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RESEARCH

Tribological Properties of Aqueous Carboxymethyl Cellulose/Uncaria Gambir Extract as Novel Anti-Corrosion Water-Based Lubricant

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A B S T R A C T

In this work, a novel anti-corrosion water-based lubricant is prepared. Carboxymethyl cellulose and Uncaria Gambir Extract, all in powder form, were dispersed in water. Corrosion, stability, and tribology test with pin-on-disk reciprocating tribometer were studied. The newly obtained lubricants were designated as Uncaria Gambir (UG), i.e. UG0 (0% volume Uncaria Gambir extract in Carboxymethyl cellulose, UG1 (1% volume Uncaria Gambir extract in Carboxymethyl cellulose, and UG2 (2% volume Uncaria Gambir extract in Carboxymethyl cellulose). The addition of Uncaria Gambir Extract into Carboxymethyl cellulose solution delivered good stability, low corrosion rate, and stable friction coefficient. UG1 has the best stability (no sedimentation), corrosion, and tribological performance. After a one-hour friction test, UG2 showed the best performance in terms of CoF, indicated by a 7.6% CoF reduction, compared to UG0. This finding indicates the potential of Uncaria Gambir Extract as an additive for an anti-corrosive water-based lubricant for machining application.

1. INTRODUCTION

The world's demand for renewable energy is increasing every year [1]. It cannot be avoided that earth will run out of natural resources. Lubricant is a substance needed to reduce friction and wear from two rubbing objects in the mechanical field. Having a low friction property is one of the crucial parameters in developing new materials [2]. It is estimated that thirty percent of the world's energy resources are required to overcome friction [3]. Nowadays, lubricants circulating in the market are mainly derived from crude oil, which will be limited in the future. Also, this type of lubricant has many drawbacks due to its toxic substance or additives [4]. Precautionary measures feed to be taken to avoid natural resource crises. Therefore, an alternative is required to maintain the natural resource of crude or mineral oil. One of the ways is by using vegetable oil. Vegetable oil is high in lubricity, viscosity, and flash point [5]. However, vegetable oil is low in oxidative stability and is relatively expensive compared to water. Employing water as a lubricating base fluid has been studied by several researchers. It is environmentally friendly, widely available, inexpensive, and has no hazardous substance, making water a potential substitute for conventional lubricants [6,7]. Recently, water-based fluids have been used as cutting fluid and hydraulic fluid due to their excellent cooling performance, low pollution, and other advantages compared to lubricating oil [8].

Although, water has low viscosity, low boiling point, and is corrosive to metal, which is unsuitable if applied as a lubricating fluid for machining application. Recent studies clarified that adding additives or chemical compounds can improve the properties of water. Nanoparticles have become one of the most common substances used for lubricant additives due to their high thermal conductivity and could enhance the viscosity of the base fluid [9,10]. For instance, adding graphene to water can reduce the friction coefficient wear rate up to 77% and 90%, respectively [11]. Adding 0.1% MoS_2 nanosheets into water greatly improved the worn volume by 45% compared to pure water [12]. Recently, research on hybrid nanoparticles has been studied frequently. Xie et al. used SiO_2/graphene combinations, providing superior results. Adding 0.1wt% SiO2 and 0.4wt% graphene in water lowered the coefficient of friction (CoF) by 49% and wore volume by 80% compared to 0.5wt% graphene in water [13]. Furthermore, Huang et al. investigated that a pair of 1:1 Graphene oxide and Al_2O_3 in the mass ratio of 0.12% reduced CoF to 60% [14].

