

The Effect of Rheological Feature of Graphene Oxide-Involved Emulsion on its Corresponding Lubricity Performance

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This study aims to investigate the effect of rheological property of graphene oxide (GO)-involved emulsion on its corresponding lubrication behavior. The friction of GO-involved emulsion could be reduced by a factor of 1.2 as compared to that of the base emulsion. The worn surface of steel specimen after lubrication demonstrated that the presence of GO shrank the wear size and depth by 7.6 % and 14 %, respectively. The welding loading of GO involved oil also gave rise to a 315 kg whereas that of the base oil only allowed 250 kg. The improved lubricity performance of GO involved emulsion was attributed to the fact that GO enhanced either anchoring reaction between oil drops (increased oil viscosity) or chemical reaction toward steel substrate. Furthermore, the variations of d_{50} oil droplet sizes for base and GO emulsion were 16.9 and 11.2 %, respectively, suggesting a slight improvement on emulsion stability with the presence of GO during lubrication. This result was plausible since carboxyl groups within GO were protonated at acidic condition such that the GO particles turned to less hydrophilic and form GO aggregates, resulting to a prevention of coalescence between oil drops. On the other hand, the I_D/I_G values of GO within emulsion before and after lubrication were 0.61 and 0.66, respectively, revealing a trivial increase in the disorderliness degrees after lubrication, which was corresponding to the change of d_{10} lattice, as determined in HR-TEM image. Based on the XPS analysis on worn steel substrate, in the case of GO emulsion, the organic O signals showed up at the expense of metal oxide (FeO and Fe₂O₃), verifying that GO participated in the formation of tribological film. Supporting evidence was also available by Fe2p1/2 spectra that the existence of Fe-C_xH_y for GO emulsion confirmed the GO binding toward the steel substrate, reinforcing the strength of tribological oil film. In addition, the protective nature of GO was also advantageous to the prevention from tribological oxidation of the steel substrate.

Keywords: Graphene oxide, Emulsion, Tribology, Interfacial rheology

1. INTRODUCTION

Oil-in-water emulsion (o/w emulsion) has been practically confirmed to provide excellent lubricity and cooling performance for metal processing, as it can form a hydrodynamic film more efficiently than that is expected from their rheological properties⁽¹⁾. Rather than the pure oil lubricant, o/w emulsion has also been made a preferred option in terms of environmental and economic aspects. In fact, numerous studies have examined the lubrication behavior and mechanism of o/w emulsion. For example, Kumar et al.⁽²⁾ indicated that the surfactant was able to compensate the repulsive force between negatively charged oil droplet and substrate, which could propel the droplets to a very close proximity to the substrate and gave rise to reduced friction. Kanagarasala⁽³⁾ also indicated that the surfactant concentration close to CMC could minimize the droplet size and subsequently allow for easy transport of droplets to the substrate, which was advantageous to the reduction of

friction. In another study, Xu et al.⁽⁴⁾ proposed that the lubrication behavior was significantly dependent on the oil distribution of droplet size, which dominated the portion of oil entering the wedge area between two nonparallel surfaces. Some field studies related to the application of o/w emulsion on the steel rolling were also carried out⁽⁵⁻⁶⁾. Dubey et al.⁽⁵⁾, for example, revealed that o/w emulsion with relatively large droplet size, high saponification value, and low emulsion stability could give relatively high film thickness and low friction, which was corresponding to the actual observation within the industry. Despite the o/w emulsion lubricant has been applied on metal processing for decades, the researches related to the emulsion lubricant are still booming, as lots of promising and state-of-the-art additives are being developed for relatively high energy efficiency and improved environmental protection during metal processing⁽⁷⁻⁸⁾.

In general, an effective additive has a high surface activity, which could offer a stable o/w emulsion along

with an excellent adherence of oil over substrate material surface, and subsequently enhances the lubricity. In this regard, the graphene-family material, possessing a 2D honeycomb network formed from one atomic layer of carbon atoms, advances a wide array of new tribology applications because of its intriguing structure and fascinating properties. More specifically, the graphene-family material could be a potential alternative to most solid lubricant additives due to its specific features including excellent lubricity, high thermal conductivity, and impermeable property⁽¹⁾. To the best of our knowledge, Berman et al. first revealed⁽⁹⁾ that graphene constituted a new class of lubrication for steel surfaces which offered decreased wear by almost 4 orders of magnitude and a reduced coefficient of friction by a factor of 6. Their following study aiming at the superlubricity contributed by graphene showed that the coefficient of friction was reduced from c.a. 1 for bare steel to 0.15 for steel covered by a low concentration of graphene flake⁽¹⁰⁾.

