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REVIEW ARTICLE

BIOLUBRICANT PRODUCTION FROM VEGETABLE OIL THROUGH TRANSESTERIFICATION USING CATALYST: A REVIEW

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ABSTRACT

Depletion of crude oil reservoirs around the world increased the costs of crude oil, and also the issues related to the environment seemed to be an emerging problem with the potential to shape human lives in profound ways. These problems are perhaps in great need of attention from the researchers to find an alternative way to build a sustainable environment. Many current articles focus on vegetable-based lubricants which are eco-friendly, biodegradable, renewable, and sustainable and have gained popularity and are accepted globally. Therefore, vegetable-based lubricant has become a potential alternative to conventional petro-based lubricant. Vegetable-basedlubricants are not widely commercialized due to their inappropriate chemical structure which leads to poor oxidative stability, poor corrosion protection, susceptibility to hydrolytic breakdown, solidification at low temperatures, poor flow, and poor viscosity index which lags them during application in odd conditions. The challenge in this field of study is to improve the above-mentioned properties and characteristics of vegetable oil without degrading their excellent tribological and environmentally applicable properties. The structural problem related to vegetable oil can be overcome by chemical modification to make it fit for the application of lubricant. Parameters like temperature, catalyst concentration, duration, and methanol oil ratio are necessary to obtain a better yield. The final ester obtained depends on the nature of alcohol and the use of a catalyst becomes necessary when the alcohol used is a complex type.

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INTRODUCTION

From time immemorial with the invention of the wheel, the forefathers used edible and non-edible vegetable oil and animal fats for lubrication. Lubrication then has become instinctual for humans with the arrival of man-made machines. It is a substance that helps to reduce friction between surfaces in mutual contact that ultimately reduces the heat generated when the surfaces move thus it protects the surfaces in close proximity and behaves as an antifriction media [1,2]. A good lubricant is capable of keeping the moving surfaces apart, has excellent thermal properties, acts as an inhibitor of corrosion and friction reducer and water repellent, possesses high wear resistance, bears wear debris and other contaminants. The word tribology is used when we are concerned about the reduction of friction and wear. Tribology is derived from the Greek word *Tribo* meaning‶rub″ or friction and *logos* meaning‶related of″or‶the logic of″. Due to global industrialization and modernization, there was a drastic increase in energy consumption which led to the progressive depletion of fuel reservoirs thereby increasing the search and development for alternative chemicals and energy sources that could replace traditional fossil fuels [3]. The lubricants produced from bio-based sources is considered to be a potential alternative to replace petro-based lubricants as it displays excellent physicochemical properties and high biodegradability [4]. It is also used to prevent rust, acts as an insulating agent in transformer applications, sealing agent against dirt, dust, and water, and transmits mechanical power in hydraulic fluid power applications. The reduction of friction and wear is considered as an important function of lubricant [5]. The exhaustive use of Petro-based lubricant has caused hazards to the environment like contamination of air, soil, and water and has become a threat to human health and well-being, welfare of plants, animals and environment as a whole as it is non-biodegradable and non-renewable [6,7]. Due to the alarming environmental pollution contributed by extensive use of petro-based lubricant, the quest for potential alternate, eco-friendly, biodegradable, renewable, sustainable and non-hazardous biolubricant has become an urgent necessity in building a sustainable environment [8,9].

