the agglomeration issue of the particles during the shearing process was seen to degrade the performance of the material, especially when subjected to long-term use of the material. In another study, cobalt ferrite ($CoFe_2O_4$) particles have been previously utilized as an additive with 50 wt.% of CIP in the MRG, and with 1 wt.% of $CoFe_2O_4$. The corresponding viscosity in the off-state condition was reduced by about 86% [13]. This finding was due to the presence of the $CoFe_2O_4$ which caused the detangling of the fibrous structures of the grease during the shearing process which subsequently assisted the particles inside of the grease to move freely as compared to the condition without the $CoFe_2O_4$. On the contrary, during the on-state conditions, or conditions induced with magnetic fields, the shear stresses and yield stress of the MRG were improved and attributed to the magnetic properties which were also owned by the $CoFe_2O_4$, which enhanced the responsiveness of the magnetic particles (CIP and $CoFe_2O_4$) towards the applied magnetic field. This finding showcased the enhancement of the MRG through the incorporation of $CoFe_2O_4$ particles. Scholars have focused on the effects of these additives on the rheological properties of the MRG.

Therefore, in this study, $CoFe_2O_4$ particles were utilized in the MRG as an additive in order to improve the transient response towards the applied magnetic fields. Considering the fact that transient response is one of the important parameters that will affect the resultant mechanical properties particularly torque, the study is vital since MRG has high potential to be used for the torque sensor applications. In fact, most sensor devices rely on materials that would exhibit high sensitivity and rapid responsiveness towards specific stimuli. Besides, previous studies also have been reported on the use of $CoFe_2O_4$ additive that would affect the mechanical and rheological properties of various materials. Owing to aforementioned matter, in this study, six samples of MRGs with different concentrations of $CoFe_2O_4$ particles particularly 0 to 5 wt.% were prepared, and the samples were characterized in terms of its magnetic properties and morphological analysis. Then, the rheological properties of MRG samples were tested under dynamic oscillatory shear mode test, with respect to the input parameters of sweep strains and sweep magnetic fields. The influence of both parameters on the resultant transient response of the MRGs with different concentrations of $CoFe_2O_4$ were discussed thoroughly. It is expected that the $CoFe_2O_4$ as an additive will significantly enhance several properties, including, but not limited to the rheological properties of the MRG. It is also hoped that it will facilitate the responsiveness towards the applied magnetic fields, resulting in the improvement of the MRG as compared to previous studies.

2. Materials and methodology

2.1. *Preparation of MRG*

Pior to the preparation of MRGs with additives, $CoFe_2O_4$ particles were synthesized using a co-precipitation method having a 68.8 emu/g magnetization value [13]. The main magnetic filler,i.e., the spherical-shaped CIP (CC-typed) from BASF Germany, was selected, as it has an average value of 1–10 μm. For the matrix medium of the MRG, the commercial lithium-based grease (Nippon Koyu Ltd, Japan) was used. It exhibited a density and viscosity of 0.92 g/cm and 0.207 Pa s, respectively. The magnetic properties for the magnetic particles were analyzed using a Vibrating Sample Magnetometer (VSM) within the range of the magnetic field from −15kOe to 15 kOe under room temperature. X-ray diffraction (XRD) pattern of the $CoFe_2O_4$ was investigated with X-ray Diffractometer (Empyream, Pan Analytical) from 20° to 80° of Bragg's angles.

In order to fabricate the MRG, the grease was firstly stirred by using a mechanical stirrer (Multimex High-Speed Dispersed) for 5 min to break down its internal structures (fibrous structures), before 50 wt.% of CIP was added to the stirred grease. The $CoFe_2O_4$ particles were introduced as an addtivive into the mixture incrementally at 1 wt.%, from 0 up to 5 wt.%. The mixture was continuously stirred for 2 h with a constant speed of 300 rpm until the homogeneity of the mixture was achieved. The pure MRG without the addition of $CoFe_2O_4$ was denoted as MRG, while the MRG-1 to MRG-5 samples consisted of different concentrations of $CoFe_2O_4$ which were labeled accordingly, as tabulated in Table 1.

2.2. *Characterization and testing of MRG*

The morphological characterization of MRG samples was examined using environmental scanning electron microscopy (ESEM, Quanta FEG 450) at magnifications of 500× and 2500×. The distribution of the $CoFe_2O_4$ particles within the CIP in the MRG were analyzed. The rheological properties of the MRG added with $CoFe_2O_4$ particles in different concentrations were tested using an oscillatory shear mode for sweep strains and sweep magnetic fields, at the off-state and on-state conditions using an Anton Paar parallel-plate rheometer (MCR 302, Austria). A 1 mm of gap distance between the upper and lower plates was set, with the sample being placed in between the plates. The diameter of the parallel plate used was 20 mm (type PP20/MRD/T1/P2). All the testing was carried out at room temperature at 25 °C. Before investigating the transient response of the MRG samples, it was important to understand the limitation of the MRG in terms of its linear viscoelastic (LVE) region by plotting the resultant moduli versus shear strains, from 0.001% to 10%. For the magnetic field sweep test, the MRG samples were tested within the range of the magnetic flux density, from 0 to 0.6 T. This was done by adjusting the applied currents from 0 A to 4 A. These tests were carried out in order to analyze the dynamic properties of the MRG samples and the deformation phase of the materials under different conditions of inputs parameters and magnetic field excitations. The transient response of each MRG sample was carried out by setting up the frequency of 1 Hz and applying magnetics flux densities of 0 T (off-state) and 0.6 T (on-state) in order to inspect the time taken for the MRG to respond towards the applied stimulus. After each test, the sample was demagnetized to remove the remanence in the sample. Each test was repeated thrice to acquire reliable and consistent data.

