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Effect of sonication to the stability properties of carboxymethyl cellulose/uncaria gambir extract water-based lubricant

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ABSTRACT

This study examined the effect of sonication on FTIR and stability at various temperatures in water-based lubricants with a mixture of Carboxymethyl Cellulose (1wt%) and Uncaria Gambir extract (1wt% and 2wt%). The sample was prepared by mixing the two materials into distilled water using a magnetic stirrer and sonicator with time variations of 5 and 10 minutes. Before mixing, the Uncaria Gambir extract solution with water is first centrifuged to remove the dregs in the Uncaria Gambir extract powder. Stability was carried out in an open room (28°C), drying oven (50°C), and refrigerator (5°C). The stability test results showed that the mixture of Carboxymethyl Cellulose and Uncaria Gambir had good stability at all temperatures after sonication for a short duration. The longer sonication duration could fuse the fibrils of Carboxymethyl Cellulose, leading to increasing particle size. FTIR results also show that there is no chemical reaction that occurs. After adding the gambier, there was a new peak at wave 800-1300 cm^{-1}, corresponding to the gambier. The results of this study indicate that the Carboxymethyl Cellulose and Uncaria Gambir solution can be a potential lubricant additive. The Carboxymethyl Cellulose can be a viscosity modifier, while Uncaria Gambir extract for corrosion inhibitor.

Keywords: Carboxymethyl cellulose; Uncaria gambir; Biodegradable lubricant; FTIR; Water-based lubricant

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1. INTRODUCTION

The need for renewable energy is increasing every year [1]. Lubricant is a substance derived from crude oil, which is a conventional resource. For that, we need alternative resources for the sustainability of the earth. Research related to bio-lubricant has been widely studied lately [2]–[5]. Bio lubricants have properties that can match conventional lubricants. Generally, bio-lubricants are prepared using vegetable oil as the base fluid. Furthermore, additives such as nanoparticles and other chemical substances are needed to match their properties with conventional lubricants. The addition of nanoparticles can increase the friction coefficient because these nanoparticles can cover the tiniest cracks in the lubricated material [6]. Vegetable oil based bio-lubricants with nanoparticle additives can reduce the friction coefficient by 15% compared to the base fluid [7]. Furthermore, nanoparticles can increase the lubricating base fluid's heat resistance and thermal conductivity. Adding an advanced nanoparticle, MXene, increased the thermal conductivity up to 1.2 times the thermal conductivity of the base fluid [8].

However, most of the nanoparticles come from metals and can cause corrosion to the lubricated object. In addition, there is a difference in the density of the nanoparticles with the base fluid. This will make the nanoparticles unstable, and sedimentation will occur [9]. On the other hand, besides its advantages, it requires a lot of processing in order to obtain the desired properties of vegetable oil-based lubricants. Not only that, the use of vegetable oil as a lubricant will potentially increase the use of free land due to increased production and increased cooking oil prices [10] . Therefore, some researchers decided to use water instead of vegetable oil. Water has excellent biodegradation properties, does not pollute the environment, and is widely available. Water-based lubricants have been studied before and can be applied as a cutting fluid and hydraulic fluid and are quite efficient compared to lubricating oil. To achieve the properties of a lubricant, it requires additives that can increase the viscosity, which is Carboxymethyl Cellulose (CMC).

On the other hand, water can cause corrosion and can be overcome by adding *Uncaria Gambir* extract. This was already done in our previous work [11]. Adding 2wt% *Uncaria Gambir* extract to water can significantly reduce corrosion compared to water lubricants without *Uncaria gambir* extract. Gambir contains tannins which can prevent the corrosion of metal. Originally used as a medicine, this plant can be processed easily by extracting gambier leaves [12], [13]. In the previous studies, water lubricants with CMC and UG have been investigated for their tribological, corrosion, and stability properties. Stability testing is only carried out at room temperature. In preparation, the samples were sonicated for 30 minutes with 600W power. In this study, FTIR testing was carried out to determine whether a chemical reaction occurred. A stability test in high and low temperatures is conducted. Each sample was sonicated with various durations of 5 minutes and 10 minutes.

