

# Field-Dependent Rheological Properties of Graphite Based Magnetorheological Grease

**Nur Alyaa Mohd Nasir**

University of Technology Malaysia

**Nurhazimah Nazmi** (✉ [nurhazimah@utm.my](mailto:nurhazimah@utm.my))

University of Technology Malaysia

**Norzilawati Mohamad**

Universiti Malaysia Sabah

**Ubaid Ubaidillah**

Universitas Sebelas Maret

**Nur Azmah Nordin**

University of Technology Malaysia

**Saiful Amri Mazlan**

University of Technology Malaysia

**Siti Aishah Abdul Aziz**

University of Technology Malaysia

**Muhammad Kashfi Shabdin**

Universiti Putra Malaysia

---

## Research Article

**Keywords:** Magnetorheological grease, graphite, non-magnetic particles, rheological properties

**Posted Date:** June 17th, 2021

**DOI:** <https://doi.org/10.21203/rs.3.rs-620537/v1>

**License:**  This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

---

# Abstract

The utilization of high viscous grease as a medium in Magnetorheological grease (MRG) has benefits in avoiding the sedimentation from occurred, however, it limits the expansion of yield stress in on-state condition thus reduced the application performance upon operation. Therefore, in present study, improvement of the rheological properties of MRG has been investigated by introduction of graphite as an additive. MRG with 10 wt% of graphite (GMRG) was fabricated and the analysis of the properties was compared to the reference sample, MRG. The microstructure of GMRG was characterized through Environmental Scanning Electron Microscope, (ESEM). While, the rheological properties including apparent viscosity, yield stress and viscoelastic properties for both samples were examined using shear rheometer under rotational and oscillatory mode. The results demonstrated slightly increment of apparent viscosity in GMRG and shows a significant improvement in term of yield stress by 38.8% at 3A with growth about 32.7% higher compared to MRG from 0A to 3A. Also, an expansion of linear viscoelastic region (LVE) from 0.01 to 0.1% was observed in the GMRG credited to the domination of elastic properties on the sample. These results obtained was confirmed based on ESEM that displayed the contribution of graphite towards constructing more stable chain structure in GMRG. In conclusion, this study highlights the influence of graphite towards improving the rheological properties of MRG. Hence, addition of graphite in MRG is a great potential to be applied in many applications in near future.

## 1. Introduction

Magnetorheological grease (MRG) is categorized as a smart material as its rheological properties can be manipulated with an application of magnetic field. MRG was invented by Rankin et al.<sup>1</sup>, capable to solve the sedimentation problem arises from magnetorheological fluid (MRF) and low magnetorheological (MR) effect in magnetorheological elastomer (MRE). The usage of grease, which is non-Newtonian fluid as a medium in MRG has enabled the magnetic particles to suspend against the gravitational force thus, sedimentation of particles can be eliminated<sup>2,3</sup>. Moreover, the grease is classified as an intermediate state between fluid and solid, provides certain freedom of movement to the magnetic particles to form columnar chains under the influence of magnetic field<sup>4</sup>. As consequences, a higher MR effect of 952.38% was recorded on MRG compared to 71.7% of solid-like state MRE<sup>5,6</sup>. Apart from that, by comparing with MRF, the MRG does not require additional sealing to prevent leakage of the devices as MRG own self-sealing property due to their thick viscosity. This property can maintain the stability of the equipment for long-term usage, thus reduce the manufacturing cost<sup>7</sup>. The merits mentioned above have directed MRG to become a potential candidate to be applied in engineering applications such as in seismic dampers, brakes and clutches<sup>8</sup>. However, the utilization of grease as a medium for MR material has made the MRG to experience high off-state viscosity and apparently, limits the expansion of yield stress in the on-state condition. This phenomenon in return has led the MRG to exhibit poor performance such as torque output<sup>9</sup>.

The improvement of rheological properties of the MRG can be undertaken by addition of additives. Kim et al.<sup>10</sup> investigated the influence of kerosene oil as an additive on the rheological properties of MRG. They discovered that the apparent viscosity of MRG was reduced by the addition of 5 wt% of kerosene, which indicated a better dispersion of CIPs in the grease medium. However, at the same time, the dynamic yield stress and viscoelastic properties of MRG were also decreased. Their findings were consistent with the study conducted by Mohamad et al.<sup>11</sup> that utilized and compared three different types of dilution oils namely kerosene oil, castor oil and hydraulic oil in MRG. Even though, they discovered the usage of these type of dilution oils was able to reduce the off-state viscosity of the MRG, however, the dynamic yield stress of the MRG were also reduced. On other words, the addition of dilution oils in MRG could lower their apparent viscosity, however, it would make the carbonyl iron particles (CIPs) less attached to the grease medium and expected to experience slipping effect under the influence of shear stress that caused drop on the resultant yield stress. Consequently, it would reduce its performance especially under the influence of low magnetic field strengths. Recently, Wang et al.<sup>12</sup>, has optimized the method to fabricate MRG through an ANOVA analysis by many parameters such as CIPs fraction and size, and silicone oil viscosity. They found that the optimum yield stress could be obtained by manipulating the CIPs fraction and silicone oil viscosity, but the influence of CIPs size was negligible. However, it was noted that as the utilized silicone oil viscosity was higher, the yield stress of MRG were seen to drop at a higher magnetic field strength. The reasons were possibly due to the utilization of high viscosity of silicone oil, up to  $1000 \text{ m}^2\text{s}^{-1}$  in MRG has contributed to the rise of MRG's apparent viscosity, which finally restricted the alignment of CIPs in the medium under influence of magnetic fields.

Apart from utilizing different types of dilution oil as an additive in MRG, there are several studies that incorporated solid type additives in order to enhance the rheological properties of MRG. For an example, Park et al.<sup>2</sup> revealed that by adding nanoparticle's additive,  $\text{CrO}_2$  in MRG has helped to improve the stability of MRG that resulted from the steric repulsion effect between CIPs and  $\text{CrO}_2$ . Though, the dynamic yield stress of MRG showed insignificant improvement. Nevertheless, Mohamad et al.<sup>13</sup> introduced another type of nanoparticle's additive in MRG namely, super-paramagnetic,  $\gamma\text{-Fe}_2\text{O}_3$ . The addition of 1 wt. % of additive capable to lower the off-state viscosity and at the same time, increased the viscosity at on-state viscosity of MRG. The result reflected the effect of nano-sized particles that filled in the voids between the CIPs under the influence of magnetic fields, thus contributed towards the formation of stronger chain-like structures inside the medium. Later, Tarmizi et al.<sup>14</sup> utilized micron-sized additive of cobalt ferrite,  $\text{CoFe}_2\text{O}_4$  that has further lowered the off-state viscosity of the MRG by up to 86% with 1 wt%. Their result reported the highest yield stress obtained, about 12 kPa with the incorporation of 5 wt.%  $\text{CoFe}_2\text{O}_4$  at 0.64 T of applied magnetic field. However, it was noted that the range of expansion of yield stress in MRG with incorporation of 5 wt%  $\text{CoFe}_2\text{O}_4$  was considered low, which was from 0.8 to 12 kPa by increasing magnetic field from 0 to 0.64 T. Apparently, this yield stress range limit the material to be applied in wide range of applications.