On the other hand, some studies have also been dedicated to the modification of o/w emulsion by the graphene-family materials. Wu et al.⁽¹¹⁾ examined the emulsifying and lubrication properties of the myristyltrimethylammonium bromide-modified graphene oxide (GO) and indicated that the improved lubricity of o/w emulsion was attributed to the strong film-forming ability contributed by GO. In another study, Liang et al. reported that a 81% reduction in coefficient of friction and 62% reduction in wear scar were provided by GO existing in the o/w emulsion. Furthermore, GO-involved o/w emulsion has been investigated in terms of the effect of pH, salt concentration, reduction of GO, and so on⁽¹²⁾. McCoy et al.⁽¹³⁻¹⁴⁾ also demonstrated the pH dependent flocculation behavior of o/w emulsion in the presence of GO, and revealed that the interfacial charge between o/w emulsion and GO interface was critical to the stability of acidic GO-involved emulsions⁽¹³⁾. Their following study presented a report on how GO behaved as a surfactant-like or a particle-like material⁽¹³⁾. In another study, the emulsion with a high salinity (5% NaCl) could be significantly stabilized for almost one year by GO, which was likely attributed to the combination of the strong electrostatic repulsion rendered by the charged edge and the weak van der Waals attraction from the extremely thin plate⁽¹⁵⁾.

Despite numerous studies have been working on the graphene-family lubricant, only limited studies examined the effect of GO on the lubricity with respect to the oil rheology, especially it appears that no study was dedicated to how GO affects the rheological behavior of o/w emulsion during lubrication. Also, the effect of emulsifying capability of GO on the lubrication performance of o/w emulsion is rarely explored. This study, therefore, aims to investigate the lubrication behavior of GO involved emulsion in terms of the rheological

aspects. Surface characterizations including SEM/TEM, Raman, and XPS analysis on the worn surface of steel substrate after lubrication were also investigated, in order to elucidate the lubrication mechanism contributed by GO.

2. METHODS

2.1 Materials, Oil and Emulsion Preparation

The base oil consisted of 10wt% naphthenic oil (T110, NYNAS), 0.6wt% dialkyl cationic surfactant (4DAC, ADEKA), 2wt% non-ionic surfactant polyoxyethylene tridecyl ether (1307, sinopol), and 0.8wt% polymer dispersant (Hypermer A70, Croda) which was balanced by palm oil while additional 0.2wt% C-SiO₂ (C-SiO₂, Enerage Inc.) added into the base oil was designated as GO-involved oil which contained c.a. 500 ppm GO. C-SiO₂ slurry used for the preparation of GO-involved oil received no further treatment. Both oils, prior to emulsion preparation, were subjected to stirring at 300 rpm for 5 min with a regular magnetic stirrer. The base and GO-involved emulsions were made by mixing 3wt% prepared oil and 97wt% D.I. water at 8,000 rpm for 30 min in a homogenizing mixer (Model 2.5, Primix). The prepared emulsions were immediately used for experiments with no further storage.

2.2 Characterization of Prepared Oil and Emulsion

2.2.1 Rheological Behavior of Prepared Oil

The thixotropic behaviors of the prepared base and GO-involved oils were determined by the hysteresis between the first and second curves which corresponded to increasing (0-600 s⁻¹) and decreasing shear rates (600-0 s⁻¹), respectively (MCR 302, Anton Paar). Each interval lasted for 10 min at 50°C with the viscosities of oil samples recorded as a function of the shear rate during the forward and backward shearing. The viscosity dependence on temperature affected by GO was also examined with the temperature ranging from 50°C to 90°C at a shear rate of 600 s⁻¹. The adsorption kinetics of GO from the bulk oil phase to the interface between water and oil phase was also examined in this study. This experiment was performed in a strain-controlled mode using MCR 302 rheometer adapted for interfacial rheometry. A biconical disk was used to oscillate region of interface between the prepared oil and water at a controlled torque while the cell remained stationary. Time sweeps were performed at 10% strain and 30 Hz frequency every 10 s for 60 min with the temperature maintained at 25°C by a Peltier plate below the cell.

2.2.2 Emulsion Droplet Size Analysis

The emulsion droplet sizes before and after lubrication tests were determined using the electrical pulse

counting techniques with a 100- μm aperture probe (MultisizerTM 3, Beckman Coulter). The narrow aperture between the electrodes constitutes the sensing zone through which suspended particles pass. The magnitude of electrical conductive variation is dependent on the droplet size distribution when passing the emulsion through a small aperture in the electrode. In other words, each particle displaces its own volume of electrolyte in the sensing zone. The volume displaced is recorded as a voltage pulse, and the height of each pulse is proportional to the volume of the particle. The device was programmed to count and determine the sizes of all particles with diameters between 2 and 60 μm at intervals of 0.2 μm .

2.3 Lubrication Tests of Emulsion Solution

The lubrication behaviors of prepared emulsions were carried out on a tribometer (MCR 302, Anton Paar), with a ball-on-three-plate test figuration. A GCr15 steel ball (Kugel Pompel, Vienna, Austria) with a diameter of 12.7 mm was selected as mating material for the plate specimen (WT%: 0.24 C, 0.41 Si, 2.3 Mn, 0.04 P, 0.04 S; balance Fe) which sited on a self-centering system. Before the tests, the plates were polished to obtain a surface roughness of $R_a \sim 0.04\mu\text{m}$ and then flushed with a stream of dry N_2 gas to remove the debris which was followed by the sonication with acetone for 10 min before placing the plates on the aforementioned self-centering system. The emulsion sample was immediately poured into the sample chamber right after the preparation. The chamber temperature and the load were kept constant during the tests at $T = 60^\circ\text{C}$ and $F = 40\text{N}$, respectively. The Stribeck curve was obtained with a logarithmical increase of the sliding speed from 10^{-7} to 1.4 m/s. Three replicates of friction and wear measurements were performed to minimize data scattering.