2. MATERIAL AND METHODS

2.1 Materials

CMC (99% purity), filtered using an 80 mesh sieve was purchased from Changsu Wealthy Science and Technology, China. UG was supplied by PT. Andalas Sitawa Fitolan, Indonesia. Analytical grade distilled water was purchased from PT. Brataco, Indonesia. Figure 1 shows the CMC and UG used in this work.

(a) (b)

Figure 1: (a) CMC and (b) UG powder

2.2 Sample Preparation

The composition of the samples is shown in Table 1. The CMC powder was stirred into distilled water using a magnetic stirrer (Daihan Scientific MSH-200) at 300 rpm and 50ºC for 30 minutes. After it was soluble, UG was dissolved with distilled water and left for 2 hours before centrifuge. When centrifuging, the residue was removed carefully, and CMC powder was added to the supernate and stirred for another 30 minutes. Sonicator (SJIA-1200W, Ningbo Yinzhou Sjia Lab Equipment, China) with 600W power was employed. The detail, including the UG morphology used in this work, is available in our previous work [14], [15].

Table 1: The composition of water-based lubricant

Samples	Sonication Time (minutes)	Fraction
CUG0-A	5	99wt% Water + 1wt% CMC
CUG1-A	5	98wt% Water + 1wt% CMC + 1wt% UG
CUG2-A	5	97wt% Water + 1wt% CMC + 2wt% UG
CUG0-B	10	99wt% Water + 1wt% CMC
CUG1-B	10	98wt% Water + 1wt% CMC + 1wt% UG
CUG2-B	10	97wt% Water + 1wt% CMC + 2wt% UG

2.2 Dispersion stability via photo capturing

The stability of the samples was investigated by capturing the samples (Canon, SX540 HS), known as the photographic capturing approach [16]. Each sample is stored in a 250 ml bottle and investigated in intervals of 0 and 24 hours. The stability is examined in three conditions: room, high and low temperatures. For the high temperature, the stability investigation is conducted in the drying oven at 50ºC (Memmert, UN-55), while the low temperature is in the showcase refrigerator at 5ºC. In the stationary condition, the settling speed of a CMC particle in a liquid follows the Stokes law [17].

$$V = \frac{2R^2}{9\mu}(\rho_p - \rho_L) \cdot g \tag{1}$$

Where V is the CMC particle's sedimentation speed; R is the CMC particle's spherical radius; μ is the water viscosity; ρ_P and ρ_L are the CMC's particle and the water density, respectively, and g is the acceleration of gravity.

2.3 Chemical stability via FTIR

The FTIR was investigated using a Spectrum Two-UATR (Perkin Elmer, United States) integrated with a detector of MIR TGS. The scanning range of the measurements is in the range of 500–4000 cm^{-1} with a scanning speed of 0.2 cm/s.

3. RESULTS AND DISCUSSION

3.1. Dispersion stability via photo capturing

Dispersion stability is one of the important parameters indicating the stability of a suspension. A suspension is stable when no precipitate or agglomeration occurs. This can happen if the substances in the solution have different densities, viscosity particle sizes, or other physical properties [8]. Literature discloses that a parameter of stable suspension via photo capturing is the sedimentation level. The suspension is unstable if more than 30% of sedimentation or agglomeration is found in the bottom, middle, or top of the vessel [18]. A suspension with unstable stability can be observed in our prior research [3], [8]. This sedimentation or agglomeration could affect the properties of the suspension and allows blockages to occur. Figure 2 shows the lubricant's stability investigation via photo capturing in various temperatures after 24 hours stored at room temperature (2a), 50ºC (2b), and 5ºC (2c). It can be seen that all sample is well-dispersed without any sedimentation or agglomeration found.