Aside from adding solid magnetic additives to improve the rheological properties of MR material, alternatively, an incorporation of non-magnetic, carbon-based additives such as graphite could also enhance the rheological performance of MR materials<sup>15-18</sup>. Graphite possesses excellent properties such as good thermal and electrical conductivities, mechanical properties, chemically inert and low density, which capable to maintain the existing mechanical properties of the material. In fact, incorporation of graphite into MRG is susceptible to induce electrical properties that make MRG to have dual-behaviour MR material. Moreover, graphite can be classified as an economical additive attributable to the high availability and low-cost production<sup>19</sup>. An experimental study conducted by Tian et al.<sup>15</sup> presented that the initial mechanical properties of MRE has been improved by addition of 20 wt% graphite. Then, a noticeable improvement of MR effect up to 60% towards field-dependent modulus of MRE has been confirmed by Shabdin et al.<sup>16</sup> with corresponding to the 33 wt% graphite. In their study, the MR effect was improved by 176% as compared to the previous study in<sup>15</sup>.

Other MR materials such as MR plastomer (MRP) and MR fluid (MRF) have also benefited from the utilization of graphite as an additive. With addition of 15 wt% graphite in MRP has increased the saturated storage modulus by 0.8 MPa compared to pure MRP, and the viscosity was remarkably improved due to strengthen effect exhibited by graphite<sup>17</sup>. In another study performed on MRF by Thakur<sup>18</sup>, a high on-state viscosity and shear stress values could be obtained with increasing the weight percentage of graphite flakes by up to 3%. The authors stated that this was caused by the contribution of graphite that involved in improving the formation of columnar chain structure by filled the empty gap between CIPs to form more strong structures, and as a result, the yield stress was elevated.

Therefore, it can be concluded that the incorporation of graphite is proven to improve the rheological properties of MR materials, and it is expected that rheological properties of MRG will be improved too. Although the off-state viscosity of MRG is presumed to slightly increases with the addition of graphite powder, the field-dependent yield stress of MRG can be enhanced due to the strengthen effect exhibited by the graphite. In this study, carbon-based additive, graphite is utilized in the fabrication of a new MRG namely GMRG. The samples are tested on the rotational and oscillatory rheological behaviour by using commercial rheometer with parallel plate measuring cell. The performances of GMRG including yield stress and viscoelastic properties are compared and discussed with reference sample, MRG without graphite.

## 2. Methodology

### 2.1 Sample Preparation

The samples consisted of soft magnetic, CIPs (OM grade, BASF, Germany) with average size and density of 5  $\mu\text{m}$  and 7.874  $\text{g}/\text{cm}^3$ , respectively, dispersed in commercial lithium grease (NPC- Highrex HD-3 Grease, Nippon Kyu Ltd, Japan). The density and viscosity of utilized grease were 0.92  $\text{g}/\text{cm}^3$  and 0.207, Pa.s respectively. The additive used in this study was irregular shape of graphite (R&M Chemicals,

EverGreen Engineering and Resources Co., Malaysia) with average size of 16  $\mu\text{m}$  and density of 1.8  $\text{g}/\text{cm}^3$ . In general, MRG was fabricated by direct mixing of CIPs and grease, while for GMRG, with the addition of graphite. Specific composition for both MRG and GMRG samples are shown in Table 1. Initially, the samples were prepared by stirring the grease for 5 minutes to open the fibrous structures. Subsequently, either CIPs and graphite (GMRG) or CIPs only (MRG) were added into the grease and stirred with speed of 300 rpm continuously for 2 hours using the mechanical stirrer until homogenized mixture was attained. All procedures were conducted at room temperature condition.

Table 1  
The composition of MRG and GMRG samples.

Samples	Percent by weight (wt%)		
	Grease	CIPs	Graphite
MRG	30	70	0
GMRG	20	70	10

## 2.2 Sample Characterization

The microstructure of GMRG were examined via environmental scanning electron microscope (ESEM; Quanta 450 FEG, FEI) to observe the distribution of the CIPs and graphite in grease medium. Thin layer of platinum was used to coat the sample prior the analysis in order to avoid the electron charging of the samples. Furthermore, the rheological properties of MRG were characterized using parallel-plate rheometer (Anton Paar, Physica, MCR 302) under rotational and oscillatory mode. The shear rheometer was equipped with MR device (MRD70/1T) to generate magnetic fields in the rheometer. For each measurement, 1 mL of sample was filled on the parallel-plate with diameter of 20 mm at a constant gap of 1 mm. The variation magnetic field strengths from 0 to 0.603 T, which corresponded to 0 to 3A of applied current were performed on all samples. The apparent viscosity and shear stress of MRG and GMRG were determined by manipulating the shear rate ranging from 0.01 to 100  $\text{s}^{-1}$  under continuous mode. In the meantime, the elasticity and energy dissipation of MRG and GMRG were evaluated through sweep strain tests. The strain was varied from 0.001 to 10% at a constant frequency of 1 Hz. All experiment were conducted at room temperature, 25°C.

## 3. Results And Discussion

### 3.1 ESEM Characterization

ESEM images of GMRG sample are illustrated in Fig. 1. Figure 1a displays an image of the sample without influence of magnetic field, Fig. 1b for sample under pre-treated with magnetic field of 0.1T, and Fig. 1c to show the enlarged views of microstructure CIPs and graphite in GMRG sample.

Generally, the CIPs and graphite were randomly distributed in grease medium in the absence of magnetic field (Fig. 1a). Then, with influence of magnetic field, the CIPs were attracted to each other through dipole-dipole force and start to align to the direction of applied magnetic field. Simultaneously, graphite particles vibrate together with magnetically influence CIPs thus involves during aligning process to form more stronger columnar chain structure (Fig. 1b). Moreover, by referring to Fig. 1c, it is confirmed that the utilized CIPs and graphite are in spherical and irregular shape, respectively. However, few graphite was captured due to the small amount of weight percent of graphite compared to CIPs. It was also observed that graphite showed a good dispersion in the grease matrix without existence of any agglomeration.

In terms of ESEM results as illustrated in Fig. 2b, an EDX analysis was performed at selected area, spectrum 16 (Fig. 2a) to confirm the elemental composition in GMRG sample.

Three types of chemical element namely, carbon, oxygen and iron were obtained as listed in Table 2 that displays weight (%) and atomic (%) of each element composition in the sample.

Table 2  
GMRG elemental compositions

Element	Weight (%)	Atomic (%)
C	75.34	89.26
O	7.02	6.24
Fe	17.64	4.49

Carbon exhibited the largest proportion, which was around 75.34 wt.%, and followed by iron and oxygen with the proportion of 17.64 and 7.02 wt.%, respectively. As elemental carbon in 2-dimensional structure of graphite have the lowest energy state, it was the most stable form of carbon in standard condition (ambient temperature and pressure), therefore it reflected to the highest constituent in the result<sup>20</sup>. Besides, large proportion of carbon was due to the hydrocarbon chain from grease matrix, where, in addition, contributed to the proportion of oxygen as well. This was due to the fact that the type of grease being utilized in this study was based on Lithium-12-hydrostearate thickener that consisted of hydroxyl (-OH) and carboxylate (-COO<sup>-</sup>) functional groups<sup>21,22</sup>. Furthermore, the second highest constituent of Fe was contributed from the CIPs that normally made up of pure Iron.

### 3.1 Effect of Graphite on MRG under rotational mode.

Figure 3 represents the apparent viscosity of MRG and GMRG as function of shear rate at various magnetic field strengths.

