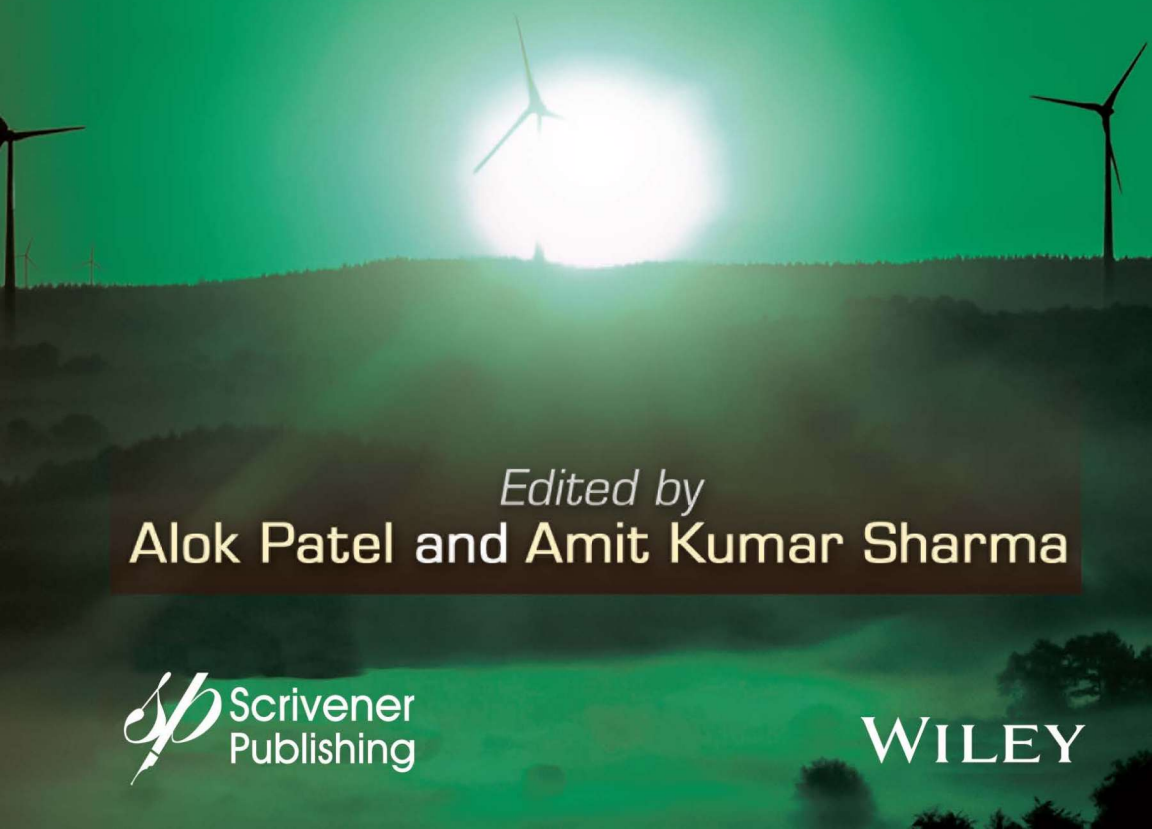




SUSTAINABLE PRODUCTION INNOVATIONS

Bioremediation and Other Biotechnologies



Edited by
Alok Patel and Amit Kumar Sharma

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Biolubricant

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Abstract

This chapter introduces the current situation of the global lubricant market and the potential application of bio-lubricants in future. The sources, properties and applications of vegetable oil and waste edible oil as the lubricant base oil were summarized. In addition, the prospect of bio-refining base oil from biomass was put forward. The upgrading process of vegetable oil, such as hydrogenation, epoxidation, esterification, transesterification and isomerization, were reviewed. In this chapter, some research achievements on the development of environmental-friendly additives through organic conversion of biological materials are reviewed. Finally, the current strategic goals of bio-lubricant are prospected: (1) to reduce pollution caused by lubricating industry, (2) to decrease the safety risk of using or producing dangerous lubricating oil, (3) to improve the utilization rate of vegetable oil raw materials through more effective conversion technologies and reaction routes, and (4) to give priority to using abundant carbon neutral biomass as raw materials.