2.4 Characterization of Worn Plate Surface

The 3D topography of the wear scar on the specimen after the friction test was characterized by Olympus OLS4000 laser scanning confocal microscope (Olympus Ltd., Tokyo, Japan). Before the observation, the worn specimens were ultrasonically cleaned with acetone followed by drying with pressured nitrogen gas. The microstructures of the specimens before and after the friction test were investigated by using a Raman analysis with a laser source of 532nm in wavelength and the scanning range of $100\sim 3500\text{cm}^{-1}$ (Horiba LabRam HR800). The chemical compositions of specimens before and after the friction test were also analyzed by X-ray photoelectron spectroscopy (XPS, The PHI Quantera A type). The Mg K α monochromatic radiation was used as the excitation source with pass energy of 29.35eV. The specimens for Raman and XPS analysis after friction were subject to only drying with pressured nitrogen gas.

3. RESULTS AND DISCUSSION

3.1 Lubrication Performances of Base and GO Involved Emulsions

As shown in Figure 1(a) (Stribeck curve), the initial peak in the curve represents the point where the lubrication system is in the transition from static friction to the kinetic state of motion⁽¹⁶⁾. No significant difference was observed between base and GO emulsions, since the transition was only dependent on the surface texture of the substrate. At low sliding speed regimes, both curves show that the friction gradually increase with increasing sliding velocity. The value of friction coefficient (CoF) moves sharply up until the breakaway point which is characteristic of elastic deformation of the system. As compared to the base emulsion, a relatively high CoF in the case of GO emulsion indicates that the introduction

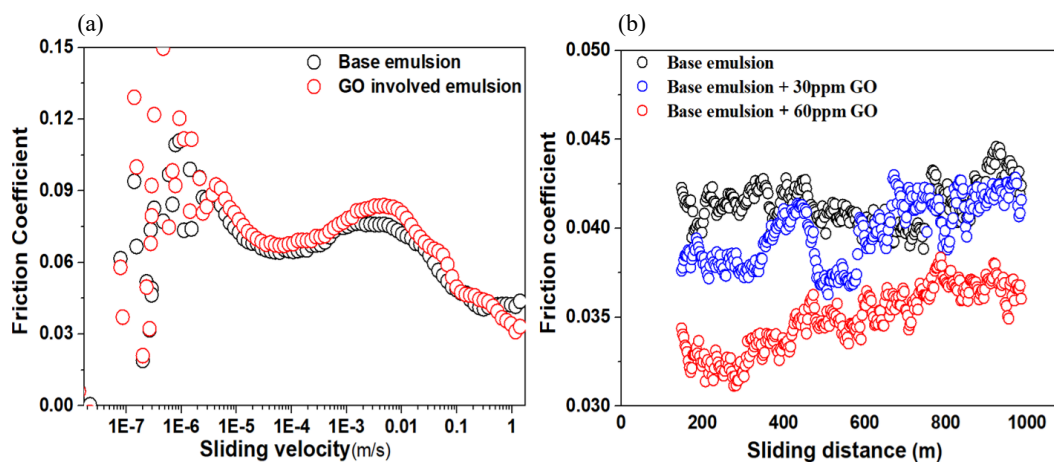


Fig.1. (a) Stribeck curve of analysis for base and GO involved emulsions, (b) the friction coefficient at a sliding speed of 1.4m/s as a function of GO amount within the emulsion.

of GO likely causes an increase in the asperity between mating surfaces in the low sliding speed regime. On the other hand, a further increase in sliding speed is indicative of the transition from boundary lubricant to hydrodynamic lubrication and there is a significant difference in the friction behavior over the high sliding speed regime. The Stribeck curves show a crossover as the sliding speed is increased beyond a certain level ($\sim 0.4\text{m/s}$), suggesting that the presence of GO in the emulsion could increase the component of hydrodynamic lubrication. A long-term observation on the CoF at a high sliding speed (1.4 m/s) shows that the friction in the case of GO emulsion is reduced by a factor of 1.2 (Fig.1 (b)). The result above clearly exhibits that the presence of GO preferentially contributes on the hydrodynamic lubrication rather than boundary one. In terms of the extreme pressure performance of base and GO involved oil, the welding loading of GO involved oil gave rise to a 315kg level, while that of base oil only allowed 250kg. Furthermore, the last none-seizure loadings of base oil and GO involved oil were 50 and 63kg, respectively. The extreme pressure property of GO involved oil was enhanced by c.a.20% as well. This result suggests an improved strength of tribofilm in the presence of GO, which could be responsible for relatively low CoF for a long-term period.

The improved lubricity contributed by GO could be further confirmed by investigating the 3D profile of

worn surfaces (Fig.2). These images present that the presence of GO shrinks the wear size and depth by 7.6% and 14%, respectively. The phenomenon is corresponding to that obtained from the friction examinations. The presence of GO in o/w emulsion literally reinforces the tribofilm strength, as evidenced by the lubrication and 3D profile analysis results. A possible mechanism is further investigated chemically and physically in the following sections.