3. Results and discussion

3.1. *Characterization of samples*

A homogenous dispersion of magnetic particles, including CIP and $CoFe_2O_4$ in the suspending medium of grease is essential in order to improve the magnetization properties of MRG. ESEM images were carried out focusing on the distribution of magnetic particles in the MRG, as shown in Fig. 1(a) and (b). As observed, the magnetic particles of CIP and $CoFe_2O_4$ are homogeneously dispersed in the grease medium. It can be seen that the shape of $CoFe_2O_4$ particles are in polygonal shape, distributed within the CIP that are in spherical shape. In fact, the size obtained for both particles are in the range of 1–5 μm which is agreeable as provided by the supplier (CIP) and from the precipitation method ($CoFe_2O_4$) as well.

Table 1 – Compositions of MRG samples.

Composition (wt.%)	MRG	MRG-1	MRG-2	MRG-3	MRG-4	MRG-5
CIP	50	50	50	50	50	50
Grease	50	49	48	47	46	45
$CoFe_2O_4$	0	1	2	3	4	5

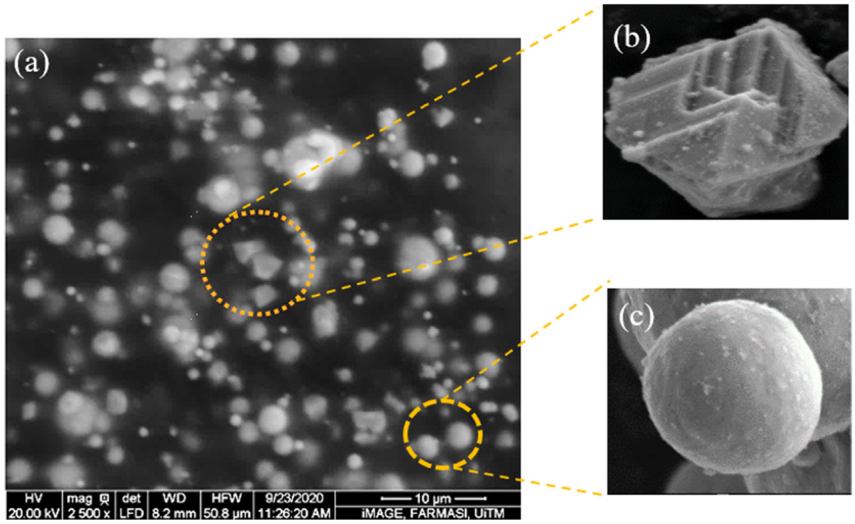

Fig. 1 – Morphological structure of MRG with $CoFe_2O_4$ under the magnification of 2500×; for (a) homogeneous dispersion of both CIPs and $CoFe_2O_4$ particles in the grease medium while, (b) polygonal shape of $CoFe_2O_4$ and (c) spherical shape of CIP.

The magnetization hysteresis loop for the magnetic particles are found to be 68.6 emu/g and 130 emu/g for $CoFe_2O_4$ and CIP, respectively as presented in Fig. 2 (a) and (b). It showed that both are soft magnetic particles with a small coercivity loop. Basically, the magnetization of CIP is higher than the $CoFe_2O_4$ due to the higher content of Fe in the particles. However, the magnetization value of $CoFe_2O_4$ can be considered high due to the high temperature applied during the sintering process. In fact, the heat energy gained during the sintering causes the crystallites to grow and join together thus forming larger particles. Particle size is directly proportional to magnetic domain size. As the magnetic domain size increases, the atomic spin numbers increase correspondingly towards the applied magnetic field, thus the saturation magnetization increases. Meanwhile, the coercivity and retentivity of the $CoFe_2O_4$ obtained are 16.397 Oe, and 0.45 emu/g, respectively. For CIP, the coercivity and retentivity is 24.03 Oe and 3.96 emu/g, accordingly. The magnetic properties of the particles have been tabulated in Table 2.

In order to determine the crystal structure of $CoFe_2O_4$, XRD is carried out in Fig. 3. All the peaks of the $CoFe_2O_4$ were discovered with sharp and narrow lines which signify high purities of crystallinity with space group Fd-3m. The peaks were observed on crystallographic planes (220), (311), (222), (400), (422), (511), (440), (620), and (533), which correspond to Bragg's angles of 30.2°, 35.5°, 37.2°, 43.2°, 53.5°, 57.1°, 62.7°, 71.1°, and 75.2°, respectively, demonstrating the formation of single-phase cubic spinel structure $CoFe_2O_4$. The most prominent peak of the $CoFe_2O_4$ at $2\theta = 35.55°$ emerge in the pattern of synthesized particles. According to Bragg's equation, the lattice constant obtained is 8.378 Å with Miller indices of 311 which is in line with other researchers [24,25].