Keywords: Biolubricant, biodegradable, non-toxic, vegetable oil, biomass, additives, tribological performance

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1.1 Introduction

Statistics show that friction consumes one-third of the world's primary energy, wear causes about 80% of machine parts to fail, and more than 50% of machine equipment accidents are caused by lubrication failure and excessive wear [1]. The resulting waste of resources has become an important issue for destroying the environment. Since the last century, with the significant increase in transportation and various industrial activities, non-renewable petroleum resources have been introduced into industrial activities as a lubricant (Figure 1.1(a)), thereby minimizing the friction between the surfaces in contact with each other. The lubricants ultimately provide smooth running, as well as expand the life of the machine component by reducing the heat generated when the surfaces have relative motion between them [2].

Generally, researchers think that lubricants are only used to make the parts 'slippery', but in real life, lubricants play a very important role because they [3]:

- Reduce the wear of parts with relative motion
- Reduce the friction between rotating and stationary parts

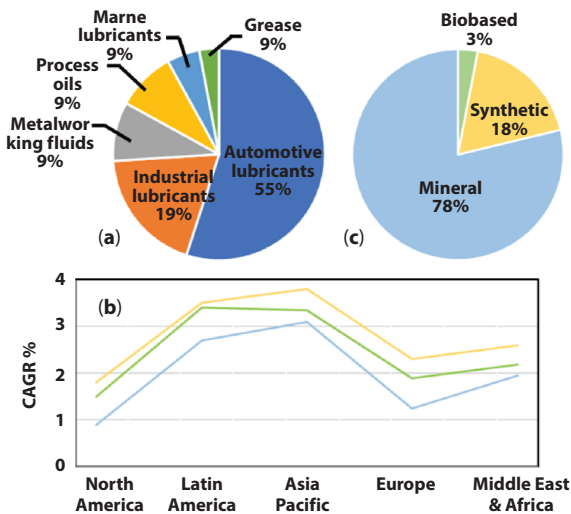


Figure 1.1 (a) Global lubricant market in terms of application; (b) CAGR of global lubricant market in terms of type and region; (c) Global lubricant market in terms of raw materials type [4]. Copyright © 2021, the royal society of chemistry, reprinted with permission.

- Absorb shock
- Minimize operating temperature
- Prevent the corrosion of metal surfaces
- Prevent contamination of the system.

However, about 50% of all lubricants in the world are scattered in the environment due to their volatility, spills, accidents, or application losses. More than 95% of these lubricants are based on mineral oil. These lubricants are now a considerable threat to the environment as well as human health because of their low biodegradability and high eco-toxicity [5].

Depending on the contact time, exposure to mineral oil-based lubricants can cause a series of skin problems, such as itching, skin discoloration, eczema, and skin thickening. In addition, as the exposure time increases, it can also cause respiratory diseases such as bronchitis, bronchopneumonia, asthma, emphysema, and tuberculosis [5]. Besides, the currently used lubricants can pose a carcinogenic risk to humans. Polycyclic aromatic hydrocarbons (PAHs) which have 3-7 rings, such as benz-a-anthracene, benz-a-pyrene, and chrysene, are the main components causing this carcinogenic effect. During the combustion cycle of the engine, toxic chemicals such as PAHs are formed [6].

The mineral oil-based lubricants are very hazardous for plants and animals as well as groundwater bodies because of their low biodegradability [7]. The lubricants that are produced from crude oil are also significantly dangerous for aquatic ecosystems. Water containing as low as one part per million of lubricant is considered to be polluted [8]. Additionally, the lubricant film on water causes a severe obstacle for exchanging oxygen between water and the atmosphere. It also reduces the amount of light that passes through the water. For these reasons, the photosynthesis process of aquatic plants will be reduced and the temperature of the water will be increased by absorbing solar radiation. These limitations cause a critical disruption in the metabolic system of the aquatic organisms [9].

In recent years, more and more attention has been paid to biolubricants. In order to protect the environment from serious damage and reduce economic losses, the use of biolubricants is increasing. Biolubricants are composed of components which are biodegradable and non-toxic to humans as well as other living organisms. Rapid biodegradability indicates that these substances are eco-friendly [10, 11]. Biolubricants can be classified into two main categories based on their chemical composition: natural oils and synthetic oils. The main ingredients of natural oils are vegetable oils or animal fats. On the other hand, researchers use various natural oils as starting substances to form synthetic oils and they show better thermo-oxidative

stability, wear resistance, and lubrication properties than mineral-based lubricants, which can be used as advanced biolubricants [12, 13].