3.2 Rheological Properties of Lubrication Oil with and Without the Presence Of GO

The Stribeck curve is the variation of friction in relation to the duty parameter which is a combination of the sliding speed, lubricant viscosity and loading level. Given that either base or GO emulsion subjects to the similar loading level and sliding speed during friction tests, the Stribeck curve is only dependent on the lubricant viscosity. In fact, the tribofilm usually has a rather high viscous modulus compared to steel substrate, e.g., they subject to shear much easier. The tribofilm, namely, behaviors as a sacrifice layer to protect underneath substrate. In other words, the rheological properties of GO-involved oil may play a pivotal role in dictating the lubrication behavior of the system after the plate-out of oil from the emulsion. The thixotropic behaviors of base and GO involved oil are demonstrated in Figure 3(a). Both cases show the shear thinning behavior while GO

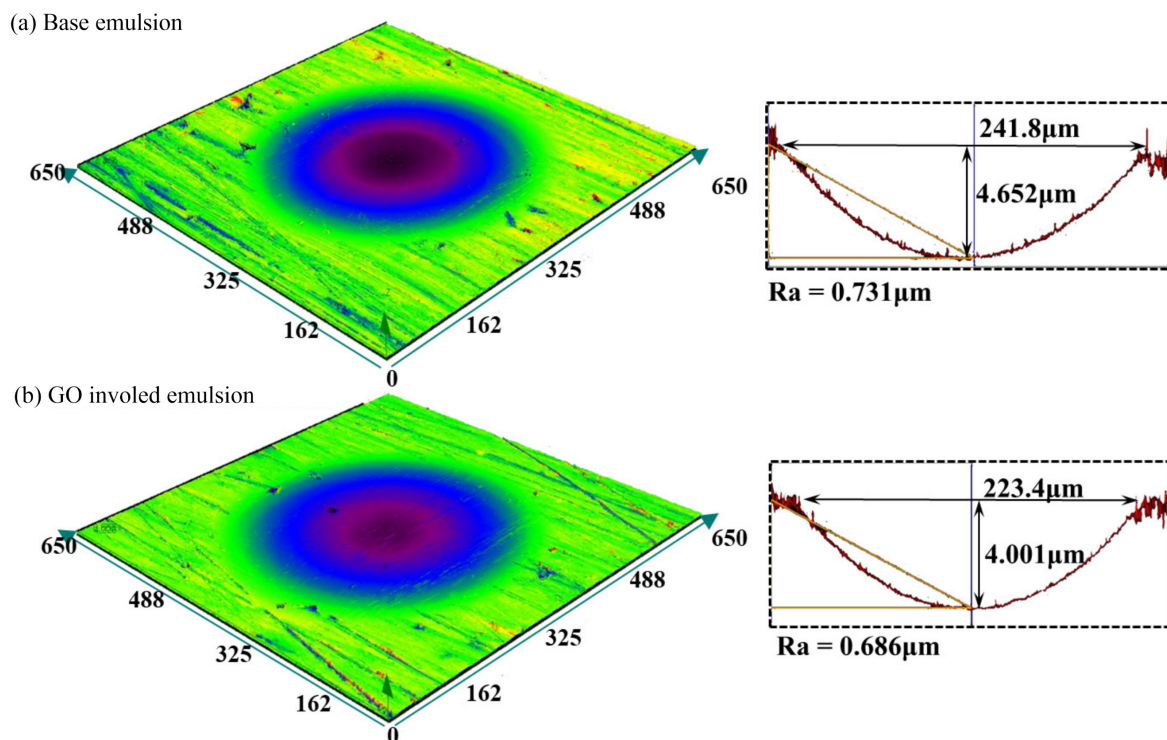


Fig.2. Analysis of worn steel surface lubricated with (a) base emulsion and (b) GO involved emulsion.

involved oil demonstrates relative high viscosities over the designated shear rate range. This result suggests that GO involvement is superior in the resistance to the applied shear, which may be attributed to that GO possibly anchors oil drops to each other. In other words, GO could allow more intermolecular interactions between oil droplets⁽¹⁷⁾. The internal structure of GO-involved oil exhibits more shear tolerance, thereby demonstrating the relatively high viscosity, which partly accounts for the improved lubricity of GO involved emulsion.

On the other hand, the thixotropic behavior is more prominent while GO is present in the oil, as demonstrated by the area constructed between the forward and backward curves shown in Figure 3 (a). This result is beyond the expectation as the internal structure of GO-involved oil should be reinforced as described above. The thixotropic curve shows no sign for network restoring of the GO involved system and these enhanced interactions and aggregates between oil droplets by GO seem to be broken with increasing the applied shear

force. In addition, Figure 3 (b) presents no difference of the temperature-dependent viscosity between bare and GO-involved oil (Fig.3 (b)), which suggests that GO has no positive effect on the thermal stability of oil. According to the demonstrated results, the strength reinforcement of the oil film by GO is likely dominated by the lubrication condition.

