The effect of antioxidant additives on the characteristics of food-grade grease using castor oil (*Ricinus communis* L.) as the base oil

E J Mayasari, R Bakri and M Fibria

1 Department of Chemistry, Faculty of Mathematics and Natural Sciences (FMIPA), Universitas Indonesia, Depok 16424, Indonesia

2 Research and Development Centre for Oil and Gas Technology (LEMIGAS), Jalan Ciledug Raya No.109, Jakarta 12230, Indonesia

Corresponding author: juitamayasari@gmail.com

Abstract. Grease has three basic components: base oil, thickening agents, and additives. Castor oil has a potential role as a lubricant base oil in grease, but it has lower oxidative stability than mineral oil. Antioxidant additives can delay oxidation reactions in food-grade grease. Li-12-hydroxystearate soap is used as a thickening agent. The food-grade grease is formulated via saponification, dilution, recrystallization, and homogenization. This research aimed to study the effects of different types (BHT, TBHQ, and high molecular weight phenolic) and concentrations (0%, 0.5%, 1%, 1.5%, and 2%) of antioxidants on food-grade grease characteristics. Several characteristics of greases, such as dropping point, pH, consistency, and corrosion resistance were observed. The results showed that different antioxidants induced differences in dropping points, and different antioxidant concentrations induced differences in dropping points and consistency. The best performance was obtained from food-grade grease formulated with high molecular weight phenolic antioxidant. This had the following characteristics: dropping point 193 °C, pH 7, consistency NLGI 2, and corrosion resistance group 1a.

Keywords: grease, food-grade grease, castor oil, antioxidant additive

1. Introduction

Grease is a semi-fluid form, a compound contains of liquid lubricant, thickening agent and auxiliaries. The liquid lubricant can be in the form of petroleum (mineral), oil derived from plants or synthetic oil [1]. Therefore, grease has three basic components: base oil, thickening agents, and additives [2]. Grease lubricants are regarded as colloidal dispersions of thickening agent contained in liquid lubricant. In environmentally friendly products, the main components of greases must be rapidly biodegradable [3]. Biodegradable greases synthesized from renewable resources such as vegetable oils offer a convenient alternative for petroleum-based greases which known to carry a toxic property toward the environment as well as non-biodegradable. Grease contains of vegetable oil mostly found in semi-solid form of colloidal dispersion from thickener (metal soap) in a liquid lubricant matrix (vegetable oil). Therefore, the current urge in tribology is an experiment to discover renewable, biodegradable, as well as highly thick oil, which meet the requisite of a lubricant [4]. Biological lubricants are particularly applied in food or pharmaceutical industry, namely in an oven or kiln in a bakery, in which the risk of contamination should be diminished. Sources of biological oils are vegetables and animals. Vegetable oils include palm, rapeseed, and castor oils, while sperm and fish oils and lanolin are sourced from animals [5]. Vegetable oils and synthetic ester-based greases give
better performances and are eco-friendly and biodegradable [6]. The neat castor oil is in lower quality compared to mineral oil in terms of its tribological properties, even though an intense review against the experimental data indicated that both are comparable. Commercial mineral oil contained additives and thus holds a better performance than neat castor oil. In this case, a more thorough experiment is to be performed by utilizing a varied of additives with castor oil to obtain a suitable formulation that can substitute mineral oil for various usages and therefore, reduce the leverage toward the environment [7].

Lubrication is essential for ensuring engine performance and efficiency. Therefore, effective and safe lubricants are necessary. Lubricants that are widely used commercially are mineral-based and synthetic oils that are harmful when in contact with food products. Food-grade grease is a lubricant based on vegetable oils that is safe for health as well as environmentally friendly, so it is beneficial to the pharmaceutical, animal feed, cosmetics, and food industries in particular [8]. Standard instrument complies with the requirement of biodegradable grease manufacture; however, the equipment needs to be completely cleaned to ensure that conventional greases and their components will not contaminate [3]. Machinery used for preparing and processing the edible stuffs, make-up products, tobacco products, and pharmaceutical products require all type of lubricants, i.e. grease, oil etc. to lubricate, for heat transfer purpose, as well as transmission of power, and protection against corrosion in a machine, including its part, equipment, and instruments. Sometimes, lubricant and food contact cannot be avoided, which will cause contamination to the food products. One way to overcome this issue is by using a food-grade lubricant [9].