As people's awareness of environmental protection has become stronger, plant-based lubricants have gradually received widespread attention. Plant-based lubricating oil is synthesized by the internal catalytic recombination of plant raw materials, which is similar to light crude oil. Compared with traditional mineral-based lubricants, plant-based lubricants have unique properties such as environmental performance, excellent lubricity, and higher viscosity index and anti-corrosion performance due to their chemical structure [14]. Moreover, they have a higher flash point that makes them suitable for high-temperature uses (over 300°C) [15]. Therefore, the development of bio-lubricants can also alleviate the fossil energy crisis. More importantly, bio-lubricants are biodegradable with little to zero toxic properties. Furthermore, their lower volatility resulted in decreased exhaust emissions [16]. Biolubricants have their own negative side. They have poor oxidative stability, which means the lubricants will oxidize quickly during the application process if untreated and become thick and polymerizing to a plastic-like consistency [16]. The free acid generated due to oxidation may increase corrosion. Chemical modification may lessen this problem but it will increase the cost [17].

It now appears that biolubricants are mainly used in total loss and high-risk industrial applications, such as railway grease and hydraulic oil. According to market research company Kline & Co, the compound annual growth rate of biolubricants is about 5%. From this, it can be seen that bio-based lubricants are showing a steady growth momentum and the market potential is huge.

1.2 Biolubricant Base Oil

Lubricating base oils are mainly divided into three categories: mineral base oils, synthetic base oils, and vegetable oil base oils [8]. A large amount of mineral base oils are widely used. However, in some applications, synthetic base oils from biomass and vegetable base oils must be used for blending products, so these two base oils have developed rapidly.

Base oil accounts for more than 70% of lubricant. The degradability of base oil determines the biodegradability of green lubricants. At present, degradable lubricant base oils mainly include synthetic oils and vegetable oils [8]. Vegetable oil is a component of natural organic long hydrocarbon chains containing polar groups, which make vegetable oil essentially amphiphilic surfactant and can be used as boundary lubricants [14]. Polar

molecules have strong affinity and interaction with metal surfaces and long hydrocarbon chains are oriented away from the metal surface to form a monomolecular layer with excellent boundary lubricating properties. Therefore, the application of vegetable oil and synthetic base oils from biomass to make it a green lubricant base oil has become the main trend of environmental protection lubrication.

1.2.1 Edible and Non-Edible Oils

Originally, vegetable and animal oil were used as lubricants for machinery and transportation [16]. At present, with the depletion of petroleum resources and people's demand for green lubrication, bio-oil is proposed as a lubricant base oil again. However, due to the complex source of bio-oil [8], poor oxidation stability, and low-temperature stability, it is now only used for niche markets such as chainsaws, railway lubricants, and other total loss lubricants. How to maintain the consistency of vegetable oils and meet performance for industrial applications has become the

Table 1.1 Physical properties of vegetable oils [19].

Vegetable oils	Density (kg/m ³)	Kinematic viscosity at 40°C (mm ² /s)	Oxidation stability 110°C, h	Cloud point (°C)	Flash point (°C)
Palm	875	5.72	4	13	165
Sunflower	878	4.45	0.9	3.42	185
Coconut	805	2.75	35.4	0	112
Soybean	885	4.05	2.1	1	176
Linseed	890	3.74	0.2	-3.8	178
Peanut	882	4.92	2.1	5	177
Rape Seed	880	4.45	7.5	-3.3	62
Jatropha	878	4.82	2.3	2.75	136
Karanja	918	4.80	6	9	150
Neem	885	5.2	7.2	14.5	44
Castor	898	15.25	1.2	-13.5	260

key to expanding and applying it more widely [8]. Compared with petroleum-based lubricant base oils, vegetable oils not only have higher flash point, viscosity index, lubricity, and lower evaporation, but also have the advantages of lower loss and good metal adhesion [18]. Table 1.1 summarizes the physical properties of different vegetable oils, mainly in terms of density, kinematic viscosity, oxidation stability, etc. [19], which are the basic requirements for vegetable oils as lubricating base oils. For vegetable oils that do not meet requirements such as the oxidation stability, pour point, and so on, they need to be upgraded through chemical modification or addition of additives. Specific modification methods and additives will be described in the following sections.