3.3 Emulsion with and Without the Presence of GO

3.3.1 Emulsion Stability After Lubrication

In addition to the rheology effect on the emulsion lubricity, the effect of oil droplet size on the lubricity of emulsion has been in dispute for years. Some studies claim a small oil droplet size is favorable to the lubricity while some verify the formation of a plate-out oil film is determined by the portion of relatively large oil droplet⁽⁴⁻⁶⁾. In our study, the oil droplet size distribution of the emulsion before and after friction tests were also examined (Fig.4). Prior to the lubrication tests, the emulsions

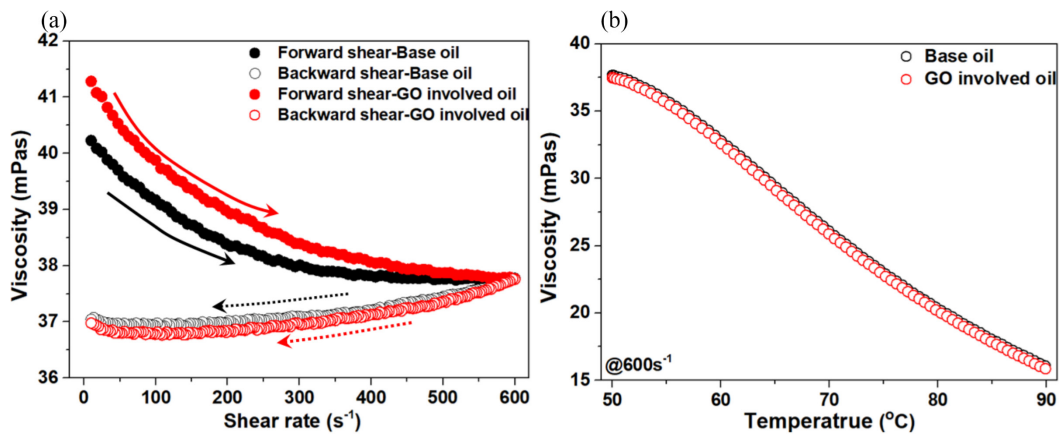


Fig.3. (a) Thixotropic analysis \ (b) temperature dependence of viscosity for base and GO involved oil.

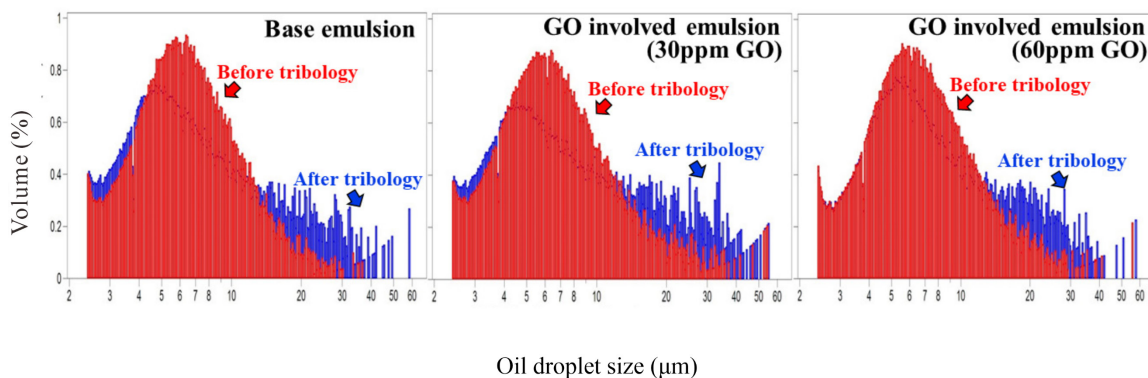


Fig.4. The size distribution of oil droplets before and after lubrication as a function of GO amount within the emulsion.

of oil droplet size distribution with and without GO show monomodal and narrow curves, with a d_{50} droplet size of 6.52 ± 5.69 and $6.30 \pm 5.55 \mu\text{m}$, respectively. Only a small portion of oil droplet shifted to a small size region in the presence of GO. Therefore, the effect of oil droplet size distribution on the lubricity could be neglected in terms of the experimental conditions in our study. Despite no significant change in the oil droplet size was observed, this result is still in parallel to some graphene-related studies which demonstrated that the relatively small oil droplet size can be achieved with the presence of the graphene-family materials⁽¹¹⁾.

Comparatively, the size distribution of the oil droplets became wider and more skewed toward higher diameter values after lubrication (Fig.4). This result is attributed to that, under lubrication shear, the oil droplets might be pressed together to enter the thin contact zone, leading to oil droplet aggregation. An increase in oil droplet size is also as a result of the thermodynamically unstable emulsion during lubrication. However, an interesting phenomenon was observed that the presence of GO appeared to improve the emulsion stability during lubrication (i.e., improved endurance to friction). The variations of d_{50} oil droplet sizes for base and GO emulsion after lubrication were 16.9 and 11.2%, respectively. This result was plausible since carboxyl groups within GO are protonated at acidic condition such that the GO particles turn less hydrophilic and form suspended GO aggregates.