Lubricant

Vegetable oil-based lubricant / Biolubricant: Many researchers have obtained various renewable feedstocks like protein from Maize gluten and rapeseed de-oiled cake for the synthesis of bioplastic sheet, lignocellulosic biomass for the synthesis of bioproducts (chemicals and industrial products), carbonized Jack fruit leaf, peel and leaves of date trees for the synthesis of efficient bio-adsorbents for removal of Nickel and Lead (II) from waste water, Seaweed, sargassumtenerimum, and various seaweed species for the synthesis of bio-stimulant for the growth of plant and its development, vegetable oil for the synthesis of biodiesel, bioconversion of coffee pulp, paper mill sludge, andlignocellulosic biomass, cassava peels, lignocellulosic wheat straw into bioethanol in a sustainable manner, agro-residues biomass for the synthesis of bioethanol and sustainable production of lubricant from vegetable oil-based resources [10-28]. Biomass is considered the main source of energy for many developing countries and it is ranked as the fourth source of energy in the world. Nowadays, these bio-based products are made commercially available in many developed countries which have become an easy approach towards creating a sustainable and renewable environment. When we talk about lubricants, the first thing that comes to our mind is mineral oil lubricants obtained from crude petroleum which is widely used worldwide. It has a wide range of industrial applications. It was reported that 30-40 million tons of lubricant is produced annually and about 50-75% of total lubricant production is poured uncontrollably into the environment [29]. About 95% of total lubricant production is petroleum based. Petro-based lubricant is composed of a mixture of paraffinic (linear/branch), olefinic, naphthenic and aromatic hydrocarbons of 20 to 50 carbon atoms and its formulation is found to be non-biodegradable, non-renewable and toxic to the environment and so has become a threat to the existence of life. Mineral-based oil reduced oxygen exchange as the oil does not dissolve in water and forms a layer on the surface of the water [30,3]. The alarming situation of the environment caused by the excessive use of petro-base lubricant has triggered many researchers and scientists to find an alternative lubricant that is biodegradable, renewable, and sustainable. Vegetable-based lubricants are found to be much more biodegradable and renewable as compared to petroleum-based lubricants [31,32].

A lubricant is considered to be biodegradable when it can be broken down by enzymes through aerobic or anaerobic processes into renewable raw materials [33].Lubricants which is used as potential alternatives to petro-based lubricants must have a high level of biodegradability [34]. The biodegradability of a lubricant takes place through several steps. In the initial process, the original bio lubricant disappears to form another compound that may or may not be completely biodegraded. This degradation is measured by the analysis of the C-H bond through IR spectroscopy [35]. In the second step, the organic compound determines the degradation of the organic compound into CO_2 and H_2O by biodegradation within 28 days [36]. The employing test developed by ASTM and OECD has shown that the percentage of oxygen consumption or carbon dioxide evolution can be monitored when the vegetable oil is inoculated with bacteria and kept under controlled conditions for 28 days. This is done to determine the degree of biodegradability of vegetable oil. Most of the vegetable oils have been shown to biodegrade by more than 70% within 28 days and as compared to petroleum oils which show biodegradability at nearly 15 to 35%. Plant oil lubricants shows a biodegradability percentage (%) of 90-100, plant oil-based esters showa biodegradability percentage (%) of 80-100, polyols and diesters showa biodegradability percentage (%) of 55-100, polyethylene glycol (PGA) shows biodegradability percentage (%) of 10-20, polyalphaolfien (PAO) shows biodegradability percentage (%) of 5-30, and polyether shows biodegradability percentage (%) of 0-25 [26,37]. Many researchers are now focusing their attention on developing technologies that use vegetable oil for lubrication as they are biodegradable, non-toxic, renewable, and consume less energy for production, and no waste is produced. Some advantages of lubricant as compared to petro-based oil [3,7,37-41] are as follows:

- Environmentally – safe, renewable, and sustainable
- Less toxic, less dermatological problems both to humans and animals
- Better efficient lubricating properties lead to lower friction losses and better boundary lubrication
- Higher viscosity index, higher flash point.
- Lesser production of vapour and oil mist
- Longer durability of equipment
- Higher boiling points which lead to fewer emissions
- Higher shear stability, and higher safety on the shop floor
- Easy maintenance, disposal, and storage
- It does not produce a greenhouse effect and does not contain sulphur and aromatic compounds
- Higher solvency for additives and lower volatility
- Lower volatility, better mental adherence