Normally, there is some relationship between the structure and performance of vegetable oils. From a molecular point of view, the main component of vegetable oil is triglyceride synthesized from glyceride and three kinds of straight-chain fatty acids, as shown in Figure 1.2. The chain length of fatty acids is usually in the range of C12-C24. These three fatty acid contents differ from raw materials and play an important role in determining the characteristics of the base oil [20]. The two main variables between fatty acids are the number of double bonds and the length of the chain. Generally speaking, a longer chain length will lead to a higher melting point and viscosity and more double bonds will correspond to a lower melting point, which will reduce viscosity and isothermal oxidation stability [21]. It has been found that monounsaturated fatty acids, such as oleic acid and palmitic acid, have a well-balanced low melting point and good isothermal oxidation stability and viscosity [22]. It can be seen from Table 1.2 that the vegetable oils with various ratios of different fatty acids, will lead to

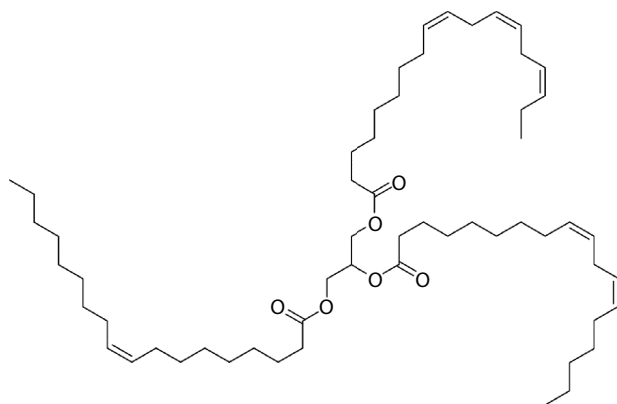


Figure 1.2 Triglyceride chemical structure.

Table 1.2 Fatty acid content of different vegetable oils [23].

	Sunflower %	Soybean %	Rapeseed %	Palm %	Olive %	Castor %	Atropa %
Palmitic Acid	6.18	11.28	4.8	47.9	11.7	2.63	15.6
Stearic Acid	2.16	2.7	1.8	4.23	3	1.51	9.7
Oleic Acid	26.13	24.39	62.7	37	77.9	4.74	40.8
Linoleic Acid	65.52	56.28	19.5	9.07	7.2	8.36	32.1
Linolenic Acid	-	5.34	8.6	0.26	-	-	-
Ricinoic Acid	-	-	-	-	-	82.8	-

changes in the physical properties. Therefore, raw materials with high oleic acid or palmitic acid content are generally favored and sought after.

1.2.2 Waste Cooking Oils

Every year, about 42 million tons of waste cooking oil (WCO) produced globally will be discharged into soil and water sources [17]. More importantly, part of it will be illegally recycled and reused in food processing and shipped all over the world for food consumption. The biggest obstacle for the development of biolubricants is the high cost of raw materials, which accounts for about 70-80% of the total cost. The cost of WCO is only 30-50% of vegetable oil and its structure is similar to vegetable oil. Hence, the WCO could be used as the biolubricant base oil. In addition, the upgrading process of vegetable oil can be used for the waste cooking oil [24].

The main components of WCO are similar to edible vegetable oils which are mainly a mixture of triglycerides and fatty acids [25]. During the frying process, triglyceride is broken to produce free fatty acids and the content of free fatty acids is related to the number of repeated frying. In addition, during the frying process, volatile compounds are produced together with oxygen and these components will be contaminated by food and kitchen

Table 1.3 Physical and chemical properties of waste cooking oils (WCO) [28].

Waste cooking oils	Acid number (mg KOH/g)	Iodine number (mg KOH/g)	Density (g/cm ³)	Viscosity @ 40°C (mm ² /s)	Flash point (°C)	Cloud point (°C)	Pour point (°C)
WCO-1	2.36	13.2	0.92	30.05	269	21	18
WCO-2	1.8	12.69	0.92	33.47	276	15	13
WCO-3	3.64	8.57	0.92	27.42	243	16.5	14
WCO-4	0.67	9.45	0.92	31.77	298	25	18.5

utensils [26]. Table 1.3 summarizes the typical physical and chemical characteristics of WCO collected from different sources [27]. The WCO from various sources has different structure and physical properties, so it needs to be reformed and upgraded to meet the lubrication requirements. The specific modification methods are detailed in the next section.

1.2.3 Microbial Oils

Raw material cost has become a shackle that restricts the development of bio-lubricants [24] and the use of microbial oil as raw material has become a new research idea. Compared with vegetable oil, microorganisms can accumulate higher oil content and the composition of fatty acids is similar to vegetable oil, so we can get some experience from the lubricating mechanism and upgrading process of vegetable oil. At the same time, microorganisms have the advantages of fast reproduction, short growth period, strong environmental adaptability, and small occupied area [29]. However, the problems of high energy consumption in the process of microbial amplification need to be resolved urgently. The current research content of using microbial fermentation to prepare bio-lubricating base oil will be summarized in the following section.