It can be observed that both samples, MRG and GMRG experienced shear-thinning phenomenon under the absence and presence of magnetic fields<sup>23,24</sup>. The apparent viscosity of both samples declined along with increasing shear rate. This phenomenon related to the destruction of CIPs alignments as well as alteration of grease medium under the influence of high shear force<sup>25</sup>. Furthermore, it can be seen, that

the apparent viscosity of both samples increased as the applied current increased from 0 to 3A, corresponding to the formation of strong columnar chain between CIPs along with the direction of applied magnetic fields<sup>5</sup>.

Apart from that, it can be realized GMRG exhibited a higher value of apparent viscosity at each magnetic field strength as compared with MRG by increasing shear rate. For an example, at off-state condition, the value of initial apparent viscosity of GMRG showed an increment about 0.049 MPa.s as compared to the MRG sample. In absence of magnetic field, the apparent viscosity of MRG was merely depended on the fibrous structure of grease matrix<sup>8,23</sup>. However, with addition of graphite in GMRG has brought into thickening effect in the grease matrix<sup>26</sup>, as employment of graphite has increased the number of solid content in sample GMRG that also contained CIPs. Consequently, a greater number of particles to polymer interaction occurred and as a results, the flow resistance of the medium increased<sup>27</sup>. Even so, the increment of the apparent viscosity in GMRG was still considered low. The reason was because of the low density of utilized graphite as compared to CIPs, which about 4 times differences.

Nevertheless, with the presence of magnetic field, GMRG depicted a slightly increased on apparent viscosity compared to MRG. From the result obtained, it can be proved that although graphite is non-magnetic particles, yet it can involve in the process of alignment with CIPs to form more stronger structures with the presence of magnetic fields. The result is in agreement with Zhang et al.<sup>28</sup>. Furthermore, such finding<sup>5</sup> was consistent with the previous studies that utilized graphite in improving the rheological properties of MR material<sup>17,18</sup>. However, GMRG exhibited unstable apparent viscosity in applied current of 3A due to the formation of thicker structure that attributed from graphite particles along with increasing of magnetic field strength. Thus, the formation might contribute to the slip of parallel plate rheometer at high shear rate<sup>29</sup>.

The shear stress assessment with shear rate range from 0.01 to 100 s<sup>-1</sup> at varied applied magnetic field strength is demonstrated in Fig. 4.

A fluctuating shear stress trend was remarked on GMRG at off-state condition. The increasing of solid content in the GMRG caused extra collisions happened between the particles and the medium under the influence of shear force. Furthermore, larger size and irregular shape owned by graphite has led to the disorder motion in the grease medium. Nonetheless, the shear stress trend of GMRG was more stable upon an application of magnetic field. Besides, at low shear rate, it was observed that both samples showed a linear increment of shear stress. This might be happened due to the CIPs in medium were not stable at low shear rate as the formation of columnar chain structure was initiated but hindered due to shear force. However, the shear stress at a higher shear rates depicted a stable trend owing to the strong dipole-dipole interaction between the CIPs, which perpendicular to the direction of shear flow. It was known that the shear stress of MR suspension manageably improved with the implementation of high shear rate<sup>30</sup>.

Additionally, the experimental results of GMRG demonstrated high value of shear stress compared to MRG in off-state condition. This result corresponded by the additional particles in GMRG that attributed from CIPs and graphite, which participated in the process of shear. The addition of graphite has contributed to increase the number of interactions between the particle-particle and particle-medium, however, resulted in more frictions between the particles and the medium. Subsequently, the interactions have induced the flow resistance of the medium and as a consequence the shear stress of GMRG sample increases<sup>27</sup>. Interestingly, an effect of graphite on shear stress was also obviously seen with presence of magnetic field. It was observed that GMRG exhibited a higher shear stress compared to MRG at all magnetic field strengths. Noted here that, as the applied magnetic field strength increases, a thicker columnar chain structure between CIPs has been formed. Simultaneously, the existing irregular-shaped graphite with high surface contact area has promoted the agglomeration of CIPs around their surface<sup>31</sup>. Subsequently as the density of graphite was much lower than CIPs, graphite was easily being escorted by CIPs in the alignment process. As the outcome, graphite particles were indirectly involved in the formation of columnar chain structure to develop more robust and stable structure, which reflected to the result of increment in the shear stress of GMRG.

From relationship between shear stress and shear rate, yield stress for both samples can be acquired through extrapolation at zero shear rate. Figure 5 shows yield stress of MRG and GMRG as a function of varying applied current from 0 to 3 A.

A linear trend of enhancement in the yield stress was observed at both samples, MRG and GMRG towards increasing magnetic field. The yield stress of MRG has shown an improvement from 1.375 to 36.051 kPa, while 4.0167 to 50.048 kPa for GMRG as increasing the applied current from 0 to 3A. The increment of yield stress was resulted from the formation of stable chain structures within the medium with escalating of magnetic field strength. This owing to the fact that the stiffness of both samples increased along with the magnetic field strength, thus hindered the free movement of CIPs within the medium. From the results, it showed that the growth of yield stress in GMRG was about 32.7% higher as compared to MRG from 0A to 3A. Hence, it proved that graphite could help to expand the range of yield stress of MRG due to their strengthen effect.

Apart from that, as can be seen from Fig. 5, the addition of graphite in GMRG shows an improvement to the yield stress at all applied currents. It was noted that the yield stress of MRG has increased from 36.051 to 50.048 kPa with an increment of 38.8% with 10 wt% of graphite at 3A. Furthermore, the yield stress of GMRG showed a dramatic increment starting from 2 to 3A, which appeared to be a different trend compared to MRG. With further increment of magnetic field strength, the CIPs in the medium that already formed thicker and stronger chain structures, has reached a stable formation of structures. As consequences, a higher force was required to break the new structure, which directly caused sudden rise in the yield stress of GMRG.

## **3.2 Effect of Graphite on MRG under oscillatory mode**

At the beginning of applied strain, the storage modulus of both samples was independent with strain amplitude, this was known as dynamic properties of linear viscoelastic (LVE) material. However, the effect of strain showed a non-linear trend as strain amplitude increases due to change in their dynamic properties at high strain<sup>32,33</sup>. This finding related to the destruction of the microstructure of the samples resulting from a strong distance dependence of dipole-dipole interaction, which known as Payne effect<sup>33,34</sup>. Generally, it is important to identify the LVE region of the material in order to be used in specific applications.

Referring to Fig. 6a, the graph shows the storage modulus,  $G'$  of MRG has increased from 0.77 to 1.74 MPa by increasing the applied current from 0 to 1A. The dramatic increment of storage modulus,  $G'$  caused from strong columnar chain structures that were formed at a stronger magnetic field. Comparing with Fig. 6b, the different gaps of storage modulus,  $G'$  between 0 and 1A for GMRG samples was lower compared to MRG samples. It was acknowledged that by the addition of in GMRG sample has increased the stiffness of the medium due to their high surface contact area that led to a stronger interaction between the grease medium and CIPs. Simultaneously, it would affect the mobility of the CIPs in the medium<sup>16</sup>. Nevertheless, it was observed that the storage modulus,  $G'$  for each sample was slightly increased along with escalated magnetic field strength.

Figure 7 illustrates the comparison between MRG and GMRG samples under shear strain at off- and on-state conditions.