The existence of such aggregates were likely to create a steric hindrance to prevent the flocculation of emulsion oil droplet⁽¹⁸⁾. A similar result was also indicated by McCoy et al.⁽¹³⁾ who attributed the remarkable increase in stability of GO involved emulsion to the charge reversal of the o/w interface. A complementary explanation is also available in that an improved stability of GO involved emulsion is partly related to the increased oil viscosity shown in Figure 3 (a). Since the involvement of GO is capable of enhancing the viscosity and emulsion stability, the plate-out volume of GO involved emulsion is relatively small at a low-speed regime, yielding in worse friction. This phenomenon is corresponding to the Stribeck curve shown in Figure 1 (a).

3.3.2 The Effect of GO on the O/W Emulsion Formation

In order to elucidate the GO effect on the o/w emulsification, the interface oscillations were made for 1 h at a frequency of 30 Hz and 10% strain. In the case of either base or GO emulsion, all the G' are several orders larger than G'' over the observation time, indicating the interface at oil and water phase is characteristic of fluid-like material. Furthermore, either G' or G'' increases with oscillation time, which is indicative of increasing strength/ numbers of interactions, i.e., emulsion formation.

Figure 5 shows that the complex viscosities of both emulsions increase with time which could be explained by the build-up of the emulsion network at the interface between oil and water phases. The inset figures demonstrate that the milky layer between oil and water phase shows up after the initial few seconds of oscillation and subsequently increases with time. Interestingly, the complex viscosity of GO emulsion is rather high as compared to the base emulsion. This result suggests that the presence of GO favors the development of the emulsion layer. Namely, the oil-water interface in the presence of GO tends to be a more rigid system as compared to that of a base emulsion. This phenomenon is plausible, as the more the oil droplet disperses throughout the water phase at the o/w interface, the higher the shear stress is exhibited. Our finding also corresponds to the report by Giudice and Shen⁽¹⁷⁾ who indicated that GO sheets significantly increased the elastic and viscous modulus of the composites; and simplified the cross-linking process between polymer chains.

In fact, whether GO is thermodynamically favorable for emulsification, appears to be dependent on the oil polarity which dominates GO adsorption on o/w interface. For example, Thickett et al.⁽¹⁹⁾ indicated that the involvement of polar oil had less driving force for GO adsorption on the o/w interface, i.e., emulsification with GO in the presence of polar oil was a less thermodynamically favorable process. In terms of this point, we found that the emulsification could be improved by replacing 5wt% of naphthenic oil with the paraffinic oil with less polar compound (Fig.5). This result is not only corresponding to the effect of oil polarity on the emulsification but is consistent with the previous reports which revealed that GO with amphiphilic characteristic is thermodynamically favorable to the emulsification⁽²⁰⁾.

3.4 Surface Characterization of Specimen Before and After Lubrication

3.4.1 HR-TEM on the Specimen After Lubrication

TEM image shown in Figure 6 presents an oil film with the thickness of 60~80nm where GO with the particle size ranging from 10 to 60 nm spread throughout the oil film. In addition, the HRTEM image of GO shows the measured lattice spacing of 0.21-0.24 nm, which is consistent with (100) plane for a graphitic facet (0.21 nm)⁽²¹⁾. A slight variation in lattice spacing of (100) plane may arise due to intercalation of -OH groups in graphene layers⁽²¹⁻²²⁾. The mechanism of slight expansion of the (100) plane is still under investigation in our following study but likely resulting from the lubrication process.

3.4.2 Raman Analysis on the Specimen Before and After Lubrication

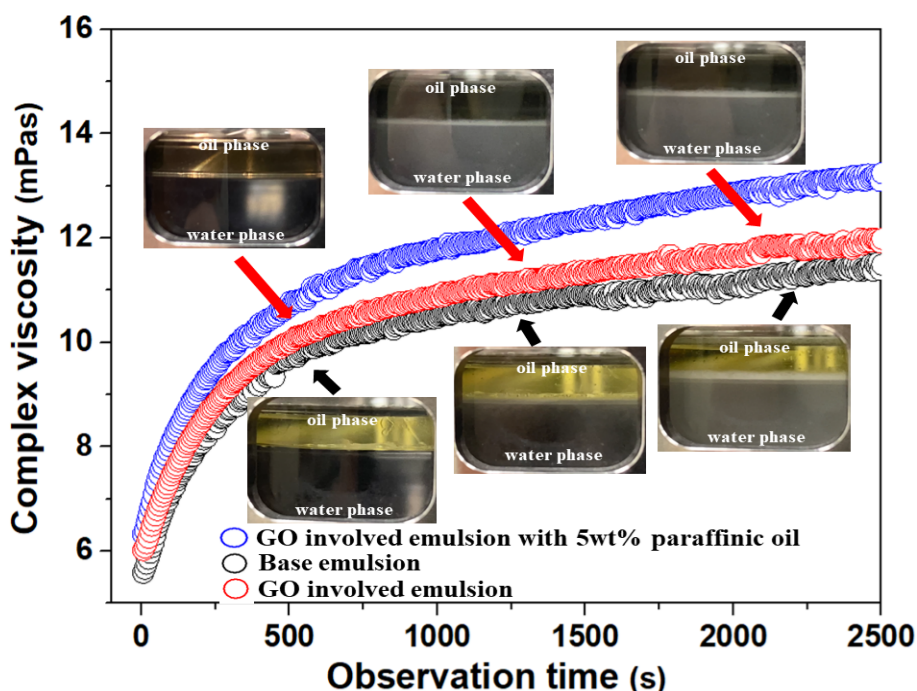


Fig.5. Time sweep rheology measurement at the interface between water and oil phases with and without the presence of GO and paraffinic oil.