3.2. *Effect of sweep strains on the rheological properties of MRG*

The main criterion which can be extracted from the graph of the storage modulus, i.e., the G′ versus shear strain, is the linear viscoelastic (LVE) region, which represents the range at which the testing was carried out without destroying the structure of the sample. Basically, a lower applied shear strain was noted below 0.1%. The values beyond this are known as

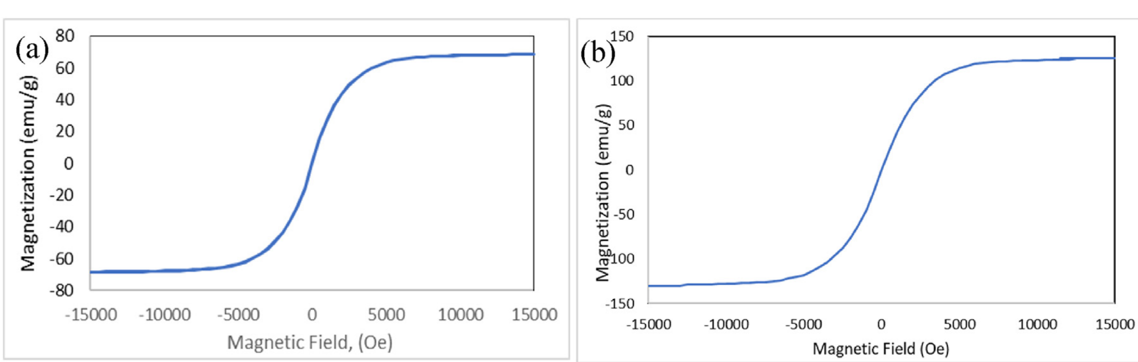

Fig. 2 – Magnetic hysteresis loop of the particles in this study which (a) $CoFe_2O_4$ recorded the value of magnetization at 68.8emu/g and (b) magnetization of CIP at 130 emu/g.

Table 2 – Magnetic properties of the magnetic particles.

Samples	Magnetization (emu/g)	Coercivity, (Oe)	Retentivity (emu/g)
CIP	130	24.03	3.96
$CoFe_2O_4$	68.6	16.397	0.45

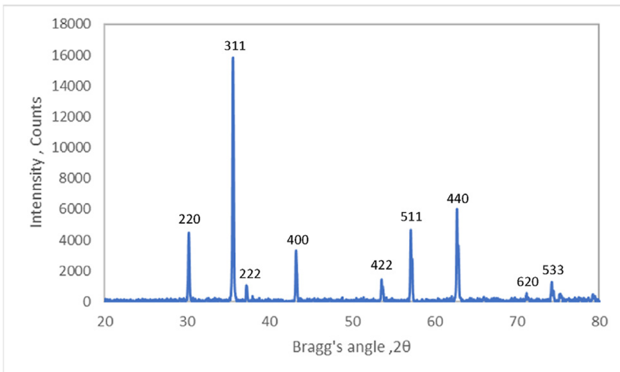

Fig. 3 – X-ray diffraction of $CoFe_2O_4$ from 20° to 80° of Bragg's angle.

high applied shear strain. The LVE region can be determined from the graph, where about 10% from the plateau of the constant G′ value was the limit of the material, which possessed a viscoelastic behaviour. The effect of applied strains on the storage modulus, G' and loss modulus, G″ of the MRG with the incorporation of the $CoFe_2O_4$ as an additive across different concentrations are depicted in Fig. 3, particularly at the off- and on-state conditions. The modulus obtained emphasizes the stored energy and is dissipated among the molecular structure of the samples as shearing acts upon it.

Based on Fig. 4 (a) and (b), both findings noted that the viscoelastic behavior of the MRG samples was dependent on the strain amplitudes, magnetic flux density, and concentrations of the $CoFe_2O_4$ particles which were added into the MRG, particularly at a constant frequency of 1 Hz. It showed that at the off-state condition (Fig. 3 (a)), the MRG with $CoFe_2O_4$ particles, regardless of any concentration, exhibited longer portions of LVE regions, designated as LVE_2in comparison to the MRG without $CoFe_2O_4$ particles, which was denoted as LVE_1. The LVE region of the MRG samples was improved from a strain limit of 0.07% (LVE_1) to 0.09% (LVE_2), by adding $CoFe_2O_4$ particles, which significantly improved the elasticity and ability of the material to attain a higher applied stress [17,26].

It can be observed in Fig. 4 (a) as well that the storage modulus for all MRG samples demonstrated an increasing trend as the concentration of the $CoFe_2O_4$ particles increased. The figure depicted that when the shear strain increased up to 10%, the storage modulus, G′ of MRG-5 attained the highest value compared to other MRG samples. This was increased about three times as compared to the MRG. This was due to the stiffer MRG-5, which had more suspended particles in the grease, for both the 50 wt.% CIP and 5 wt.% $CoFe_2O_4$ additives. In fact, the value of the storage modulus of the MRGs were 0.152, 0.279, 0.374, 0.395 and 0.459 MPa for the MRG-1, MRG-2, MRG-3, MRG-4, and MRG-5, respectively. This indicated that the MRG samples were stiffer with more $CoFe_2O_4$. On the other hand, the increasing applied shear strains from low (0.001%) to a higher value (10%) led to the deterioration of the entanglement of the grease, which caused the drop of the G′ of the MRG, particularly at more than 0.1% of the applied strain. The internal viscous friction between the collision of the CIP converted the elastic energy of the material into frictional heat, forming a lower viscous grease with the increased shear strains. Therefore, the capability of the MRG to store more energy dropped further [27].