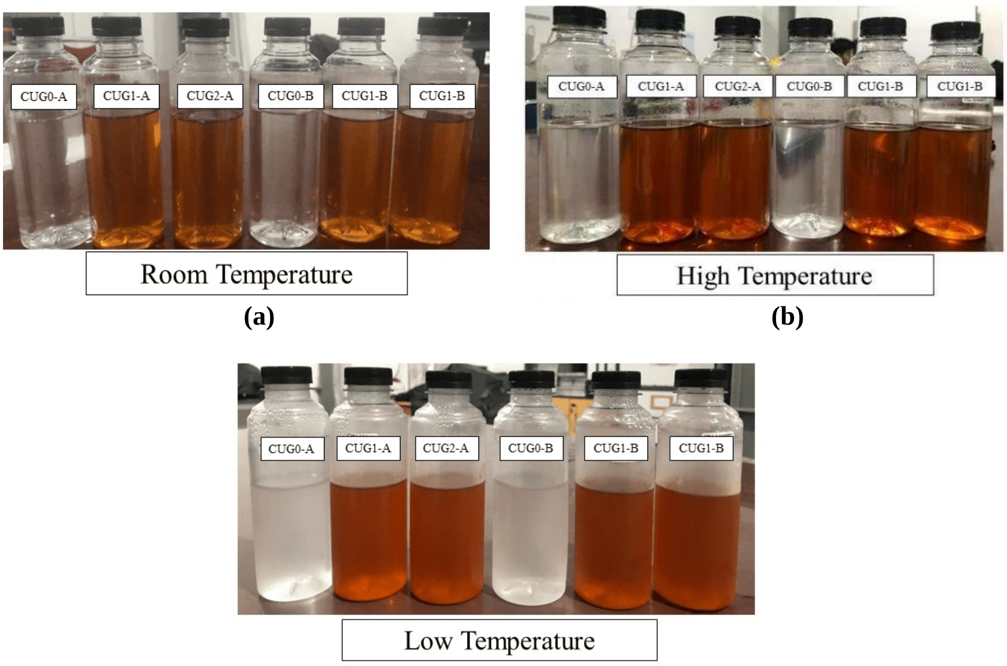

Figure 2: Stability of the samples after 24 hours in (a) Room (b) High, and (c) Low temperature

CMC is hydrophilic in nature with high solubility in water [19]. The addition of UG causes a color change in the solution. After adding UG, the sample remained stable at all temperatures. This is because Gambir is hydrophilic and can dissolve in water [19]. In this previous study, samples were sonicated for 30 minutes, and samples with 2wt% gambier underwent sedimentation [11]. This is probably due to the long ultrasonic duration. It is stated that sonication could increase the crystallinity of cellulose membranes. The increase occurred in the amorphous region and turned into crystalline. However, if conducted for a long duration, the bundled cellulose fibrils could fuse with one another, and the particle size becomes higher [20]. The larger particle size will lead to faster sedimentation speed which, at worse, agglomeration [17]. In that case, finding a suitable duration for sonication is vital to reduce the particle size of CMC and UG [21].

Reducing the duration of sonication has been shown to prevent agglomeration that occurred in previous studies. In Figures 2 (a, b, and c), it can be seen that all of the samples have good stability. This indicates that 5 to 10 minutes of sonication delivers better stability than 30 minutes. The particle size of CMC and UG might be lowered efficiently due to sonication, thus, lowering the sedimentation speed. On the other hand, the sedimentation speed should be faster at low temperatures (Figure 2(c)), but sedimentation is still not visible. This may be due to CMC being dissolved in water, so there is no van der Waals force or attractive force between CMC particles.

3.2. Dispersion stability via FTIR

The dispersion stability of the samples can be analyzed with FTIR by investigating its functional groups of the compound. If one of the groups of UG and CMC are present in the FTIR graph of CUG1 or CUG2, than the suspension is stable. In this test, the 10 minutes sonicated samples is employed. 10 minute sonicated samples is expected to have better results according to the literature [22]. The FTIR spectra of all lubricants are shown in Figure 3. For comparison, an additional sample of UG0 was carried out in this test. This sample is Gambir solution without CMC. It can be seen that all samples show a similar form of the spectrum. O-H stretching vibration in the range of 3100-3500 cm^{-1} corresponds to a characteristic of the CMC. A band of –C=O groups at the peak in the range of 800-1300 cm^{-1} confirms the absence of -C=C- aromatic and C-H, Ar-H corresponds to UG [23]. As expected, for all samples, no significant change is acquired in the peak of 3200 cm^{-1}. This means that the hydrophilicity of CMC and UG is proven. There is no chemical reaction that occurs since there is no new or shifting peak. In the CUG2-B sample, there was a slight change in intensity in the 1200 cm^{-1} wave area, which corresponds to the Gambier group. This result indicates that the sample with the Gambir mixture (CUG1 and CUG2) is well dispersed and chemically stable, which is consistent with Figure 2.