However, even though it can drastically improve properties, some nanoparticle mentioned above is not environmentally friendly and very expensive. One of the most abundant biopolymers is cellulose which is produced from algae, tunicates, certain bacteria, and plant-based materials [15,16]. This type of compound is also suitable as a lubricant additive. It has been proved that adding 0.6wt% of Bacterial Cellulose to the Polyolester Oil (POE) could enhance the wear rate by 49% compared to pure POE [16]. Furthermore, sonicated cellulose with nano-sized particles could improve CoF by 25% compared to pure water [17]. Carboxymethyl cellulose (CMC), also known as cellulose gum, has many advantages if applied as a lubricant additive: high solubility with water, biodegradable, non-toxic, and potentially as viscosity modifier [18,19]. A recent study has confirmed that CMC applied in rapeseed oil decreased wear depth up to 79% compared to its base fluid [20]. Although, applying water as a lubricant could corrode metal, which would be very detrimental [21]. Moreover, reports on the study of lubricant that investigating its corrosion resistance is very rare. *Uncaria gambir* extract (UGE) can be used to improve the corrosion resistance of water. *Uncaria Gambir* is hydrophilic and contains 80% catechin, which is reported to exhibit anti-oxidative activity [22]. It has been reported that UGE in a cooling-water solution remarkably provides a corrosion inhibition efficiency of up to 70% [23]. Although, the catechin of Uncaria Gambir has a cross-linking capability that, if applicate in a high amount, will decrease the cellulose solubility in water. The undissolved cellulose will affect the stability of the solution because agglomeration will occur. Therefore, it is crucial to determine the optimum concentration of UGE. In order to evaluate lubricant performance as a machining application, tribological and corrosion resistance tests are essential to be conducted. Moreover, lubricant with anti-corrosion capability is needed to maintain the cutting tool's lifespan [24]. As the author is aware, the tribological and corrosion test of aqueous lubricant anti-corrode derived from CMC and UGE have not yet been explored. This present work report aqueous lubricant of CMC/UGE performance. The corrosion test, stability, tribology, and morphologies of the worn object were investigated.

2. MATERIALS AND METHOD

2.1 Materials

CMC with 99.6% purity and filtered with an 80-mesh sieve was purchased by Changsu Wealthy Science and Technology, China. UGE, which was used in our previous work, was supplied by PT. Andalas Sitawa Fitolan, Indonesia, comprises 90% predominant catechins [22]. Analytical-grade distilled water was purchased from PT. Brataco, Indonesia.

2.2 Lubricants preparation

The composition of the samples is shown in Table 1. The CMC powder was mixed with distilled water using a plate stirrer (Daihan Scientific MSH-200) at 300 rpm and 50C for 30 minutes. Soon after it was soluble, UGE powder was dissolved in distilled water and centrifuged for 30 minutes. The precipitate was removed carefully, and its supernate was added to the CMC-distilled water mixture and stirred for another 30 minutes. Sonicator (SJIA-1200W, Ningbo Yinzhou Sjia Lab Equipment, China) with 600W power and 30 minutes duration was employed to enhance the stability of the samples [25]. The detail, including the UGE morphology used in this work, is available in our previous work [22,26].

Table 1. The composition of water-based lubricant.

Samples	Mass Fraction (%)		
	CMC	UGE	Distilled Water
UG0	1	0	99
UG1	1	1	98
UG2	1	2	97

2.3 Stability of the lubricants

The stability of the samples was investigated by capturing the samples (Canon, SX540 HS) which known as the photographic capturing approach. Each sample is stored in a 20 ml glass vial at various time intervals of 0, 24, and 48 hours.

2.4 Corrosion test

A simple yet effective qualitative corrosion test is conducted in this work, consistent with the previously reported work [27–29]. Gray cast iron disk (FC25) was immersed with the lubricant in beaker glass. It was placed in a drying oven at 55 ± 2 °C for 24 h. The corrosion level of the samples was evaluated according to the GB6144-85 reference standard [29]. An Optical Microscope (Primotech, Zeiss, Germany) was used to investigate the disk's surface soon after the test.

2.5 Tribological properties

The tribological performances of the lubricant samples were measured with a pin-on-disk reciprocating tribometer which was also utilized in our previous work [15]. A gray cast iron (FC25) with 30 mm in diameter and 5 mm in thickness was utilized as the disk. It was polished with emery paper until grid #320 for 10 minutes at 200 RPM. The pin, directly taken from a bearing (AISI52100), is 4 mm in diameter and 20 mm in length. A normal force of 11.76 N was applied as the load. The disk movement completed in 3 s with a sliding distance of 15 mm. The sliding speed of the test was between 0 to 0.2 m/s. The friction force was acquired by using a load cell. The friction tests were conducted for one hour. The topography of the wear surface was examined using Optical Microscope (Primotech, Zeiss, Germany).