However, there are some disadvantages of vegetable-based oil due to undesirable physical properties viz. poor thermal stability due to the presence of saturated fatty acids, poor oxidation stability due to the presence of mono and poly-unsaturated fatty acids, hydrolytic instability, and poor viscosity index. These shortcomings can be overcome by chemical modification. Therefore, research has been carried out to improve its physical properties so that it can be used as an alternative to petro-based lubricants [28]. Factors like climatic conditions and geographical location seem to affect the ability of the plant to produce lubricants. For example, Asia uses palm and coconut oils to make lubricants. Soybean oils are mostly used by the US to make biolubricant and Europe depends on rapeseed and sunflower oils [42]. India has a great prospectof producing edible and non-edible vegetable oils which remain untapped. Coconut, Olive, Rapeseed, Palm, Soybean, Sunflower, Peanut, Corn, Linseed, etc. are considered edible whereas Jatropha, waste cooking oil, Karanja, Rice bran, Mahua, Castor, Neem, Madhucaindica (Mahua), etc. are considered as non-edible [43-45]. Rapeseed, Soybean, cottonseed, palm, peanut, safflower, sunflower, coconut, tallow, and also waste cooking oil are good raw materials for the production of lubricantsas theycontributeto a sustainable environment [46]. Among all the vegetable oil Canola oil is found to be more viscous than the other commonly tested oils. Its flow rate is lower than diesel at the same pressure and it dropped to almost zero at -4˚C. Castor, Palm, and Coconut oils are found to be potential alternative lubricants to two-stroke engine oils. 5% volatile organic compounds emissions to the environment can be reduced with the use of castor oil as a biolubricant [47]. Different vegetable oils employed for various industrial applications and health benefits are tabulated below in Table 1. According to the report, Soybean oil displayed better power-transmitting properties than mineral oils [48]. Several vegetable oils such as rapeseed oil, palm oil, Moringa oil, Passion fruit oil, and Rubber seed oil displayed excellent properties when used as hydraulic oils [49].

Lubrication properties of Vegetable oil: The triglyceride present in vegetable oil provides many desirable qualities for lubrication. Most of vegetable oils are considered to be amphiphilic due to the presence of polar groups, allowing them to adhere to metal surfaces and possess good lubricityand nonpolar groups in the same molecule [4,58]. The polar group contains at least one ester functional group and the nonpolar group is are hydrocarbon chain with degrees of unsaturation. The accurate chemical composition of these two groups is responsible for the tribological and other properties of vegetable oils. For example, better oxidative stability is exhibited by triglycerides having a lower degree of unsaturation [65]. The strong affinity of the base oil's high polarity with lubricated surfaces enables plant-based oil to function as highly effective boundary lubricants [66]. It also has low volatile organic compound emission (VOC) [41]. The fatty acid that is present in biolubricant prevents corrosion and wear between the rubbing zone surfaces by forming a layers and stable film [34]. Viscosity is the vital characteristic of a biolubricant as it prevents contact between the two surfaces [46]. The untreated oils and fats (high viscosity) cause operational problems (deposits on its interior parts) which lead to certain disadvantages thathave to be taken care such as poor thermal and oxidative stability, poor corrosion protection, and very susceptibility to hydrolytic breakdown due to the presence of ester functionality [67,68]. It also undergoes solidification at low temperatures, cloudiness, precipitation, poor flow (low-temperature property), and low viscosity index [26,28,32]. Poor thermal and oxidative stability is due to the high degree of multiple C=C unsaturation in the fatty acid from oleic, linoleic, and linolenic acid moieties of vegetable oilsand it is also an active site for many reactions including oxidation [59,69-70]. The greater the level of unsaturation, the more susceptible the oils undergo oxidation [71]. The unstable nature of plant-based oil is influenced by the free acid content in vegetable oil [27]. Different methods have been acquired to improve the properties of vegetable oils like blending in which two or more oils are mixed to achieve the required properties, fractionation for separating the oil into two or more fractions depending upon the end-use requirement, domestication of wild crops to convert the wild crop into crops that can be cultivated commercially, genetic modification of crops to get desired properties, and chemical modification in which various reactions like hydrogenation, esterification/transesterification of fatty acid with different alcohol moiety to improve the properties of vegetable oil [42]. A lot of research is being carried out to enhance the physicochemical properties of vegetable oil. Some of the reported physicochemical properties of vegetable oils with their respective lubricants are shown in Table 2. Till date, research indicates that vegetableoil modified both chemically and genetically are found to exhibit an outstanding potential to be used as a lubricant [28]. Severalvegetable-based lubricants have been developed for various sectors of industry.