Castor oil is inedible and is used almost entirely for pharmaceutical and industrial applications [10]. Lubricating grease based on castor oil is suitable for use on metal surfaces. Castor oil is not consumed in foodstuffs so would not impact food security [11]. Research performed on crude castor seed oil confirmed the presence of ricinoleic, oleic, palmitic, stearic, and dihydroxystearic acid; this indicated that it could be modified for use as food additives as well as in transportation, cosmetic, and pharmaceutical industries [12]. Castor oil was found to be a rich source of ricinoleic acid, as a major fatty acid, and has significant potential as industrial oil [13].

Castor oil has a potential role as lubricant base oil in grease, but it has lower oxidative stability than mineral oil. According to Mohammed [14], natural antioxidants are among the most important additives used in greases. Selected antioxidants were found to be effective in improving the oxidative stability of coconut oil [15]. Antioxidant additives can delay oxidation reactions in food-grade grease [8].

This research aimed to study the effects of different types (BHT, TBHQ, and high molecular weight phenolic (HMWP)) and concentrations of antioxidants on food-grade grease characteristics.

### 2. Materials and methods
Lithium hydroxide and 12-hydroxystearic acid were used to prepare a lithium 12-hydroxystearate soap lubricating grease (thickening agent) containing 17% soap. BHT (butylated hydroxytoluene), TBHQ (tert-butylhydroquinone), and HMWP were used as antioxidant additives at percentages of 0%, 0.5%, 1%, 1.5%, and 2% by weight of the total formulation. Castor oil and all the components were kindly supplied by PPPTMGB “LÉMIGAS.”

A food-grade grease was formulated by a saponification–dilution–recrystallization–homogenization process. The greases were prepared by mixing lithium 12-hydroxystearate soap, castor oil, and the additive, heating to 80 °C under constant stirring at 400 rpm for 90 min, and then allowing the hot mixture to cool to room temperature. Desirable characteristics of greases relate to their dropping point, pH, consistency, and corrosion resistance, as well as oxidation stability for castor oil, and these properties can be measured. The experimental greases produced were characterized using ASTM (American Society for Testing and Materials) test methods.

### 3. Results and discussion
True grease consists of oil or another fluid lubricant that is mixed with a thickener such as a soap to form a solid. Lithium grease has many useful properties such as load carrying ability, high-temperature tolerance, and shear stability; thus, it is considered valuable and is commonly used in most parts of the world [14]. Table 1 lists the tribological properties of food-grade greases.
Table 1. Characteristics of Food-Grade Greases.

|                      | Dropping Point (°C) | pH | Worked Penetration (0.1 mm) | Consistency NLGI | Corrosion Resistance |
|----------------------|---------------------|----|-----------------------------|------------------|----------------------|
| G-0                  | 191                 | 7  | 271                         | 2                | 1a                   |
| G-BHT0,5             | 189                 | 7  | 290                         | 2                | 1a                   |
| G-BHT1               | 189                 | 7  | 287                         | 2                | 1a                   |
| G-BHT1.5             | 190                 | 7  | 285                         | 2                | 1a                   |
| G-BHT2               | 190                 | 7  | 281                         | 2                | 1a                   |
| G-TBHQ0.5            | 188                 | 7  | 295                         | 2                | 1a                   |
| G-TBHQ1              | 190                 | 7  | 291                         | 2                | 1a                   |
| G-TBHQ1.5            | 190                 | 7  | 281                         | 2                | 1a                   |
| G-TBHQ2              | 192                 | 7  | 275                         | 2                | 1a                   |
| G-HMWP0,5            | 192                 | 7  | 289                         | 2                | 1a                   |
| G-HMWP1              | 193                 | 7  | 285                         | 2                | 1a                   |
| G-HMWP1.5            | 193                 | 7  | 265                         | 2                | 1a                   |
| G-HMWP2              | 194                 | 7  | 258                         | 3                | 1a                   |

3.1. Dropping point
Grease’s heat resistance indicator is represented by the dropping point parameter. When the grease’s temperature rises, the penetration property increases to the point it liquefies and lost its desirable consistency. The dropping point refers to the temperature where the grease is adequately fluid to drip, suggested the upper temperature boundary in which the structure of grease remains, instead of maximum temperature which can be applied [16]; this is usually 20 °C–30 °C below the dropping point [4].