On the other hand, the loss modulus, G″ for the MRG samples slightly increased with the increment of the applied strains until it reached a threshold point, at about 1% strain, and then significantly decreased. Increment of G″ for all the samples can be clarified in terms of energy dissipation (as

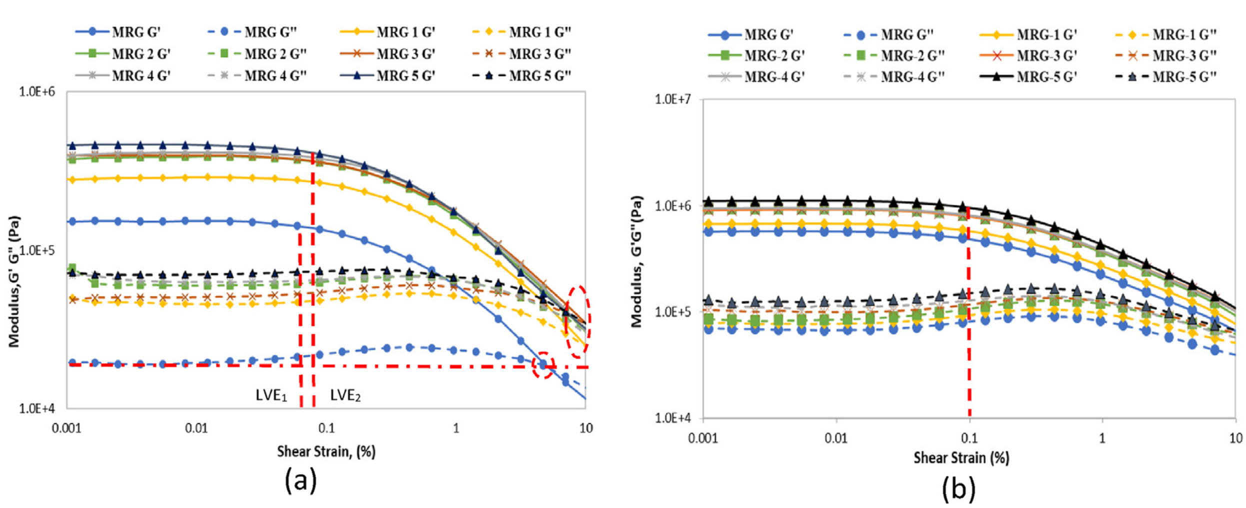

Fig. 4 – torage and loss modulus of MRG under different values of strain for all samples at (a) off-state condition (b) on-state conditions where the dotted line represents the LVE region and the round-dashed line represents the crossover between the storage and loss modulus of the MRG samples.

heat) with increasing shear strains. The phenomenon caused a rupture of the MRG's structure, and slightly less energy could be stored. This indicated that the storage modulus of the MRGs dropped. Lower energy could be stored by the MRG, which simultaneously decreased in the loss modulus due to the movement of the CIP with increased shear strains. As can be seen from Fig. 4 (a) as well, the cross-over point, $G' = G''$ (equilibrium modulus) for the MRG sample was at 7% strain, which was marked as circular dotted lines. Meanwhile, the equilibrium modulus obtained for the MRG with the $CoFe_2O_4$ was at 10% of the strain, which was marked by red oval dotted lines. This phenomenon indicated that the MRG samples entered a viscous flow region, and developed a transition behavior from a viscoelastic solid to a viscoelastic liquid [26]. At this intersection point, the weak bond of the grease started to break, which resulted in the separation of the base oil in the grease medium, particularly at higher applied strains. Nevertheless, the presence of $CoFe_2O_4$ in the MRG for 1 to 5 wt.% contributed to the delay of the transition regions with respect to the enlargement of the LVE region, where more strains could be endured by the samples before degradation of the MRG's structure took place. Somehow, at the off-state condition, the behaviors of the MRG samples mainly depended on the natural structure of the MRG itself, and not the induced magnetic fields.

Based on Fig. 4 (b), it is shown that when 0.6 T was applied to the samples, the behavior of the MRG samples exhibited a similar off-state condition, as shown in Fig. 4 (a). The critical strain of all the samples was discovered at 0.1% strain, which with respect to the LVE region of all the samples, this LVE value was similar to the dynamic properties of the MR gels [26,28,29]. This region manifests with longer elastic deformation of all the samples under the applied shear strains and magnetic fields, at which the internal structure of the materials is then returned to its original condition upon removal of both the applied stresses. Furthermore, the increment of G' for all MRG samples when the external magnetic field was applied were doubled in comparison to the samples with no magnetic fields (off-state condition). It is noted that the induced magnetic field caused stiffer MRGs, which was attributed to the attraction between the magnetic particles, which became stronger via magnetic forces and was able to obstruct the hydrodynamic forces. Thus, this resulted in the formation of chain-like structures of the particles in the grease medium [30]. Indeed, with the presence of $CoFe_2O_4$ in the samples for both the magnetic and polygonal shape of the $CoFe_2O_4$ is believed to assist and promote its formation and the strong chain-like structures for both particles, i.e., the CIP and $CoFe_2O_4$. The shape of the $CoFe_2O_4$ is expected to promote the dipolar–dipolar interactions of the particles via the shape's axes during the alignment of the particles when stimulated with the magnetic field. The magnetic anisotropy of the $CoFe_2O_4$ can form one or more axes depending on the ease and favorability of the spontaneous direction of the magnetization, and thus would lead to a larger contact surface with the spherical shape of the CIP.