***Chemical structure of vegetable oil*:** Triacylglycerides (TAG, 98%) are the major components of vegetable oils and animal fats [26]. The other minor components are diacylglycerol (DAGs, 0.5%), monoacylglycerol (MAGs, 0.2%), Fatty acids (FAs, 0.1%), sterols (0.3%), tocopherols (0.1%) [72]. Schematic representations of major and minor components of vegetable oils are shown in Fig. 1. During processing, these minor components are removed from biolubricant. The physical and chemical properties of vegetable oil result from the constituent fatty acids since fatty acids make larger portions of triacylglycerols [73]. Triglycerides are glycerol molecules that have three long-chain polar fatty acids attached at the hydroxyl groups through ester linkages and theycontribute to the physical characteristics of the plant-based oil [27,74]. TAG structure provides desirable qualities in lubricant [69]. Vegetable oil with more double chains shows better performance in pour point and low oxidative stability as compared to vegetable oil with fewer double chains. The composition of vegetable oil varies depending on the kind of oil, the pre-harvest condition, and the type of soil and climate [55].The low volatility of plant-based oil is due to the high molecular weight of the triacylglycerol molecule. Vegetable oil properties are directly proportional to the fatty acid composition of triacylglycerol which is closely related to their source [75]. Tri-saturated fatty acids in plants are found mostly in tropical species like palm oil and lauric fats like coconut and palm kernel fats. They are not suitable for lubrication due to their high melting point.

Common properties of Vegetable oil

Fatty Acids: The fatty acids chain length usually ranges from C_{12} to C_{22} and accounts for 85% of the plant oils' weight which determines their properties. The ratio and position of carbon-carbon double bond in vegetable oil is determined by its fatty acid composition. The fatty acid which has a carbon-carbon single bond in its backbone structure is called saturated fatty acids. Stearic acid, palmitic acid, myristic acid, lauric, and butyric acid are examples of saturated fatty acids and the structure is shown in Fig. 2. Palmitic acid (C16:0) and stearic acid (C18:0) are considered as the most important saturated fatty acids. They show resistance to oxidation and exhibit high pour point. Saturated fatty acids exhibit high melting points and are the most chemically stable due to the molecule's conformation. Therefore, the fatty acid's melting point increases as the number of carbon in the chain increases [76]. The fatty acids which have one carbon-carbon double bond in their backbone structure are called mono-unsaturated fatty acids. Oleic acid is considered a mono-unsaturated fatty acid and its structure representation is shown in Fig. 3. Examples of mono-unsaturated fatty acids are Erucic acid, Oleic acid, and Palmitoleic acid. They possessed reactive sites for chemical modification and exhibited extremely low pour points. Mono-unsaturated fatty acids in the oil are responsible for the resistance to rancidity and are liquid at room temperature. However, they are less stable to oxidation than saturated fatty acids [77]. Polyunsaturated fatty acids have more than one carbon-carbon double bond in their backbone structure. Linoleic and linolenic acids are considered poly-unsaturated fatty acids and their structure is represented in Fig. 4. Oleic acid (C18:1), Linoleic acid (C18:2), Linolenic acid (C18:3) are considered the most important unsaturated fatty acids [37]. The higher the number of unsaturated, the greater the biological and oxidative instability of the poly-unsaturated fatty acids [78]. The fatty acids which have a hydroxyl group (-OH) in their backbone structure are called hydroxyl fatty acids. Ricinoleic acid is considered a hydroxyl fatty acid as it possesses one hydroxyl group and its structural representation is shown in Fig. 5.

The hydroxyl group in the fatty acids is responsible for low pour point and reactive sites for chemical modification [79]. Table 3 shows the fatty acids composition in commonly used vegetable oil. It also affects the properties of bio-based lubricants in terms of thermal-oxidative stability, viscosity and viscosity index, and low-temperature behavior. Fatty acid becomes oilier and less soluble in water with the increase in the length of the carbon chain. Short non-branched fatty acids with 6 carbon atoms possess better solubility properties in water due to the presence of polar-COOH group [80]. The plant oils' properties and fatty acids composition of triacylglycerol are closely related to their source [75]. Cottonseed oil is classified as a poly-unsaturated oil as it consists of palmitic acid (20-25%), stearic acid (2-7%), oleic acid (18-30%) and linoleic acid (40-55%) [81]. It is used as a salad oil, frying, margarine manufacture, and manufacturing shortenings for cakes and biscuits. Palm oil, olive oil, cottonseed oil, peanut oil, and sunflower oil are considered unsaturated fatty acids as they contain a high proportion of mono-unsaturated oleic acid and poly-unsaturated linoleic acid [82]. They are liquid at room temperature and the melting point is low. Other vegetable oils fall under various classes such as erucic acid oil, oleic acid oil, linoleic acid oil, and lauric acid oil due to their dominant fatty acids content in vegetable oil. They are used in salad dressing, margarine, shortenings, etc. Soybean oil is classified as linolenic acid oil since it highly contains unsaturated linolenic acid. It is considered an important oil as it has many applicants in the current situation. Castor oil contains triacylglycerols of ricinoleic acid [83]. Coconut oil is classified as saturated fatty acids (92%), particularly lauric acid and is solid at room temperature and has a sharp melting point, unlike other fats and oils. The saturated and unsaturated fatty acid compositions (%) of vegetable oils are given in Table 4. The physicochemical properties of vegetable oils are dependent on fatty acid distribution. A number of double bonds and their position within the aliphatic chain affects the properties of the oil. Biolubricant performance can be enhanced by the removal of double bond and the glycerol molecules from triacylglycerides [37].