3. RESULTS AND DISCUSSIONS

3.1 Stability of the lubricants

The qualitative sedimentation measurement via photo capturing was used to observe the lubricant's stability, as presented in Figure 1. This method has been employed in our previous works [15,16,30]. After the preparation of the lubricants, it can be seen in Figure 1(a) that CMC has good stability in water due to its hydrophilicity [31]. Visually, the addition of UGE did not affect the stability of the lubricants on the first day. Figure 1(b) shows the stability of the samples after being left overnight. There was sedimentation for UG2. This is probably due to the excessive UGE that contains 80% catechin [26]. This catechin has a cross-linking capability that decreases the CMS's solubility to water leading to agglomeration [32,33]. Moreover, catechin has limited solubility in water [34]. After two days (Figure 1(c)), the UG2 was completely agglomerated and sedimented. On the other hand, UG0 and UG1 showed good stability.

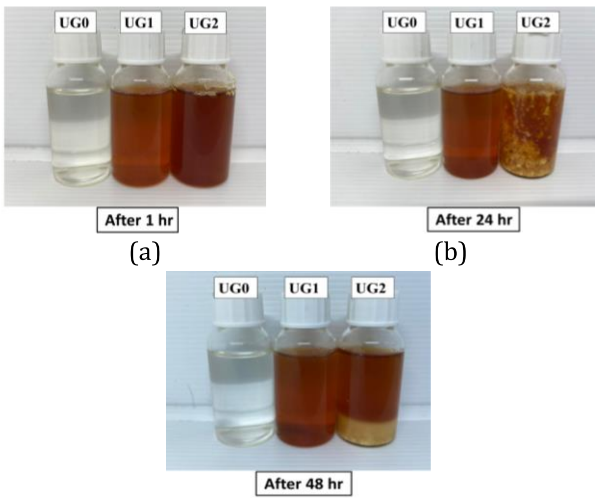
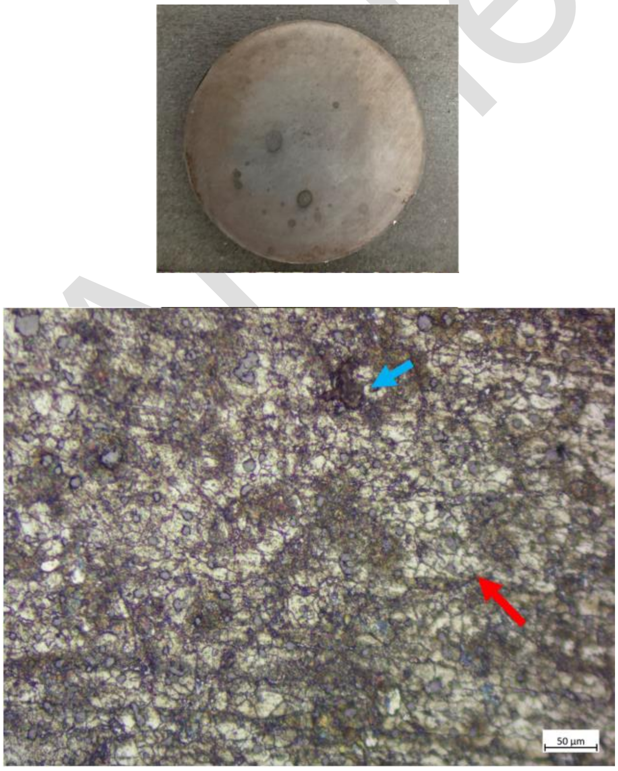