Food-grade grease with added BHT antioxidants has a lower dropping point than conventional grease with and without added TBHQ and HMWP antioxidants. This is because BHT has a lower melting point than TBHQ and HMWP, which have melting points ranging from 125 °C to 129 °C, while that of BHT is only 70 °C.

3.2. pH
The expected pH of food-grade grease is neutral (7). Addition of antioxidants did not affect the pH. All the food-grade greases produced had a pH of 7.

3.3. Consistency
Grease’s physical and chemical properties of are greatly regulated by its consistency or hardness [17]. The thickening agent provides consistency characteristic in grease and keeps the oil properly [1]. Rigidity or consistency is the grease’s most significant characteristic. Effective lubrication depends on the consistency of grease, which in turn relies on the applied thickening agent’s type and quantity as well as the base oil’s viscosity. The consistency of grease reflects in the resistance against deformation by a specified force. Penetration is a parameter for measuring the consistency of grease. Penetration of the grease that works and does not work is measured by ASTM D 217 and D 1403 methods. The Consistency or grade numbers, ranging from 000 to 6 has been established by NLGI related to a stated range of penetration number [16].

The greases with BHT had a lower consistency than the others, while those with HMWP had a higher consistency. Increasing the concentrations of antioxidants tended to increase the consistency of food-grade grease.

3.4. Corrosion resistance
Corrosion in air is caused by oxigen reactions, generally starting by the formation of peroxides and hydroperoxide, followed by the breakdown of fatty acids and the conversion into aldehydes or ketones as well as the formation of free fatty acids (rancidity). Oxidation process can increase the peroxide
Addition of antioxidants did not affect corrosion tests. All the food-grade greases produced showed good resistance to corrosion (group 1a). Group 1a indicates that there is no corrosion.

3.5. Oxidation stability
Oxidation causes breakdown of the chemical and physical properties of materials. For example, samples containing fats and oils can become rancid, exhibiting increased acidity that can lead to corrosion and rusting. Oils can thicken, becoming more viscous, which affects their lubricating properties. Sludge can also form, blocking nozzles and filters, and further compromising lubrication. Lacquering can affect cooling and friction properties, and dyes can undergo discolouration. To slow these processes, antioxidants are added to inhibit oxidation reactions [18]. The vegetable oil’s oxidation stability decreases toward the double bond number, and the poor low-temperature properties is even worse as the number of double bonds decreases [19].

Figure 1 shows the performance of three different antioxidants at various concentrations in a castor oil in the RBOT test at 150 °C. It can be seen that all the antioxidants are effective in inhibiting oxidation, although BHT and HMWP are more effective than TBHQ. BHT is the most effective antioxidant.

4. Conclusions
A food-grade grease, NLGI Grade 2 (depth of penetration 265–295 mm (0.1 mm)), can be manufactured using 17% lithium soap thickener. Increased concentrations of antioxidants tend to increase the dropping point and consistency of food-grade grease. Food-grade grease with added BHT antioxidants has a lower dropping point. HMWP antioxidant additives increased the dropping point. Addition of antioxidants did not affect the pH or corrosion test. All the food-grade greases produced have a neutral pH (7) and good corrosion resistance (group 1a). All the antioxidants effectively inhibit oxidation, but BHT and HMWP are more effective than TBHQ. BHT is the most effective antioxidant. The best performance was shown by food-grade grease formulated with HMWP antioxidant. This food-grade grease had the following characteristics: dropping point 193 °C, pH 7, consistency NLGI 2 (multipurpose grease), and corrosion resistance group 1a.

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