The storage modulus,  $G'$  of GMRG displayed a higher value compared with MRG at all magnetic field strengths. This finding reflected to a strong viscoelastic behaviour exhibited by GMRG samples. Meanwhile, the addition of graphite as in sample GMRG has shorten the range of LVE region in off-state condition. It was because graphite contributed to a stiffer GMRG, thus sensitive to a low strain, which could cause microstructural damage. However, a broaden LVE region ( $< 0.1\%$ ) was observed in the GMRG sample compared to MRG sample ( $< 0.03\%$ ) at on-state condition. This was because the irregular shape of graphite contributed to good wettability between graphite and grease, which led to an excellent dispersion of graphite in grease medium<sup>31</sup>. As a result, graphite was also involved in the alignment process together with CIP, thus provided more stable structures at high magnetic field strength.

Additionally, Fig. 8 presents the loss modulus,  $G''$  of MRG and GMRG at small strain ranging from 0.001 to 10% at different applied currents.

The loss modulus,  $G''$  for each sample showed a lower value at strain  $< 0.1\%$  but increased dramatically at strain  $> 0.1\%$ . Both samples displayed fluctuating loss modulus at high magnetic fields strength in a low applied strain,  $< 0.1\%$ . Moreover, the peak of the graph becoming more obvious with increased of magnetic field strength. This suggested that at high magnetic field strength, more energy dissipated through heat that promoted to more inter-particle interactions between CIPs<sup>35</sup>. On the other hand, the increasing loss modulus at a higher magnetic field might due to the complicated structure attributed from combination of grease's matrix fibre structure with induced-magnetizable chain structure<sup>36</sup>.

On the other hand, the comparison of loss modulus,  $G''$  between both samples in off- and on-state conditions are illustrated in Fig. 9.

It was observed that in absence of magnetic field, the loss modulus of GMRG was higher than MRG, indicated that more heat was dissipated by GMRG. In contrast, with presence of magnetic field, it was noticed that MRG sample displayed a higher and more fluctuated loss modulus compared to GMRG at a very low strain ( $< 0.1\%$ ). The possible reason might relate to the stable structure acquired by GMRG at escalated applied magnetic field strength caused by the existing of graphite particles. However, at strain  $> 0.1\%$ , GMRG showed a much higher value of loss modulus compared to MRG due to strong Payne effect experienced by GMRG.

Based on above results, it can predict that the addition of graphite has a significant effect towards the interaction between CIPs and grease medium. It was assumed that some of the CIPs have attracted to the graphite to establish bonding between them (Fig. 1b). Furthermore, the positive improvement on rheological properties of GMRG was related to the excellent interfacial interaction of graphite with grease medium. Mechanism of the movement of graphite during CIPs alignment under the presence of magnetic field in GMRG is illustrated in Fig. 10.

Without an application of magnetic field, the spherical CIPs and irregular shape graphite were dispersed randomly<sup>16</sup> in grease medium as shown in Fig. 10a. At this stage, the viscosity of the samples was primarily depended on the fibrous structure of the grease matrix. Due to a good adhesion of graphite with grease medium, the apparent viscosity and shear stress of GMRG was supposed to be higher, however, it has a drawback by the low density of graphite. Thus, resulted in a small difference with MRG.

Conversely, with an application of magnetic field, the CIPs were magnetized and started to attract to each other as displayed in Fig. 10b. The movement of CIPs was according to the direction of magnetic field and at the same time, the gap between the particles was reduced. The process of alignment was involved the CIPs, where the graphite was also followed by the movement of CIPs that attached to it. With further increased of magnetic field, the inter-particle interactions were stronger due to the closer gap between the CIPs, resulted in a thick columnar chain structure driven from dipole-dipole force. In the meantime, more CIPs accumulated around the graphite's surface due to rough and high surface area contact of irregular graphite<sup>31</sup>. Consequently, the graphite tended to 'move' together with magnetizable CIPs and thus, involved in the formation of columnar structure within the matrix as shown in Fig. 10c. Apparently, a stronger interaction structure between CIPs, graphite and matrix led to the improvement of the rheological properties in the GMRG sample.

## 4. Conclusion

A new type of MRG comprised of 10 wt% graphite (GMRG) was prepared in this study and their rheological properties were compared with referring to MRG. The results showed that GMRG sample has improved rheological properties in terms of apparent viscosity, shear stress, yield stress and viscoelastic

properties. The GMRG has a better apparent viscosity in both off- and on-state conditions that reflected to the contribution of graphite that filled the spaces between CIPs. Interestingly, the yield stress of GMRG has showed 38.8% improvement, which demonstrated the contribution of graphite towards stable structure. Furthermore, an existing of graphite in GMRG had result in widen the yield stress from 0A to 3A for about 32.7% higher compared to MRG. Thus, this study has provided an information that by the addition of graphite as a carbon-based additive, has altered and improved the microstructure in strengthening the CIPs columnar structure that enhanced GMRG properties.

## Declarations

### Acknowledgement

This work was supported by the Ministry of Higher Education under Fundamental Research Grant Scheme (FRGS/1/2020/TK0/UTM/02/57). This work was also supported by Skim Pensyarah Lantikan Baru (SLB2111).

### Author contributions

N.A.M.N: Prepared-wrote the original manuscript, carried out the experiment, analyzed the data. N.N., N.M: Supervised the work, contribute in writing, reviewing, and editing the manuscript, funding acquisition. N.A.N.: Supervised the work, contribute in writing, reviewing, and editing the manuscript. U., S.A.M, S.A.A.A., M.K.S.: Involves in writing, reviewing, and editing the manuscript.

### Competing interests

The authors declare no competing interests.