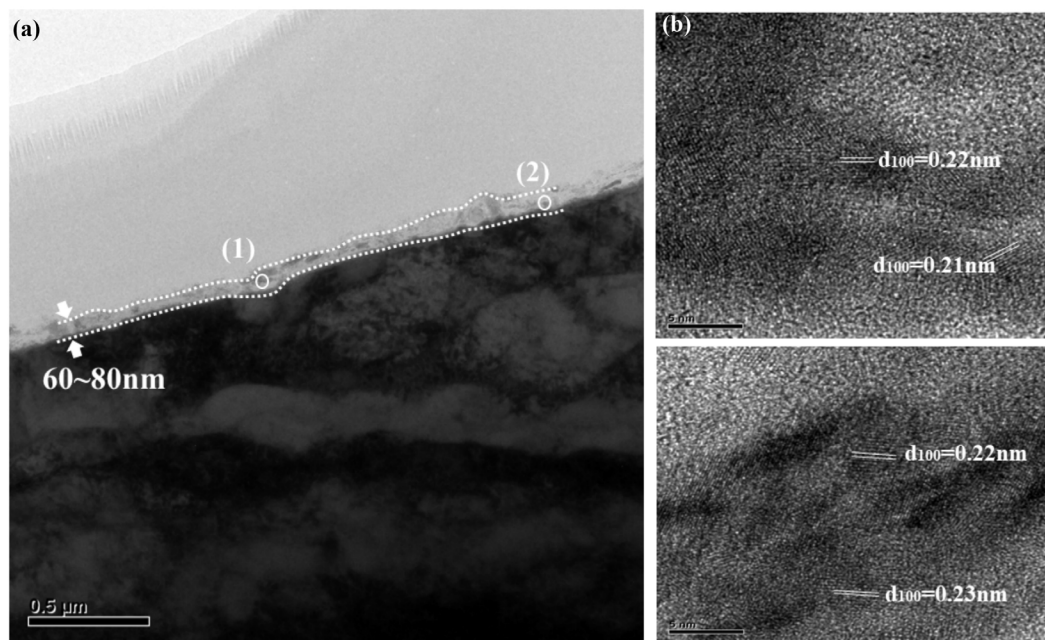


Fig.6. (a) The cross-sectional HR-TEM observation of worn steel surface lubricated with GO involved emulsion \ (b, c) lattice spacing of GOs.

In the case of GO emulsion, the peaks showing at ~ 1350 and ~ 1580 cm^{-1} are characteristic of D-band and G-band of GO, respectively. D band represents the disorder structure (sp^3 carbon) and defects, while G bands corresponds the order structure (sp^2 carbon

atoms) in hexagonal lattice (Fig.7)⁽²³⁾. The intensity ratio of D and G bands (I_D/I_G) is an indication of the degree of defects and chaos of carbon-based materials. The I_D/I_G values of GO emulsion before and after lubrication are 0.61 and 0.66, respectively, suggesting a slight increase

in the disorderliness degrees of GO after lubrication. This result is corresponding to the lattice change after lubrication which is shown in HR-TEM image. Another possibility related to the increased disordered structure of GO is attributed to either the increased amount of graphitized wear debris entrapped in the worn spots, or GO transformation into a disordered graphitic structure⁽²⁴⁻²⁵⁾. Consequently, the less degree of Van der Waals forces in graphite's atomic structure can easily be broken to allow the layers of GO to slide off and deposit onto a substrate surface to provide improved lubrication. On the other hand, as compared to the base emulsion, the GO signal on the worn surface in the case of GO-involved emulsion is prominent, revealing that GO may spread over the oil film, which is partly responsible for the enhanced lubricity of GO-involved emulsion.

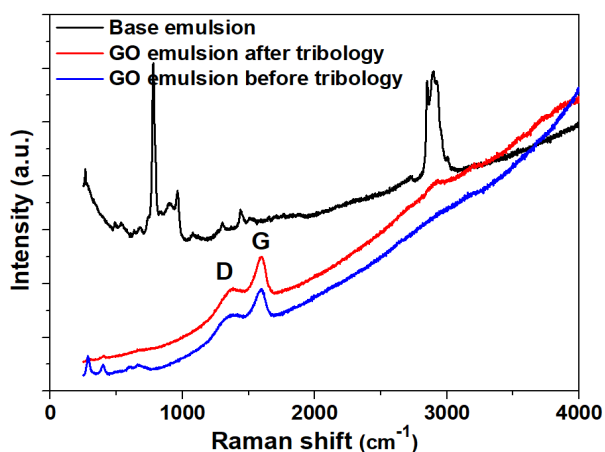


Fig.7. Raman spectra of a worn steel surface lubricated with base and GO involved emulsions.