The polygonal shape of the $CoFe_2O_4$ promoted good wettability, which resulted in the optimal dispersion of the particles in the grease medium, and reduced the grease resistance as soon as strains and magnetic fields were applied to the MRG. As a result, the movement of the magnetic particles in the grease medium became easier, and formed a stronger particle chain alignment when external magnetic forces were applied. This subsequently improved the LVE region of the MRG samples. However, the strong interaction between the particles was seen to break when the strain amplitudes were further increased. This gradually led to the MRG samples being able to flow much better, especially beyond the LVE region, due to the Payne effect, which was in line with the silica and Gr [31,32]. In addition, there was no cross-over point observed between the G' and G". The $G'>G''$ for all the MRG samples, particularly at 10% strains, denoted that all the samples were attained as viscoelastic materials even at higher amplitudes strain values. In fact, the addition of the magnetic additive $CoFe_2O_4$ in the MRG samples contributed to the enhancement in the modulus of the materials.

3.3. *Effect of sweep magnetic field on the rheological properties of MRG*

Fig. 5 depicts the storage modulus versus magnetic flux density for samples with different compositions of $CoFe_2O_4$, at a shear strain of 0.001%. The absolute MR effect can be calculated by using the following equation:

Absolute MR effect, ΔG: G'_{max}-$G'_{initial}$

Where G'_{max} is the maximum value of storage modulus and $G'_{initial}$ is the initial value of the storage modulus of the MRG. Based on Fig. 5, as expected, the storage modulus of the samples increased along with the applied magnetic flux density, as the induced magnetic fields caused a stronger interaction between the magnetic particles in the grease. From the observation, the MRG sample showed the lowest initial value of the G' as compared to samples with $CoFe_2O_4$ particles. This indicated that the lower stiffness of the of MRG attributed to a lower amount of particles in the grease medium. The dispersed particles in the MRG were referring to the CIP only.

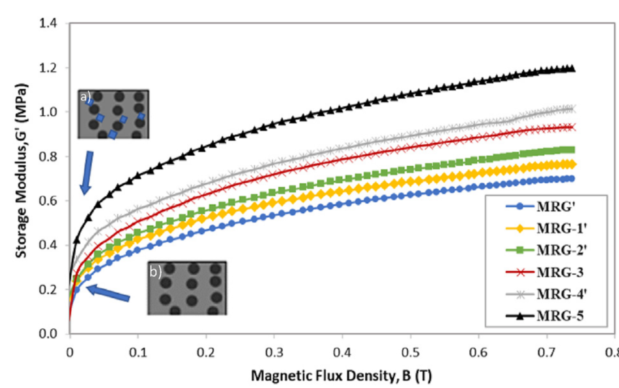

Fig. 5 – Storage modulus of MRG samples as a function of magnetic flux density from 0 T to 0.75 T. All the MRG samples demonstrated that the storage modulus directly proportional to the magnetic flux density. The schematic illustrations represented the initial structuration of the particles in grease medium which a) with $CoFe_2O_4$ and b) without $CoFe_2O_4$.

Table 3 – Summary of storage modulus of MRG samples under magnetic field sweep.

Samples	G'_o, (MPa)	G'_{max}, (MPa)	Absolute MR effect, ΔG
MRG	0.10678	0.69772	0.59094
MRG -1	0.16075	0.76607	0.60532
MRG -2	0.16986	0.82889	0.65903
MRG -3	0.17655	0.93053	0.75398
MRG -4	0.23318	1.0149	0.78172
MRG -5	0.25445	1.1969	0.94245

However, the MRG-5 resulted in the highest initial value of the G′ as compared to other MRGs with $CoFe_2O_4$, by almost 77% in terms of increment in the MRG without the $CoFe_2O_4$. This was due to the higher concentration of particles in the grease medium. This resulted in a stiffer MRG sample, as illustrated by the schematic diagram in Fig. 5. In addition, at the highest magnetic flux density, the G'_{max} values for all the samples increased as the composition of the $CoFe_2O_4$ particles increased, particularly for the samples with 0.6977, 0.7661, 0.8289, 0.9305,1.0149 and 1.1969 MPa, for 1 to 5 wt.% respectively. These values indicated that the presence of magnetic