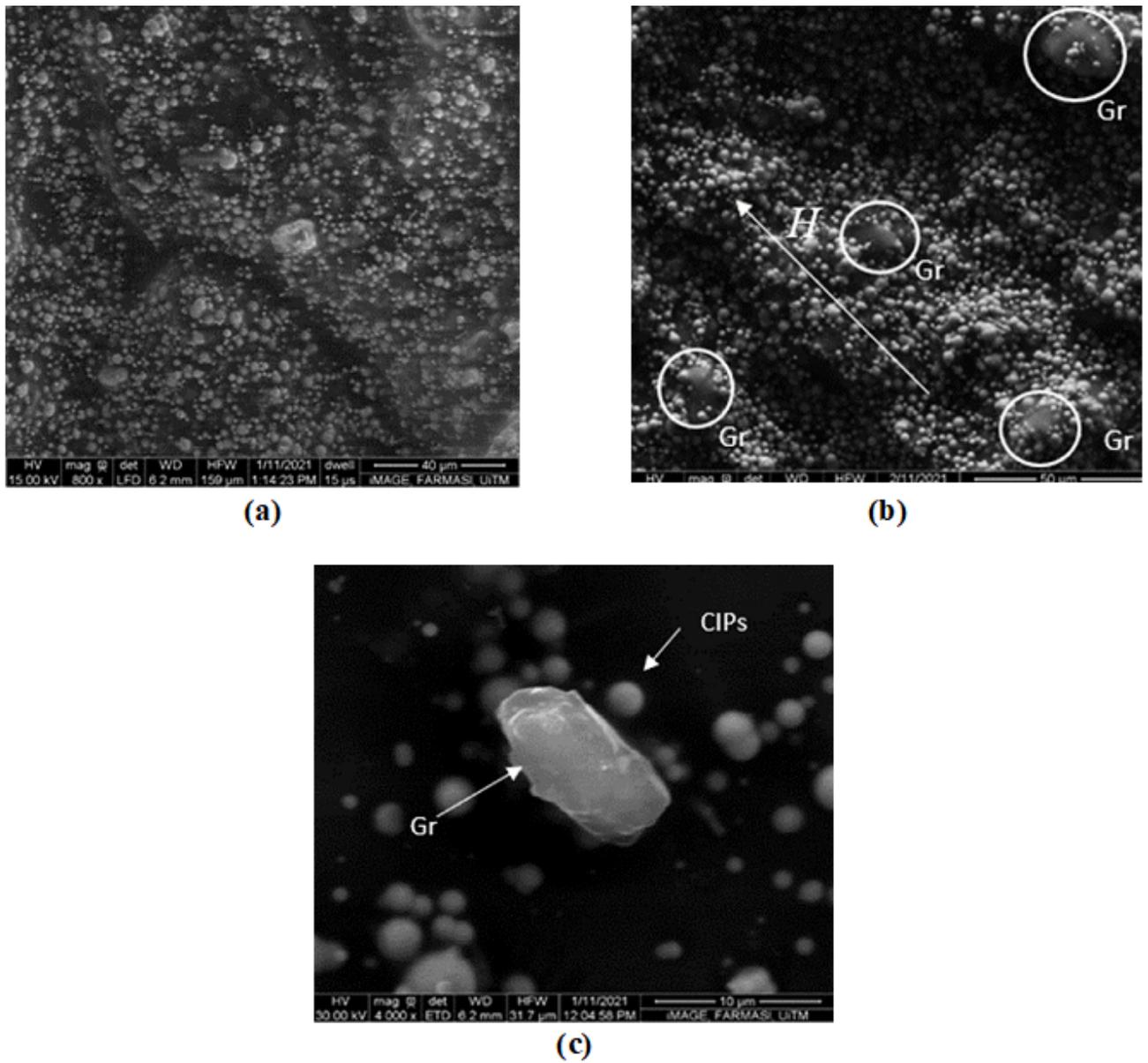
## References

1. Rankin, P. J., Horvath, A. T. & Klingenberg, D. J. Magnetorheology in viscoplastic media. *Rheol. Acta* **38**, 471–477 (1999).
2. Park, J. H., Kwon, M. H. & Park, O. O. Rheological Properties and Stability of Magnetorheological Fluids using Viscoelastic Medium and Nanoadditives. *Korean J. Chem. Eng.* **18**, 580–585 (2001).
3. Karis, T. E., Kono, R. N. & Jhon, M. S. Harmonic analysis in grease rheology. *J. Appl. Polym. Sci.* **90**, 334–43 (2003).
4. Wang, H., Li, Y., Zhang, G. & Wang, J. Effect of temperature on rheological properties of lithium-based magnetorheological grease. *Smart Mater. Struct.* **28**, 035002 (2019).
5. Mohamad, N., Mazlan, S. A., Ubaidillah, Choi, S. B. & Nordin, M. F. M. The Field-Dependent Rheological Properties of Magnetorheological Grease Based on Carbonyl-Iron-Particles. *Smart Mater. Struct.* **25**, 095043 (2016).

6. Burhannuddin, N. L. *et al.* Physicochemical characterization and rheological properties of magnetic elastomers containing different shapes of corroded carbonyl iron particles. *Sci. Rep.* **11**, 868 (2021).
7. Sahin, H., Gordaninejad, F., Wang, X. & Fuchs, A. Rheological behavior of magneto-rheological grease (MRG). *Act. Passiv. Smart Struct. Integr. Syst.* **2007** 6525, 65250D (2007).
8. Ahamed, R., Choi, S. B. & Ferdaus, M. M. A state of art on magneto-rheological materials and their potential applications. *J. Intell. Mater. Syst. Struct.* **29**, 2051–2095 (2018).
9. Sukhwani, V. K. & Hirani, H. A Comparative Study of Magnetorheological-Fluid-Brake and Magnetorheological-Grease-Brake. *Tribol. Online* **3**, 31–35 (2008).
10. Kim, J. E., Ko, J. Do, Liu, Y. D., Kim, I. G. & Choi, H. J. Effect of medium oil on magnetorheology of soft carbonyl iron particles. *IEEE Trans. Magn.* **48**, 3442–3445 (2012).
11. Mohamad, N. *et al.* Dilution dependent of different types of redispersing oils on magnetorheological greases. *Int. J. Eng. Technol.* **8**, 107–111 (2017).
12. Wang, K., Dong, X., Li, J., Shi, K. & Li, K. Effects of silicone oil viscosity and carbonyl iron particleweight fraction and size on yield stress for magnetorheological grease based on a new preparation technique. *Materials (Basel)*. **12**, 7–9 (2019).
13. Mohamad, N., Ubaidillah, Mazlan, S. A., Choi, S. B. & Halim, N. A. Improvement of magnetorheological greases with superparamagnetic nanoparticles. *MATEC Web Conf.* **159**, 8–12 (2018).
14. Tarmizi, S. M. A. *et al.* Incorporation of cobalt ferrite on the field dependent performances of magnetorheological grease. *J. Mater. Res. Technol.* **9**, 15566–15574 (2020).
15. Tian, T. F., Li, W. H., Alici, G., Du, H. & Deng, Y. M. Microstructure and magnetorheology of graphite-based MR elastomers. *Rheol. Acta* **50**, 825–836 (2011).
16. Shabdin, M. K. *et al.* Material characterizations of gr-based magnetorheological elastomer for possible sensor applications: Rheological and resistivity properties. *Materials (Basel)*. **12**, 391 (2019).
17. Pang, H., Xuan, S., Liu, T. & Gong, X. Magnetic field dependent electro-conductivity of the graphite doped magnetorheological plastomers. *Soft Matter* **11**, 6893–6902 (2015).
18. Thakur, M. K. Influence of Graphite Flakes on the Strength of Magnetorheological Fluids at High Temperature and Its Rheology. *IEEE Trans. Magn.* **56**, 4600210 (2020).
19. Radouane, N. *et al.* Thermal, electrical and structural characterization of zinc phosphate glass matrix loaded with different volume fractions of the graphite particles. *J. Non. Cryst. Solids* **536**, 119989 (2020).
20. Sengupta, R., Bhattacharya, M., Bandyopadhyay, S. & Bhowmick, A. K. A review on the mechanical and electrical properties of graphite and modified graphite reinforced polymer composites. *Prog. Polym. Sci.* **36**, 638–670 (2011).
21. Porfir'ev, Y. V. *et al.* Effect of Thickeners on Low-Temperature Greases. *Chem. Technol. Fuels Oils* **55**, 540–551 (2019).