3.4.3 XPS Analysis on the Specimen Before and After Lubrication

For deconvolution analysis of XPS spectra, C element on the worn surface after lubrication is characterized into three peaks at binding energies of 284.6, 285.3, and 288.7eV which are typical of C-C, C-O, and C=O bindings, respectively. No obvious difference of peak intensity was observed between the deconvolution analysis of base and GO emulsion. However, the peaks of C1s spectra in the case of GO emulsion shift to a relatively high binding energy as compared to that of a base emulsion (Fig.8 (a) and 8 (d)). The decrease in electron density of C element is possibly owing to the binding of C=O within GO to steel substrate. This result is also consistent with the previous study reported by Wu et al.⁽¹¹⁾ who indicated that GO could be bound more tightly with the steel surface through the formation of chemical bonds.

Further solid evidence is also available from O1s and Fe2p1/2 spectra. By comparing the O1s spectra shown in Figures 8 (b) and 8 (e), the organic O signals show up at the expense of metal oxide (FeO and Fe₂O₃) in the case of GO emulsion, verifying that GO participates in the lubrication, and then simultaneously prevents the steel specimen from lubrication oxidation. Regarding the Fe2p1/2 spectra (Fig.8 (c) and 8 (f)), while no obvious Fe-C_xH_y signal was observed in the case of the base emulsion, the existence of Fe-C_xH_y for GO emulsion also revealed the GO binding toward the metal surface, resulting to the formation of GO involved oil film⁽¹¹⁾. In combination with the previous results, it could be concluded that GO reacting with the specimen

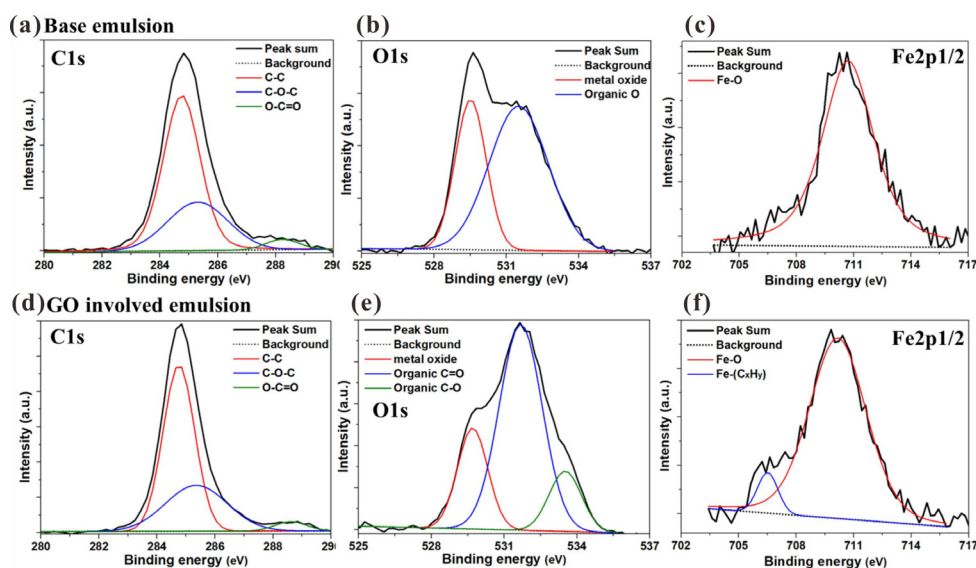


Fig.8. Partial XPS spectra of C1s, O1s and Fe2p1/2 peaks for a worn steel surface Lubricated with (a)(b)(c) base and (d)(e)(f) GO involved emulsions.

steel surface during the formation of oil film, was in favor of the lubricity between mating surfaces.

4. CONCLUSION

In this study, the effect of graphene oxide (GO) on the lubricity of oil-in-water emulsion was investigated in terms of the rheological aspect. Our results demonstrated that GO could be a promising additive for oil-in-water emulsion as a result of the unique antiwear and friction reduction performance. According to the Stribeck curve, a relative high friction was observed in the boundary lubrication while a marked reduction either in friction or wear was attained in high sliding speed regimes (elasto-hydrodynamic lubrication). The lubricity of GO emulsion increased with increasing in sliding velocity, which was attributed to the increased viscosity of oil and favorable affinity of GO toward steel substrate, as supported by the thixotropic and XPS results. In addition, Raman analysis and HR-TEM observation showed that the microstructure of GO was only slightly affected during lubrication, revealing that GO may be applicable for the long-term use. On the other hand, the interfacial rheological results revealed that the emulsification between oil and water phase in the presence of GO was improved while the presence of less polar oil could give rise to an additional driving force for emulsion formation. Our study, however, showed that GO provided only trivial contribution on the emulsion stability as compared to that of the base emulsion, which may be also a desirable characterization for lubricity improvement. Furthermore, the protective nature of GO prevented steel substrate from oxidation (tribo-corrosion) when presenting at sliding contact interface. To sum up, upon considering the corresponding rheological feature of oil contributed by GO, this promising material opens up a new realm of lubrication application which was previously unattainable.

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