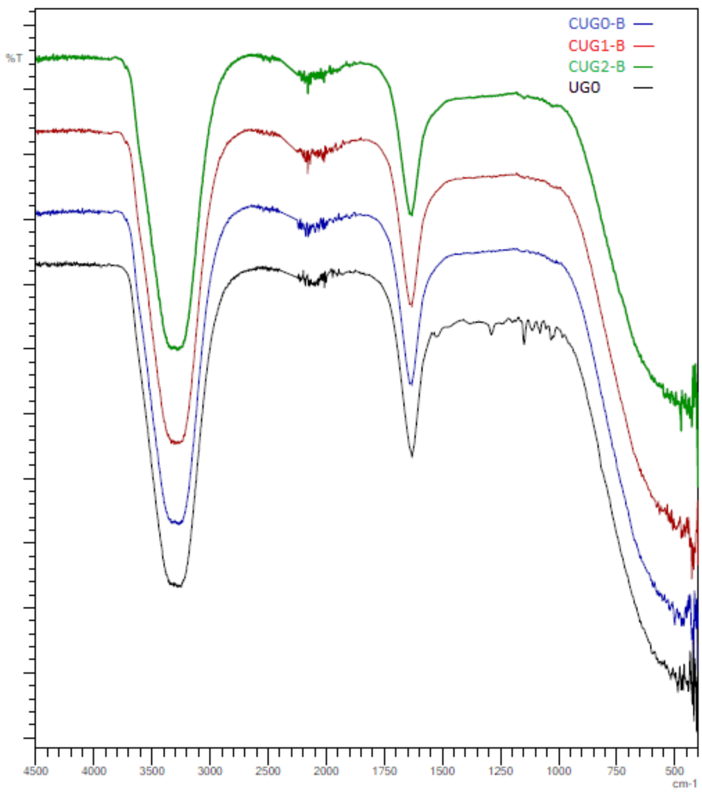

Figure 3: FTIR of the samples sonicated in 10 minutes

4. CONCLUSION

This study investigated the effect of sonication on FTIR and stability at various temperatures in water-based lubricants with a mixture of CMC and UG. Stability testing was carried out at room-high and low temperatures. The stability test results showed that the mixed CMC and UG lubricants had good stability after being sonicated for a short duration. FTIR results also show that there is no chemical reaction that occurs. Photo capturing is the qualitative method in investigating stability performance. In the following work, it is necessary to investigate suspension stability with UV-Vis, Zeta Potential, or DLS method. It is also necessary to investigate the suspension's particle size to ensure the sonication's effect. It is also possible to add the CMC and UG to a different base fluid. Additionally, this compound might be suitable as a grease additive since CMC has a relatively high viscosity. The results of this study indicate that the combination of CMC and UG can be a very potential lubricant additive. It can also be used as an economic coolant in machining applications.