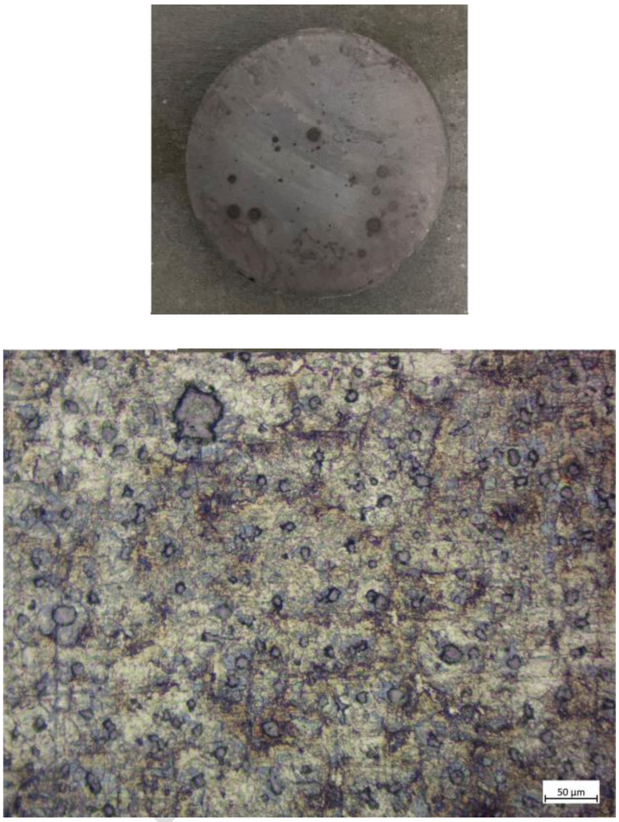
Fig. 1. Stability observation of the lubricants after 1 hour of preparation (a); after 24 hours of preparation (b); after 48 hours of preparation (c).

3.2 Corrosion test

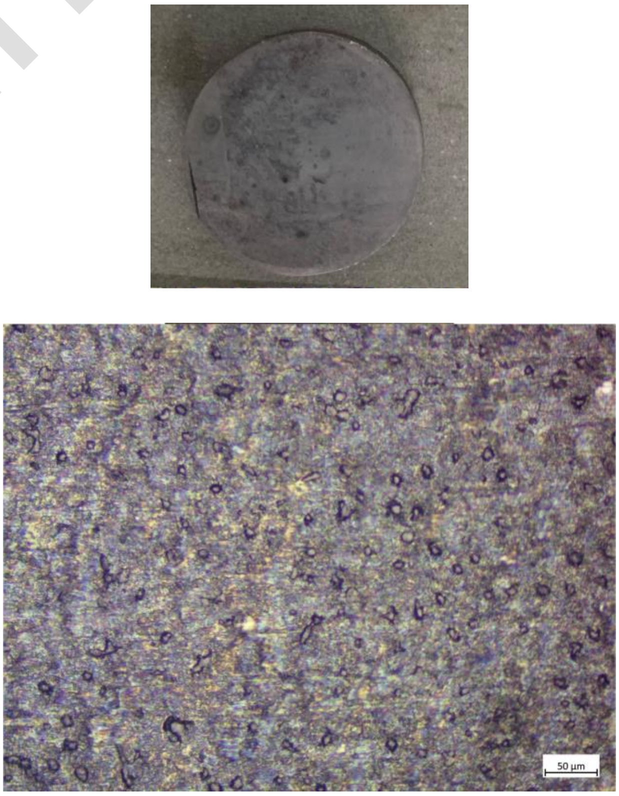
To investigate whether UGE can reduce metal corrosion by water, a corrosion test was conducted. According to Figure 2(a), it can be seen that the surface of the cast iron is presented with a crack (red arrow), black spot or passivated pit (blue arrow), and discoloration occurred (Figure 2(a)), indicating that the corrosion of disk immersed with UG0 is severe [29,35]. On the other hand, the disk lubricated with UGE has weaker corrosion compared to UG0. In Figure 3(b), it is observable that there is no discoloration. Figure 2(b) proves that UG1 delivers no crack and passivated pit to the grey cast iron disk. Even so, pores are present. Increasing the amount of UGE reduces the corrosion rate of the disk. Figure 2(c) shows the performance of the corrosion test with UG2. Compared to UG1 and UG0, there is no crack available, it has fewer pores, and the surface is relatively smooth, indicating that UGE partially alleviated metal corrosion in water. Some of the previous work also investigated mild steel immersed in an aqueous UGE solution. The SEM EDX analysis showed that UGE could inhibit oxygen elements up to 81% compared to immersion without UGE [36]. Furthermore, it was stated that catechin could reduce the corrosion rate of iron by up to 39% [37].