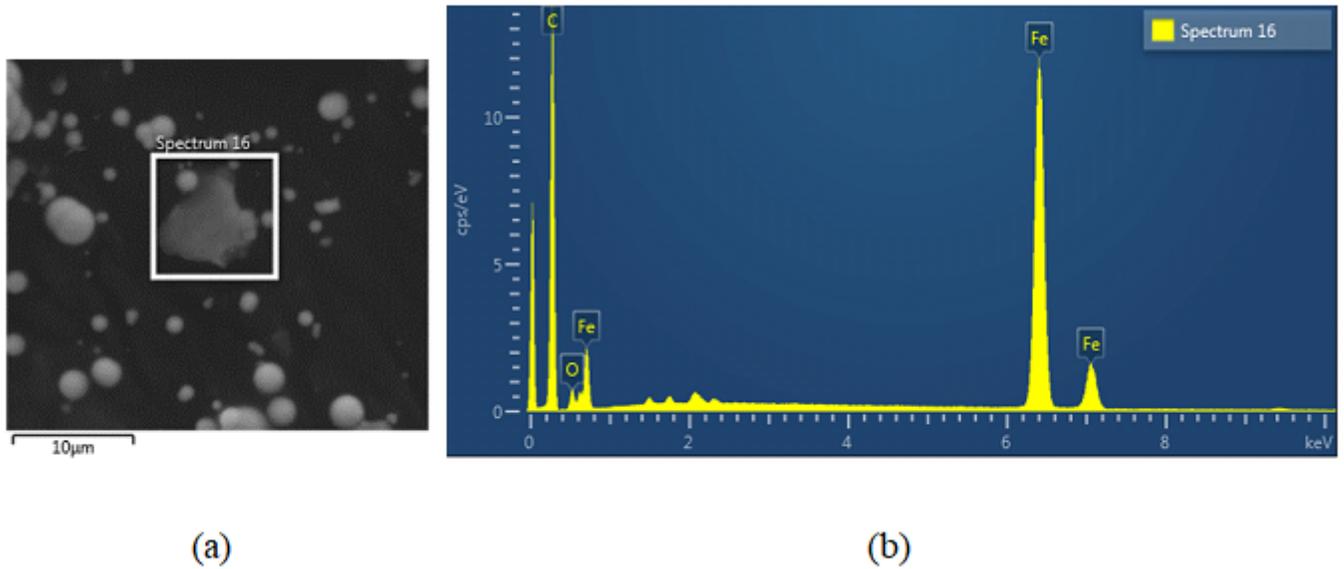
22. Paszkowski, M., Olsztyńska-Janus, S. & Wilk, I. Studies of the kinetics of lithium grease microstructure regeneration by means of dynamic oscillatory rheological tests and FTIR-ATR spectroscopy. *Tribol. Lett.* **56**, 107–117 (2014).
23. Park, B. O., Park, B. J., Hato, M. J. & Choi, H. J. Soft magnetic carbonyl iron microsphere dispersed in grease and its rheological characteristics under magnetic field. *Colloid Polym. Sci.* **289**, 381–386 (2011).
24. Kavlicoglu, B. M., Gordaninejad, F. & Wang, X. Study of a magnetorheological grease clutch. *Smart Mater. Struct.* **22**, 125030 (2013).
25. Wang, X. & Gordaninejad, F. Study of magnetorheological fluids at high shear rates. *Rheol. Acta* **45**, 899–908 (2006).
26. Czarny, R. & Paszkowski, M. The influence of graphite solid additives, MoS<sub>2</sub> and PTFE on changes in shear stress values in lubricating greases. *J. Synth. Lubr.* **24**, 19–29 (2007).
27. Khan, S. A. & Lazoglu, I. Development of additively manufacturable and electrically conductive graphite – polymer composites. *Prog. Addit. Manuf.* **5**, 153–162 (2019).
28. Zhang, W. L., Kim, S. D. & Choi, H. J. Effect of graphene oxide on carbonyl-iron-based magnetorheological fluid. *IEEE Trans. Magn.* **50**, 2500804 (2014).
29. Prasad, M. H. & Gangadharan, K. V. Synthesis and Magneto Mechanical Properties of MR Grease. *Int. J. Eng. Res. Technol.* **3**, 2369–2372 (2014).
30. Tian, Y., Jiang, J., Meng, Y. & Wen, S. A shear thickening phenomenon in magnetic field controlled-dipolar suspensions. *IEEE Trans. Inf. Theory* **39**, 1031–6 (1993).
31. Baptista, R. *et al.* Effect of high graphite filler contents on the mechanical and tribological failure behavior of epoxy matrix composites. *Theor. Appl. Fract. Mech.* **85**, 113–124 (2016).
32. Ubaidillah, B., Sutrisno, J., Purwanto, A. & Mazlan, S. A. Recent Progress on Magnetorheological Solids: Materials, Fabrication, Testing, and Applications. *Adv. Eng. Mater.* **17**, 563–97 (2015).
33. Xu, Y., Gong, X. & Xuan, S. Soft magnetorheological polymer gels with controllable rheological properties. *Smart Mater. Struct.* **22**, 075029 (2013).
34. Gong, X., Xu, Y., Xuan, S., Guo, C. & Zong, L. The investigation on the nonlinearity of plasticine-like magnetorheological material under oscillatory shear rheometry. *J. Rheol.* **56**, 1375–1391 (2012).
35. Helgeson, M. E., Wagner, N. J. & Vlassopoulos, D. Viscoelasticity and shear melting of colloidal star polymer glasses Viscoelasticity and shear melting of colloidal star polymer glasses. *J. Rheol.* **51**, 297–316 (2007).
36. Wang, H. *et al.* Characterization of nonlinear viscoelasticity of magnetorheological grease under large oscillatory shear by using Fourier transform-Chebyshev analysis. *J. Intell. Mater. Syst. Struct.* **32**, 614–631 (2020).

## Figures



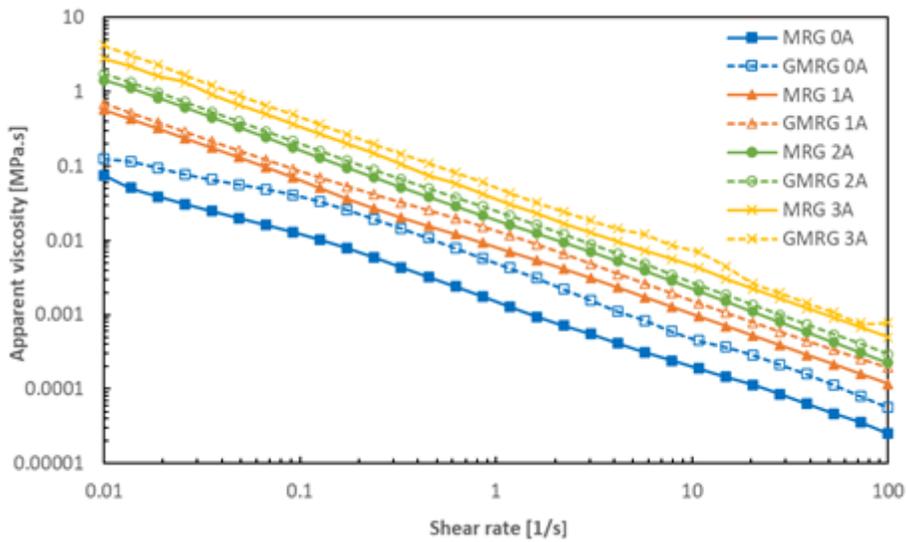
**Figure 1**

ESEM image of GMRG samples; (a) with the absence and (b) under pre-treated of magnetic field of 0.1T, and (c) with enlarged view of CIPs and graphite under absence of magnetic field at magnification of 4000x.



**Figure 2**

(a) Selected microstructure area and (b) EDX graph for sample GMRG.



**Figure 3**

Apparent viscosity of MRG and GMRG at shear rate range from 0.01 to 100s<sup>-1</sup> in different applied currents.