Viscosity: The viscosity of a lubricant is a very important property for hydrodynamic lubrication and it's a vital parameter to be considered. A lubricant with a high viscosity requires a large force between two moving surfaces and a lubricant with low viscosity damages the devices [97]. Most of the biolubricants display high viscosity and the viscosity increases with the length of the hydrocarbon chain of the carboxylic acid or alcohol in ester biolubricants [98]. Viscosity is also directly related to temperature. According to the report, it was said that biolubricant with high viscosity indexes show fewer viscosity modifications with the increase in temperature. Biolubricant branching also affects the viscosity index since increased branching in the alcohol or the carboxylic acid lowersthe viscosity index [59].

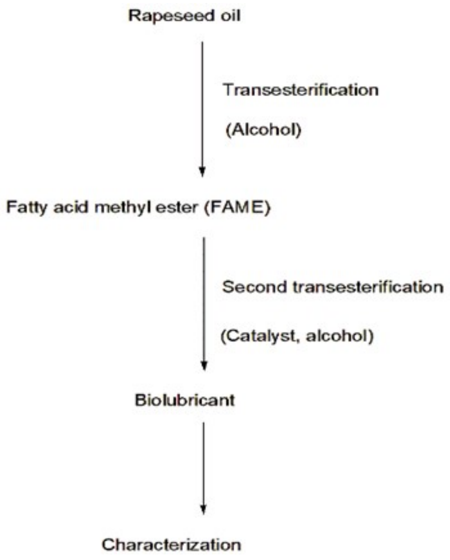
Pour point: The pour point is the temperature below which the liquid loses its flow characteristics. In biolubricant, the pour point and viscosity index are directly related to one another. According to the report, ternary alcohols such as trimethylolpropane (TMP) decrease the pour point of the biolubricant. The use of neopentyl polyols which is a branched alcohols, has become a potential alternative to obtain biolubricant with low pour points and higher oxidative stability levels [45,55]. The pour point of the oil is decreased by the presence of -C=C- bonds, however, it has become vulnerable to the oxidation process. The optimum pour point is obtained from saturated fatty acids with short hydrocarbon chains because an increase in the length of the carbon chain causes a higher pour point. In plants, the carbon chains generally contain between 16 and 18 atoms indicating that the saturation of these acids causes them to become solid at around 65-75°C [37]. The pour point temperature of the oil is not affected by the position of the -C=C- unsaturated bonds much but its conformation slightly influences the pour point temperature. The hydrocarbon chain with cis-configuration, with hydrogen atoms on the same side is observed to have a lower pour point than the hydrocarbon chain with trans-configuration [59].

Low oxidative stability of vegetable oil: A lubricant exhibiting strong oxidation resistance minimizes the occurrence of deposits, sludge, and corrosive by-products in applications such as grease, engine oil, and industrial oil [99]. The double bond (-C=C-) present in the fatty acids of vegetable oil is responsible for the low oxidation stability of the oil. The higher the composition of the unsaturated fatty acids, the more susceptible the vegetable oil towards the oxidation process. The low oxidative stability of vegetable oil can be improved by chemically modifying the vegetable oil by saturating the (-C=C-) bonds through epoxidation [100]. The presence of saturated fatty acids contributes to high oxidation stability. Vegetable oil that undergoes the process of oxidation increases the viscosity of the oil and produces a varnish and sludge deposit which degrades the quality of the vegetable oil. The oxidative stability increases with an increase in the number of substituents at mid and end-chain ester and decreases the number of hydroxyl groups. Oxidative instability can also be improved by transesterification using the catalyst and methyl ester [101].