(a)



(b)



(c)

Fig. 2. Optical microscope images of the surface of gray cast iron disk after corrosion test in (a) water; (b) UG1; (c) UG2.

3.3 Tribological test

Figure 3(a) shows the CoF of the pin-on-disk test lubricated with all samples (UG0, UG1, and UG2) in full sliding time, while Figure 3(b) for the sliding time of 3000 to 3600 s. Figure 3(a) shows that UG0 has the lowest CoF at about the first 1500 seconds of the test, indicating good lubrication performance. These results prove that CMC can be a friction modifier that enhances water's anti-friction properties due to its high viscosity nature [38]. However, at sliding time between 1400 to 1500, the CoF increases. This is probably due to the high corrosion rate of the rubbing surface, leading to rough surface formation. This is consistent with Figure 2(a). After running the test for 30 minutes, the CoF of UG0 is increased continuously. According to Figure 3(b), which shows the last 5 minutes of running, it can be seen that UG0 has reached the maximum CoF value. The average CoF (sliding time of 3000 to 3600s) of UG0 is calculated and is found to be higher than UG1 and UG2. Figure 4 shows the surface of the pin after the test. It can be disclosed from Figure 4(a) that a high corrosion rate occurs, indicated by the scar (red arrow). This finding is consistent with the corrosion test.

On the other hand, lubricant with the addition of UGE has a relatively high value of CoF compared to UG0. Referring to the stability test, this is probably due to the high amount of catechin in gambir that decreased CMC's water solubility to water [32]. The insoluble CMC will form agglomeration, forming new asperities that could increase the friction and wear because of the failure to form stable tribofilm [39,40]. Agglomeration of CMC particles could reduce the lubricity performance or mending effect ability [16,41]. Although, the CoF acquired from these lubricants is relatively stable. This is because UGE could inhibit the formation of ferrous hydroxide, providing a low corrosion rate [42]. According to Figure 4(c), the pin has less corrosion compared to the pin lubricated by UG0, indicating that UGE protects the pin and disk from corrosion. Previous research shows that a lubricant with good corrosion inhibition could stabilize the CoF value [24].

Meanwhile, in Figure 4(c), deep wear is present in the middle of the wear scar of the pin. This is probably due to the highest UGE fraction that leads to a higher sedimentation rate of the UG2 sample, according to Figure 1(c). CMC's agglomerated particle could not enter the thin film between the pin and the disk. Moreover, the UG2 could not fill the scars or grooves of the rubbing surface, increasing the surface roughness [16].