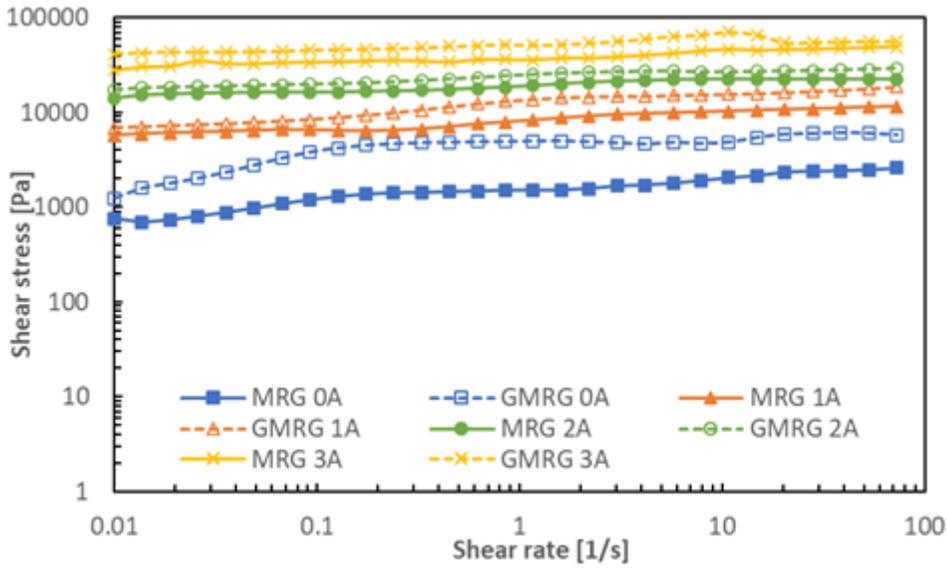


Figure 4

Comparison of shear stress between MRG and GMRG at varied magnetic field strengths.

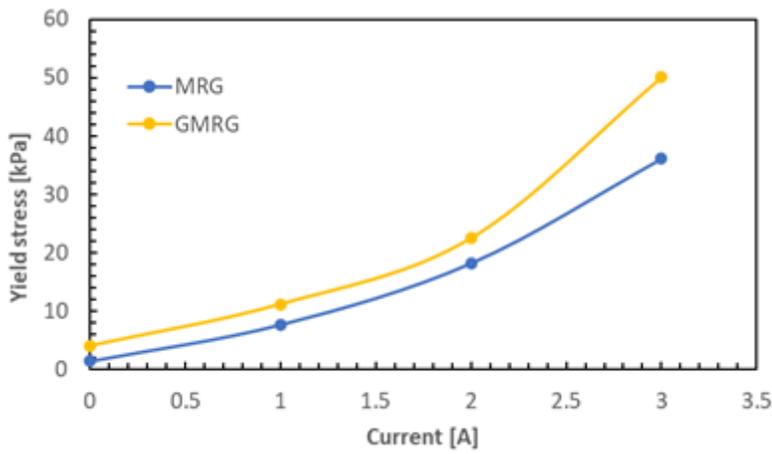
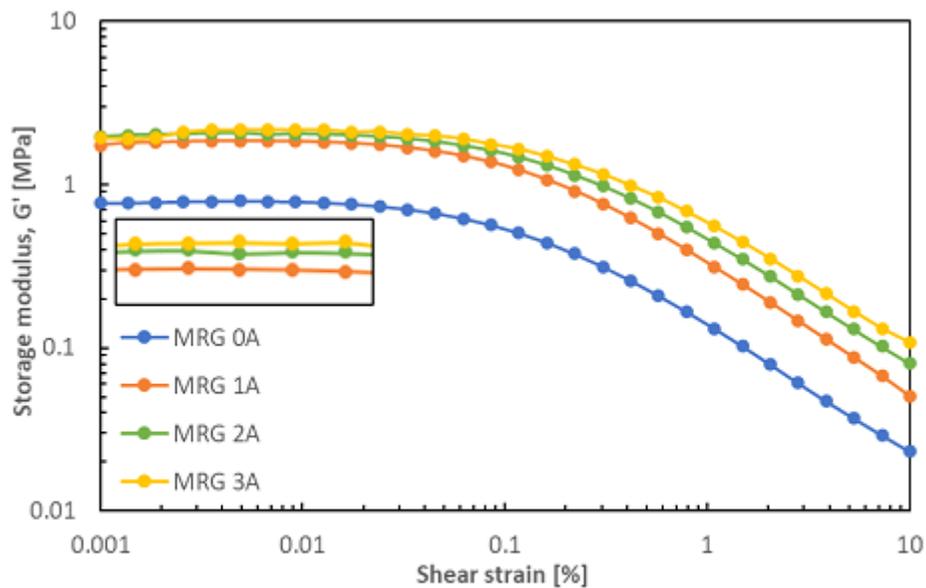
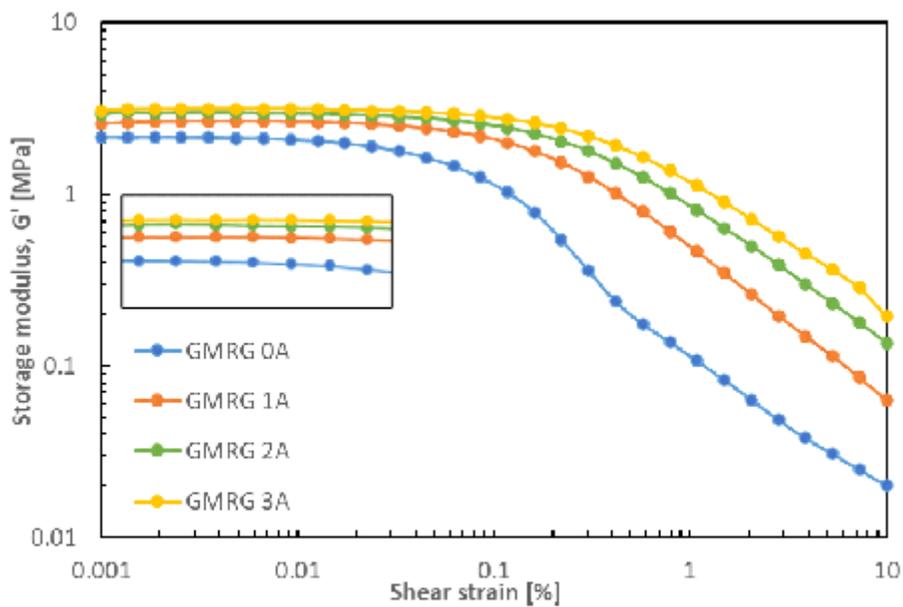


Figure 5

Yield stress of MRG and GMRG under various applied currents from 0 to 3A.



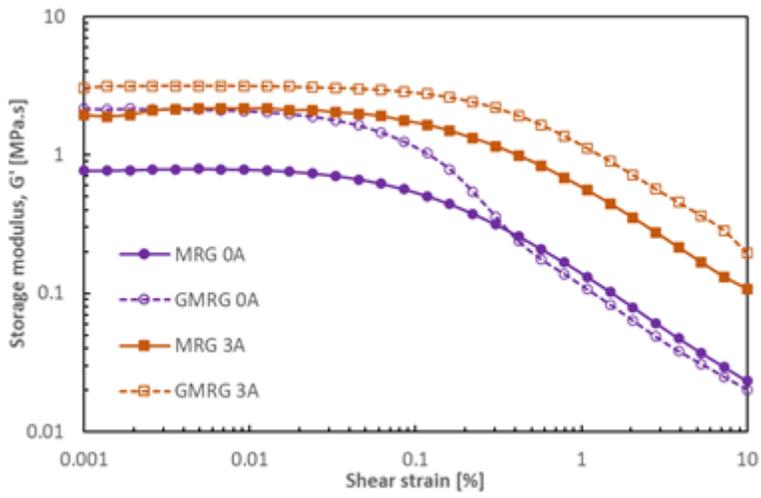
(a)



(b)