At present, microbial oils are mainly produced by fungi, microalgae, and oleaginous yeast. Microalgae oils are composed of high content of polar oils and non-glycerol oils [30]. The length of fatty acids is between C12-C22 and the number of double bonds is between 0-6 [31]. Unsaturated fatty acids are almost *cis*-heterologous. The oleaginous yeast oil mainly stores oil in the form of triglycerides in liposomes in cells. Yeast fatty acids are mainly composed of oleic acid, stearic acid, palmitic acid, linoleic acid, and hexadecenoic acid [31]. The main fatty acids of yeast oils are very similar to the fatty acid composition of vegetable oils, allowing them to replace vegetable oils to prepare bio-lubricants and other chemicals [32]. For example, Patel *et al.* researched that oil obtained from oil-producing microorganisms can replace squalene and docosahexaenoic acid (DHA) in fish oil and the residual lipid produced as a by-product can be used as a new type of biolubricant in Figure 1.3 [33]. The disc wear volume and ball wear diameter lubricated by PEG 200 and leftover lipids are depicted in Figure 1.4 and these findings suggest the potential of using leftover lipids to replace mineral oils in a number of applications.

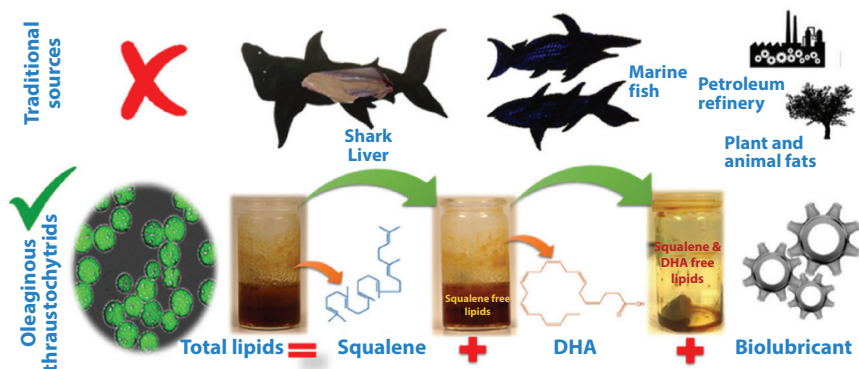


Figure 1.3 Schematic diagram of preparation of lubricating oil from microalgae [33]. Copyright © 2020, American Chemical Society, reprinted with permission.

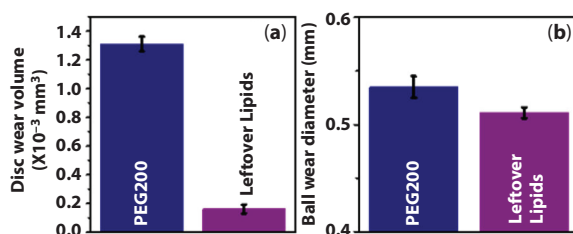


Figure 1.4 Performance Indicators of PEG 200 and Leftover Lipids used as Lubricants: (a) Disc wear volume; (b) Ball wear diameter. load, 2.0 GPa; test duration, 1 h; room temperature [33]. Copyright © 2020, American Chemical Society, reprinted with permission.

1.2.4 Lignocellulose Base Oil

Lignocellulose is the most abundant biomass raw material in the world. It mainly consists of cellulose, hemicellulose, and lignin [34, 35] and consists of a sustainable raw material. Nowadays, fossil fuels are gradually depleting and researchers are more and more performing in-depth research on them. Among them, lignocellulose can be converted to chemicals and fuels through catalytic reaction [35, 36]. Recently, the preparation of high-value lubricating oil through bio-refining has become a research hotspot.