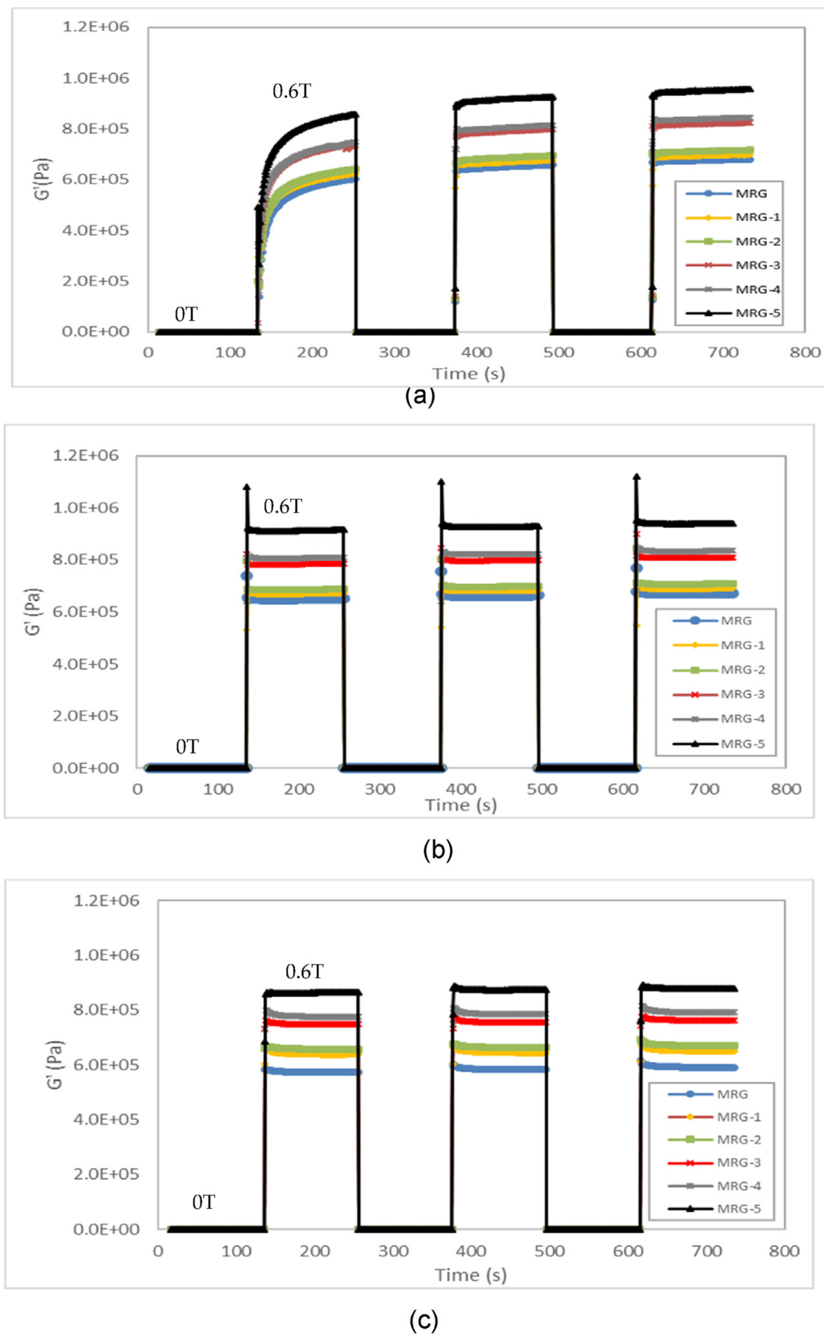

Fig. 6 – Transient behaviour of MRG samples under different value of applied strains (a) 0.01%, (b) 0.05% and (c) 0.1% when the magnetic field was removed and applied.

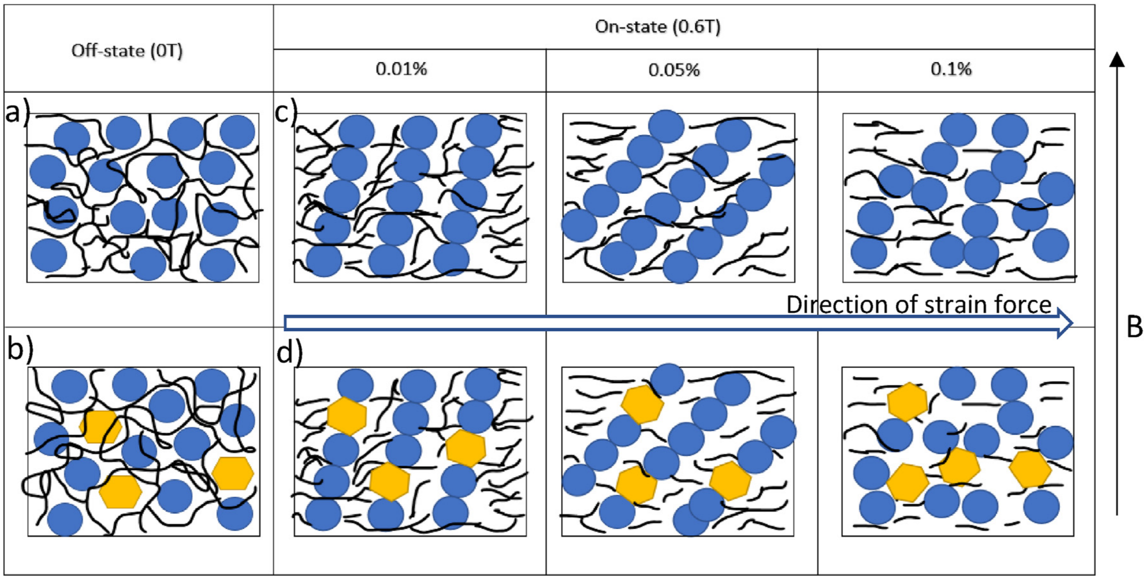

Fig. 7 – Illustrations of the pure MRG and MRG with $CoFe_2O_4$ during off state and on state conditions. In (a) and (b), it illustrated the conditions of MRG and MRG with $CoFe_2O_4$ when there is no magnetic field applied on it. (c) and (d) showed that when magnetic field applied on MRG samples, the particles in grease medium which consist of fibrous structure were aligned. As the shear strain increase, the fibrous structure of the grease starts to loosen and breakage of weak molecular bond of the grease. (d) This demonstrated that with the presence of $CoFe_2O_4$, the fibrous structure of the grease more straightened and shortened compared with (c).