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DECLARATIONS

Author contribution

D. Rahmadiawan: Writing - Original Draft, Writing -Review & Editing, Conceptualization, Formal analysis, Investigation, Resources, Visualization. H. Abral and I.A. Laghari: Conceptualization, Formal analysis, Investigation, Supervision. F. Ilhamsyah and Y. A: Writing - Review & Editing, Resources, Visualization, Supervision

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Competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

REFERENCES

[1] C. D. Rakhmania, S. R. Sari, Y. I. Azhar, A. Sugita, and M. Tominaga, “Cellulose Nanofiber Platform for Electrochemical Sensor Device: Impedance Measurement Characterization and Its Application for Ethanol Gas Sensor,” *Teknomekanik*, vol. 5, no. 1, pp. 57–62, 2022. https://doi.org/10.24036/teknomekanik.v5i1.12872

[2] M. M. Hussain, A. P. Pratap, and V. R. Gaval, “Study of vegetable oil based biolubricants and its hydrodynamic journal bearing application: A review,” *Tribol. Ind.*, vol. 43, no. 4, pp. 511–523, 2021. https://doi.org/10.24874/ti.979.10.20.02

[3] D. Rahmadiawan, H. Abral, N. Nasruddin, and Z. Fuadi, “Stability , Viscosity , and Tribology Properties of Polyol Ester Oil-Based Biolubricant Filled with TEMPO-Oxidized Bacterial Cellulose Nanofiber,” *Int. J. Polym. Sci.*, vol. 2021, no. Natural Fiber-Reinforced Biopolymer Composites, 2021. https://doi.org/10.1155/2021/5536047

[4] M. A. Hossain, M. A. Mohamed Iqbal, N. M. Julkapli, P. San Kong, J. J. Ching, and H. V. Lee, “Development of catalyst complexes for upgrading biomass into ester-based biolubricants for automotive applications: A review,” *RSC Adv.*, vol. 8, no. 10, pp. 5559–5577, 2018. https://doi.org/10.1039/C7RA11824D

[5] M. A. N. Barsari and A. Shirneshan, “An Experimental Study of Friction and Wear Characteristics of Sunflower and Soybean Oil Methyl Ester under the Steady-State Conditions by the Four-Ball Wear Testing Machine,” *J. Tribol.*, vol. 141, no. 4, 2019. https://doi.org/10.1115/1.4042390

[6] B. Zakani, S. Entezami, D. Grecov, H. Salem, and A. Sedaghat, “Effect of ultrasonication on lubrication performance of cellulose nano-crystalline (CNC) suspensions as green lubricants,” *Carbohydr. Polym.*, vol. 282, no. May 2021, p. 119084, 2022. https://doi.org/10.1016/j.carbpol.2021.119084

[7] N. F. Azman, S. Samion, M. A. A. Moen, M. K. Abdul Hamid, and M. N. Musa, “The anti-wear and extreme pressure performance of CuO and graphite nanoparticles as an additive in palm oil,” *Int. J. Struct. Integr.*, vol. 10, no. 5, pp. 714–725, 2019. https://doi.org/10.1108/IJSI-03-2019-0026

[8] D. Rahmadiawan, N. Aslfattahi, N. Nasruddin, R. Saidur, A. Arifutzzaman, and H. A. Mohammed,

"MXene Based Palm Oil Methyl Ester as an Effective Heat Transfer Fluid," *J. Nano Res.*, vol. 68, pp. 17–34, 2021. https://doi.org/10.4028/www.scientific.net/jnanor.68.17

[9] Z. Fuadi, D. Rahmadiawan, R. Kurniawan, F. Mulana, and H. Abral, "Effect of Graphene Nanoplatelets on Tribological Properties of Bacterial Cellulose / Polyolester Oil," vol. 8, no. March, pp. 1–11, 2022, https://doi.org/10.3389/fmech.2022.810847

[10] D. Rahmadiawan, N. Nasruddin, P. Primawati, S. R. Primandari, P. Purwantono, and R. A. Nabawi, "Life Cycle Assessment of Synthetic Polyolester Oil-Based Biolubricant Filled with Bacterial Cellulose Nanofiber," *Int. J. Life Cycle Assess.*, pp. 1–6, 2022.