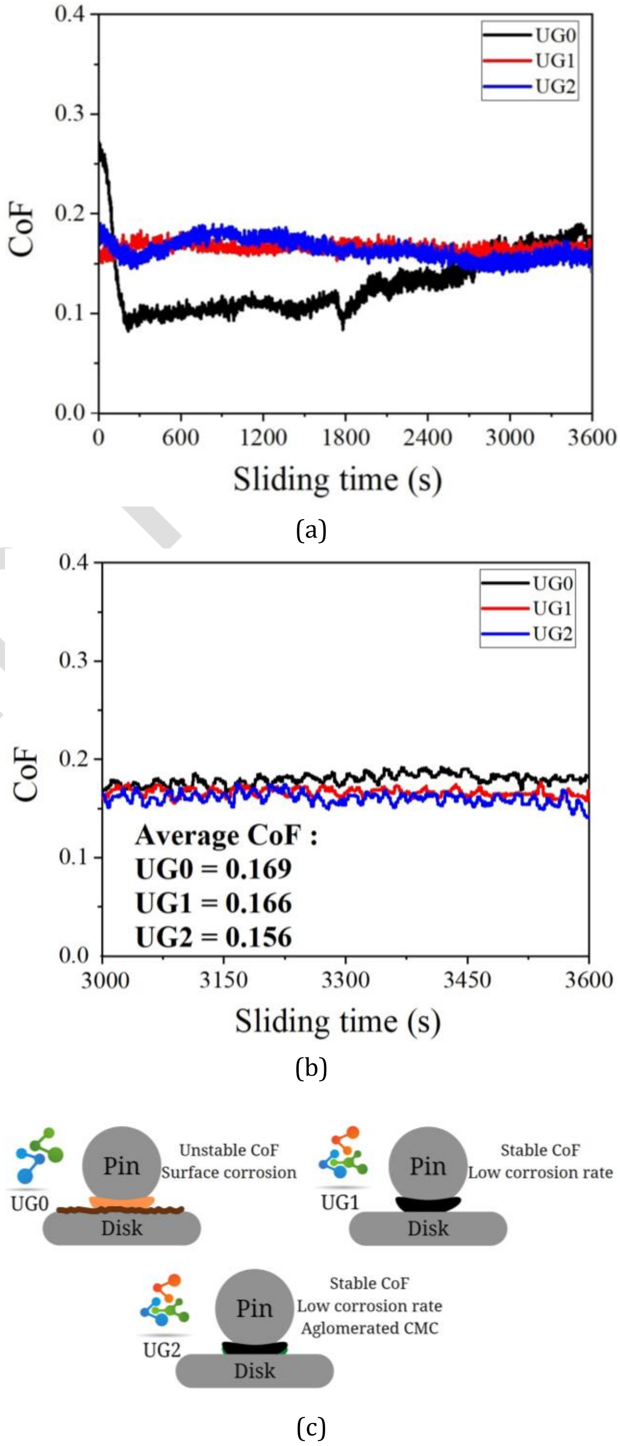
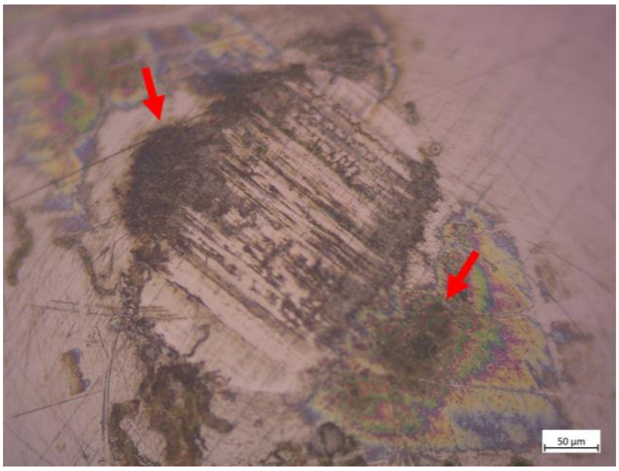
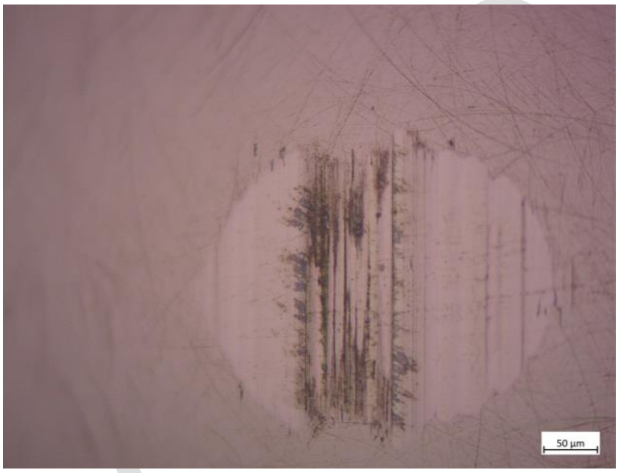


Fig. 3. Friction coefficient vs time for all water-based lubricant samples in full sliding time (a) sliding time of 3000 to 3600s (b), wear-mechanism of UG0, UG1, and UG2 (c).



(a)

(b)



(c)

Fig. 4. Optical microscope image of the pin lubricated by UG0 (a) UG1 (b), and UG2 (c)

4. CONCLUSION

A novel anti-corrosion type of water-based lubricant was prepared and used UGE and CMC as an additive to study its tribological and anti-corrosion properties. UGE and CMC are hydrophilic and can disperse well in water. The bubble test revealed that the prepared UG1 and UG2 samples significantly enhanced anti-corrosion properties and more stable CoF than UG0. The UG1 lubricant sample has the best performance with stable CoF, good stability, and a low corrosion rate. These results suggested that this water-based lubricant could be utilized in machining applications that will improve the lifespan of the cutting tool.

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