At present, a large number of research focuses on the use of catalytic technology to prepare cellulose lubricants with branched carbon chains. Upgrading a low-carbon biomass matrix to a high-carbon product requires C-C coupling and deoxygenation strategies to obtain a PAO similar structure product. It is achieved through a two-step catalytic reaction

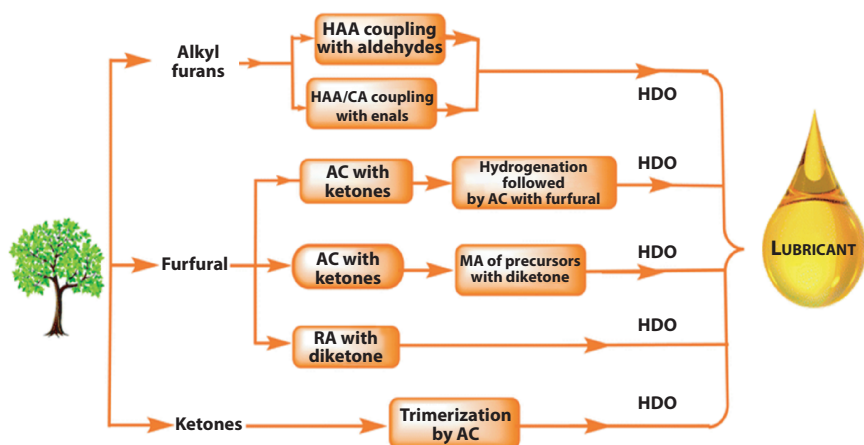


Figure 1.5 Pathways to Produce Lubricating Base Oils (LBOs) from Biomass Feedstock [4]. HAA = Hydroxyalkylation/alkylation, CA = Conjugate addition, AC = Aldol condensation, HDO = Hydrodeoxygenation, MA = Michael addition, RA = Robinson annulation. Copyright ©2021, The Royal Society of Chemistry, reprinted with permission.

process [4], as shown in Figure 1.5. The first step is to prepare high-carbon compounds as the precursor of lubricating base oil by cellulose or hemicellulose derivatives through carbon-carbon bond polycondensation and the second step is to produce deoxygenated lubricating oil in the process of hydrogenation catalytic reaction [19].

Bell *et al.* [37] used a Pd/C and acidic catalysts (silicon-supported 4-ethylbenzenesulfonic acid, EBSA/SiO₂) to synthesize ketones derived from biomass (aldehydes and ketones) through reductive etherification/reductive o-alkylation and to synthesize a series of ether LBOs, as shown in Figure 1.6. From Table 1.4, the physical properties of the synthesized products are similar to commercial oils, indicating that this method could produce a green lubricant based oil.

Lignin has a highly complex aromatic polymer structure. It is a polymer composed of three basic phenylpropane monomers and has abundant

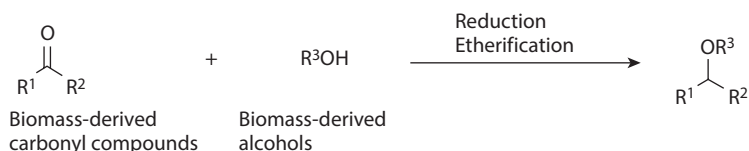


Figure 1.6 Preparation approaches for synthetic lubricants [37]. Copyright © 2017, Wiley, reprinted with permission.

Table 1.4 Properties of (hemi) cellulosic lubricating base oils (LBOs) compared with select commercial analogues [4, 38].

LBOs	KV100 (cSt)	KV40 (cSt)	VI	PP (°C)	Noack volatility/ wt%	DSC oxidation onset T/°C
(2-Heptanone+2-Decyl-1-tetradecanol) Ether	3.1	10	150	-60	6	186
(Butanal+2-Decyl-1-tetradecanol) Ether	3.1	11.1	143	-21	11.9	210
(Isobutanal+2-Decyl-1-tetradecanol) Ether	3.3	12.2	142	-12	11.5	210
(Cyclopentanone+2-Decyl-1-tetradecanol) Ether	3.6	14.2	147	-30	10.8	189
(Octanal+2-Decyl-1-tetradecanol) Ether	3.3	11.9	146	-27	3.9	213
(2-Ethylhexanal+2-Decyl-1-tetradecanol) Ether	3.5	12	145	-36	5.1	206
ExxonMobil PAO4	4.1	19	126	-66	18.8	221
Mineral group II base oil (150N)	5.3	30.6	106	-13	14.5	N/A

oxygen-containing functional groups such as carboxylic acids, phenols, and alcohols [38]. Therefore, some volatile or corrosive fatty acids (such as acetic acid) will be produced during the depolymerization process of lignin, resulting in the instability of bio-oil. Hence, the corrosive problems need to be solved before lubrication application.

Mu *et al.* [39] has converted lignin into bio-oil through depolymerization and esterification reactions with high liquid production rate

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