additives in the MRG samples considerably improved the storage modulus, G' of the MRGs by strengthening the formation of the particle's chain alignments with increased magnetic field intensities [33,34]. In fact, the magneto induced, or commonly known as absolute MR effect (or ΔG), showed an increment with the increasing amount of $CoFe_2O_4$ in the MRG samples. This was also related to the enhancement of the G' in the MRG samples with $CoFe_2O_4$ which resulted in the greater responsiveness of the samples, which was attributed to the enhancement of the ordered degree of the particle's alignment towards the manipulation of the applied magnetic fields. This complied with the previous studies done by Li et al. [35]. In brief, the rheological properties of the MRG in terms of the storage modulus and MR effects were proportional to the concentration of the additives and the magnetic field strength applied to the samples. The results of the storage modulus, G' for G'_o (0 T) and G'_{max} at 0.75 T are tabulated in Table 3.

3.4. *Transient response of MRG samples*

The transient response test was carried out to investigate the responsiveness of the MRG samples towards the tunability of the shear strains at applied magnetic fields from 0 to 0.6 T. The test was applied at a frequency value of 1 Hz, and the strains were set at 0.01%, 0.05% and 0.1%, respectively. The storage modulus was obtained as the output of the set parameters. The transient response of the MRG samples was then calculated at about 63% of the storage modulus from the off-to on-state conditions [17,24].

Fig. 6 shows the transient response of the MRG samples in terms of storage modulus under the periodical stepwise magnetic fields at different applied shear strains. As can be seen from the depicted figures, the value G′ rapidly increased when the condition changed from the off-state to the on-state condition (magnetized), and became totally de-magnetized when the magnetic field was removed. This phenomenon

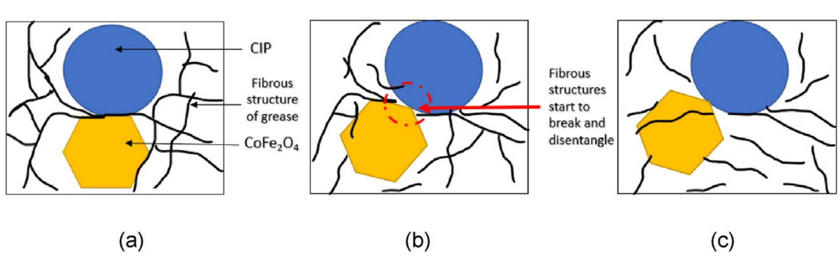

Fig. 8 – Schematic illustrations on what happened to the fibrous structure of grease during the shearing process of MRG with the presence of $CoFe_2O_4$. (a) When the magnetic applied, the particles tend to align within the fibrous structure of the grease. (b) At low shear strain, the magnetic particles move along with the direction of the forces simultaneously the edge of $CoFe_2O_4$ 'cut' and disentangle the grease's fibrous structure. (c) At high shear strain, the fibrous structures of grease are shortened and disentangle while the $CoFe_2O_4$ are still intact with CIP.

Table 4 – Transient response (s) of the MRG samples with variations of shear strains (%).

Samples	Time taken (s) to respond from off-state (0 T) to on-state (0.6 T) condition		
	0.01%	0.05%	0.1%
MRG	8.15	1.0	0.58
MRG-1	6.53	0.74	0.54
MRG-2	6.35	0.62	0.60
MRG-3	6.20	0.56	0.60
MRG-4	5.85	0.52	0.56
MRG-5	5.65	0.47	0.54

can be seen via the sharp vertical line of the off-state (0 T) as it transitioned into the on-state (0.6 T) condition, or vice-versa. This indicated that the MRGs were able to store the magnetic energy, and simultaneously removed the energy shortly after due to the low remanence properties depicted by the materials. The value of the storage modulus, G' for the MRG exhibited the lowest value as compared to the MRG-1, MRG-2, MRG-3, MRG-4, and MRG-5 samples, which showed a stiffer MRG sample with the applied magnetic field. The interaction of the particles in the grease medium improved in the presence of $CoFe_2O_4$, which resulted in a stronger particle alignment, as discussed in the previous section.