[11] D. Rahmadiawan, Z. Fuadi, R. Kurniawan, H. Abral, F. Ilhamsyah, and A. Arafat, "Tribological Properties of Aqueous Carboxymethyl Cellulose / Uncaria Gambir Extract as Novel Anti-Corrosion Water-Based Lubricant," *Tribol. Ind.*, 2022. https://doi.org/10.24874/ti.1357.08.22.10

[12] N. Erizon, J. Jasman, S. Syahrul, and W. Wakhinuddin, "Manufacture of Gambir Leaf Press Impac System in Efforts to Increase Productivity of Gambir Farmers," *Teknomekanik*, vol. 2, no. 2, pp. 68–75, 2019. https://doi.org/10.24036/tm.v2i1.3972

[13] J. Jasman, N. Erizon, I. Irzal, and J. Ardi, "Making Print Tool Gambir Sap Parallel Slinder System," *Teknomekanik*, vol. 2, no. 2, pp. 61–67, 2019, https://doi.org/10.24036/tm.v2i1.3872.

[14] H. Abral *et al.*, "Anti-UV, antibacterial, strong, and high thermal resistant polyvinyl alcohol/Uncaria gambir extract biocomposite film," *J. Mater. Res. Technol.*, vol. 17, pp. 2193–2202, 2022. https://doi.org/10.1016/j.jmrt.2022.01.120

[15] H. Abral, A. Kurniawan, D. Rahmadiawan, D. Handayani, E. Sugiarti, and A. N. Muslimin, "Highly antimicrobial and strong cellulose-based biocomposite film prepared with bacterial cellulose powders, Uncaria gambir, and ultrasonication treatment," *Int. J. Biol. Macromol.*, vol. 208, no. March, pp. 88–96, 2022. https://doi.org/10.1016/j.ijbiomac.2022.02.154

[16] M. S. Kumar, V. Vasu, and A. V. Gopal, "Thermal conductivity and rheological studies for Cu–Zn hybrid nanofluids with various basefluids," *J. Taiwan Inst. Chem. Eng.*, vol. 66, pp. 321–327, 2016. https://doi.org/10.1016/j.jtice.2016.05.033

[17] Hiemenz, P. C. and R. Rajagopalan, *Principles of Colloid and Surface Chemistry, revised and expanded*. CRC press, 2016.

[18] L. Samylingam *et al.*, "Thermal analysis of cellulose nanocrystal-ethylene glycol nanofluid coolant," *Int. J. Heat Mass Transf.*, vol. 127, pp. 173–181, 2018. https://doi.org/10.1016/j.ijheatmasstransfer.2018.07.080

[19] D. Rahmadiawan *et al.*, "The Enhanced Moisture Absorption and Tensile Strength of PVA/Uncaria gambir Extract by Boric Acid as a Highly Moisture-Resistant, Anti-UV, and Strong Film for Food Packaging Applications," *J. Compos. Sci.*, vol. 6, no. 11, p. 337, 2022. https://doi.org/10.3390/jcs6110337

[20] P. Paximada, E. A. Dimitrakopoulou, E. Tsouko, A. A. Koutinas, C. Fasseas, and I. G. Mandala, "Structural modification of bacterial cellulose fibrils under ultrasonic irradiation," *Carbohydr. Polym.*, vol. 150, pp. 5–12, 2016. https://doi.org/10.1016/j.carbpol.2016.04.125

[21] H. Abral, V. Lawrensius, D. Handayani, and E. Sugiarti, "Preparation of nano-sized particles from bacterial cellulose using ultrasonication and their characterization," *Carbohydr. Polym.*, vol. 191, pp. 161–167, 2018. https://doi.org/10.1016/j.carbpol.2018.03.026

[22] M. Asrofi, H. Abral, A. Kasim, A. Pratoto, M. Mahardika, and F. Hafizulhaq, "Characterization of the Sonicated Yam Bean Starch Bionanocomposites Reinforced By Nanocellulose Water Hyacinth Fiber (Whf): the Effect of Various Fiber Loading," *J. Eng. Sci. Technol.*, vol. 13, no. 9, pp. 2700–2715, 2018.

[23] G. Yeni, K. Syamsu, O. Suparno, E. Mardliyati, and H. Muchtar, "Repeated extraction process of raw gambiers (Uncaria gambier Robx.) for the catechin production as an antioxidant," *Int. J. Appl. Eng. Res.*, vol. 9, no. 24, pp. 24565–24578, 2014.