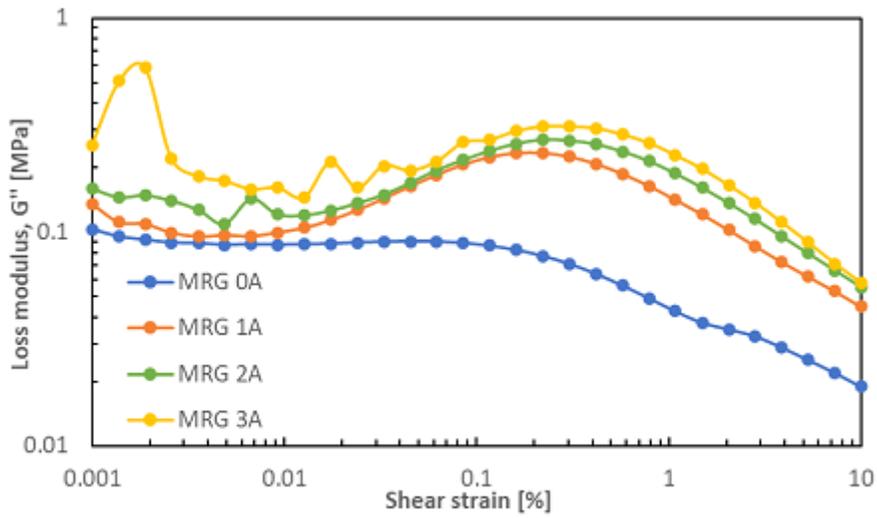
Figure 6

Storage modulus,  $G'$  function of shear strain for (a) MRG and (b) GMRG at different applied currents.

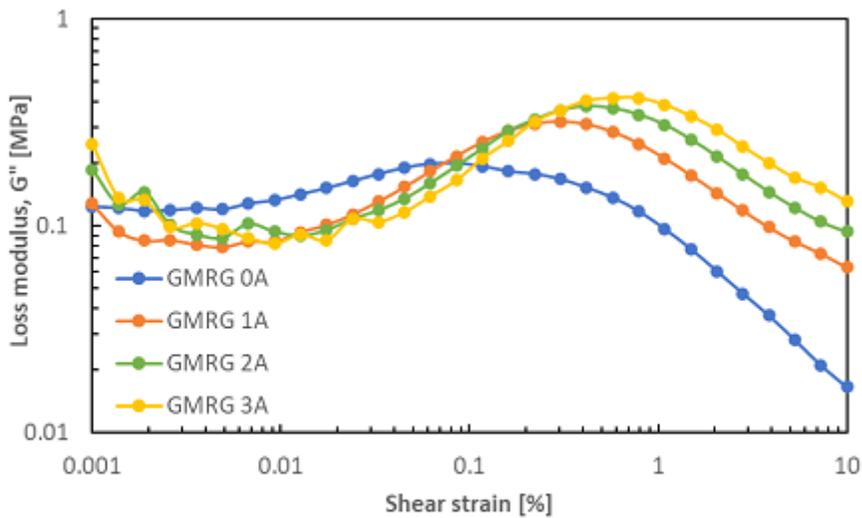


**Figure 7**

Storage modulus,  $G'$  as a function of shear strain for MRG and GMRG at off- and on-state conditions.



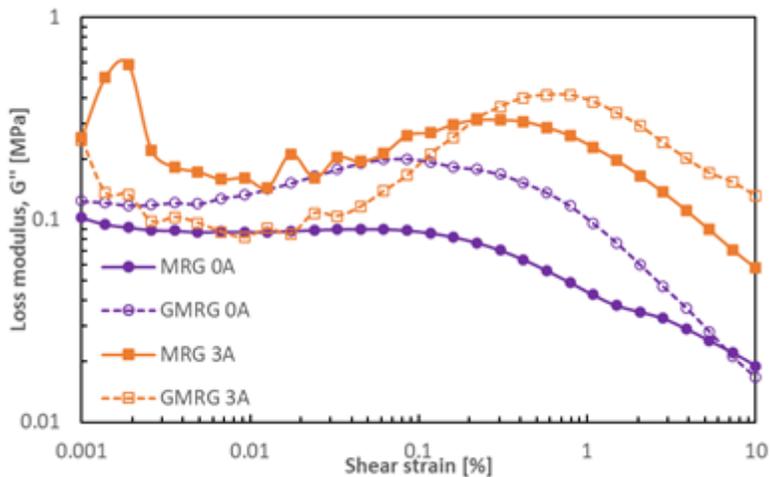
(a)



(b)

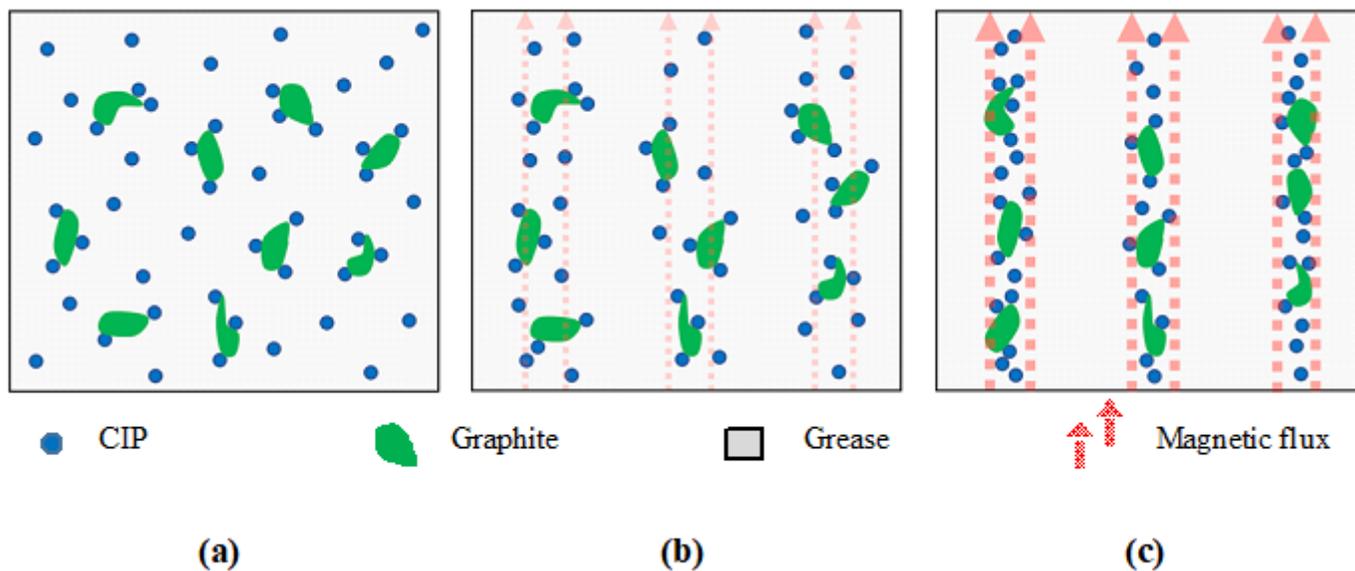
Figure 8

Loss modulus,  $G''$  of (a) MRG and (b) GMRG as function of shear strain at different applied currents.



**Figure 9**

Loss modulus,  $G''$  function of shear strain for MRG and GMRG at off- and on-state conditions.



**Figure 10**

Schematic arrangement of CIPs and graphite in MRG: (a) in the absence of magnetic field, (b) with the introduction of magnetic field, and (c) with further increases of magnetic field.