It was observed in Fig. 6 (a) that the G′ values for all the MRG samples showed an increment over several periodical stepwise magnetic fields, particularly at a 0.01% applied strain, which indicated that the strength of the particle's chain structures was increasing. In fact, at lower applied shear forces, this might be insufficient to support the mobility of the magnetic particles to form the particle's chain alignment in the grease medium due to the restrictions from the fibrous structure of the grease, as illustrated in Fig. 7. When a 0.05% strain was applied to the MRGs, as in Fig. 6 (b), the obtained G′ showed a consistent value under the periodical stepwise magnetic fields whereas the overshoot peak indicated that the particle's chain of the CIPs in the MRG experienced higher forces, which were from the strain and magnetic fields applied, and became much more stabilized after a few seconds. A similar finding was observed for the higher applied strain of 0.1% (Fig. 6 (c)), as a much more stable particle chain alignment was achieved. In terms of transient responses, the MRG, which was incorporated with the $CoFe_2O_4$ performed at a rapid response upon the application of magnetic fields, compared to the MRG without $CoFe_2O_4$, which was true for all the applied strains. As calculated from the obtained graphs, the MRG-5 with the highest load of $CoFe_2O_4$ showed much faster responses, by 5.65s at 0.01% and 0.47s at 0.05% of strains, as compared to the MRG, which responded at 8.15s for the 0.01% strain and 1.0s at the 0.05% strain. These results denoted that the presence of $CoFe_2O_4$ enhanced the response time of the MRG samples by improving the particle's mobility in the grease medium, and hindered the hydrodynamic forces during shearing. The achievement was also attributed to the decrease in the MRG's viscosity with such an addition, which disrupted the fibrous structure of the grease. This made it easier for the particles to generate the alignment as shown in Fig. 8 [17]. Beyond the LVE limit of the MRG samples, particularly for the 0.1% applied strain sample, the MRG samples performed at the fastest responsiveness, as calculated and presented in Table 3. This finding is due to the disintegration of the particle's chain structure which broke the weak molecular bonding between the grease at higher applied shear strains [36] Although the transient response of the MRG was quicker with higher applied forces, the behavior of the MRG exceeded the LVE limit, at which the MRG was no longer seen to be in the viscoelastic behavior.

The schematic diagram on the dispersion of magnetic particles in the grease medium based on the current finding is shown in Fig. 7(a–d). In the absence of magnetic field, the magnetic particles are dispersed within the fibrous structure of the grease thickener. As the magnetic field was induced to the MRG sample, the magnetic particles in the medium would be aligned following the direction of magnetic field, with the $CoFe_2O_4$ particles would tend to align based on their ease axis of magnetization among the CIPs. As the shear strain was applied simultaneously and increased regularly, the fibrous structure of the grease started to loosen and breakage of the weak molecular bond of the grease occurred due to the collision that generated by movement of the particles, as illustrated in Fig. 7 (c) and (d). Besides, as shown in the magnified schematic diagram of Fig. 8 which respective to the presence of $CoFe_2O_4$ particles in the MRG, the polygonal shape particularly the edge of the particles would aid to cut and disentangle the fibrous structure of the grease which release the base oil within the grease thickener. Subsequently, the responsiveness

Table 5 – Comparison of transient response between previous and current study.

Based matrix	Magnetic Filler	Additives	Conditions		Transient Response	Research
Grease	Plate-like CIP	–	Strain	0.01%	6s	[27]
				0.05%	2s	
	Bi-dispersed CIP	–		0.01%	5s	
				0.05%	2s	
Elastomer	CIP	Fe–Ni coated	Current	1 A–5A	0.34s–1.27s	[37]
Gel	CIP	1 wt.% of laponite	Magnetic field	0.18 T	~12s	[38]
				0.35 T	~10s	
Fluid	CIP	1 wt.% of iron powder coated with hydrophilic carbon shell	Magnetic field	38 kA/m	NA	[39,40]
				69 kA/m	NA	
Grease	CIP	5 wt.% of $CoFe_2O_4$	Strain	0.01%	5.65s	Current study
				0.05%	0.47s	

of the magnetic particles towards the applied magnetic fields could be improved and the transient response of the MRG has been enhanced. The summary of time response of MRG has been tabulated in Table 4. Meanwhile, brief comparison between the current and previous studies related to transient response of MR materials has been shown in Table 5 which indicates the rising focus and importance of the study recently. It is noted that, the presence of additives has improved the transient response of MR materials respectively, which is in-line with the finding of this present study.

4. Conclusion

To sum up the findings, the utilization of $CoFe_2O_4$ in the MRG was shown to be able to successfully enhance the properties and behavior of the MRG, at both the off-state and on-state conditions. With the incorporation of $CoFe_2O_4$ particles, the LVE region of the MRG was enlarged to 0.09% from 0.07%, as compared to the MRG without $CoFe_2O_4$, particularly at the off-state condition. It was shown that the additive improved the elasticity and delayed the deformation of the MRG when a shear strain was applied. At the on-state condition, the presence of $CoFe_2O_4$ assisted and promoted a stronger particle chain alignment among the CIPs, which resulted in a stiffer MRG. This simultaneously enhanced the storage modulus and the absolute MR effect. In addition, along with the increment of the $CoFe_2O_4$ content in the MRG, the transient response of the material when subjected to the 0.6 T of applied magnetic field performed faster, particularly within the LVE. This was improved by 56%, as compared to the MRG without the $CoFe_2O_4$. The enhancement of the rheological performance of the MRG with a higher concentration of $CoFe_2O_4$, as well as the transient response towards the manipulation of the magnetic fields, achieved the intended target, at which a wider potential for the related MR devices was seen. All the results from this study suggest that the rheological properties and transient responses of the MRG depend on the concentrations of the $CoFe_2O_4$ as an additive. In future works, it is recommended to concentrate on the delay time of the materials and their responses towards the stimulus. It is suggested to investigate the performance of the MRG with the different types of additives.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Nur Azmah binti Nordin reports financial support was provided by Universiti Teknologi Malaysia Malaysia-Japan International Institute of Technology. Ubaidillah reports financial support was provided by Sebelas Maret University.

Acknowledgment

This study was funded by through Universiti Teknologi Malaysia (UTM) Zamalah (Vot No. 0060 N) and Hibah non APBN 2023 by Universitas Sebelas Maret.

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