Poor low-temperature properties of vegetable oil: When the vegetable oil is subjected to low-temperature conditions (24˚C), it undergoes solidification. The solidification does not occur rapidly at a particular temperature but occurs over a wide range of temperatures. The poor low-temperature properties of vegetable oil can cause cloudiness, precipitation, poor flow ability, increased viscosity, and rheological behavior [102]. Many researchers think that the composition of saturated fatty acids in vegetable oil greatly influences the poor low-temperature performance of the oil because at low temperatures the saturated fatty acids chains of carbon atoms tend to bundle rapidly than the unsaturated fatty acids turning into a crystalline form. The poor point (PP) and cloud point temperatures can identify the low-temperature properties of vegetable oil [26,45]. The fatty acid chain with a large branching group at the mid and end points creates a barrier called a stearic barrier inhibits crystallization and results in a lowering of pour point and cloud point temperature. It was reported that thermo-oxidative stability can be improved through chemical modification of vegetable oil, thereby allowing their use in a wider range of operating conditions. A lot of research is being carried out to improve the physicochemical properties of vegetable oil so that it may become an alternative to petro-based lubricants. The excellent wear/friction characteristics and broad temperature range stability can be achieved through chemical modification of triacylglycerides [103]. The properties of vegetable oils can be improved chemically by altering their structure [104]. Additives are also used to enhance the properties of lubricants [105]. Various routes have been developed for the chemical modification of vegetable oils which can be used as a perfect lubricant shown in Table 5.

Production of biolubricant

Transesterification: Many chemical reactions are involved in the production of biolubricant. Among the various reactions, transesterification (from triglycerides to fatty acid complex ester via fatty acid methyl esters) is considered one of the most important reactions in obtaining biolubricant. Table 6. shows the synthesis of biolubricant from various vegetable oil through transesterification. Transesterification is a process in which the triglycerides react with alcohol (methanol, ethanol, etc) in the presence of a catalyst to produce fatty acids alkyl estersand this process lowers the viscosity of the oil or fat [122,123]. The process can be catalyzed by acids, bases, and enzymes [124]. This process is a sequential reaction in which triglycerides are first reduced to diglycerides which in turn are reduced to monoglycerides then monoglycerides are further reduced to fatty acids ester (Glycerol) [125]. In each successive step of the reaction, a mole of ester is released. The general equation for transesterification of a triglycerideis shown in Scheme 1. The melting point and boiling point of fatty acids, methyl ester, mono-, di-, and triglycerides exhibit an upward trend with the growth in carbon chain length but decrease with an escalation in the number of double bonds. The ascending order of melting points among tri-, di-, and monoglycerides is attributed to the polarity of the molecules and the presence of hydrogen bonding [126]. Heterogenous transesterification is found to be more advantageous than homogenousas the catalyst can be reused again, also the removal of the catalyst from the reaction product is not required and there is no formation of saponification products [127,128]. The reaction can be termed as an inter-esterification when there is an exchange of acyl between two molecule and intra-esterification when there is an exchange of acyl group within a molecule [129]. The conversion of acyl group that is bounded in acylglycerols to methyl esters can be determined by using gas-liquid chromatography (GLC) on packed columns [130]. The exchanged of acyl moiety either between an ester and acid is called acidolysis, ester and alcohol is called alcoholysis, two esters is called acyl exchange [131]. The biodiesel produced are referred as FAME since methanol is the most widely used alcohol in transesterification [132]. The steps involved in the production of biolubricant are as follows:



In the first transesterification, FAME was found to be obtained from vegetable oil as an intermediate product. When one mole of triglycerides of vegetable oil reacts with three moles of methanol, it produces three moles of FAME and one mole of glycerol [133]. The use of excess alcohol increases the yield of the alkyl ester and allows its phase separation from the glycerol. After the completion of transesterification, water can be added to the reaction mixture for the easy separation of glycerol [134]. The use of a catalyst becomes necessary when a complex alcohol is used in the reaction. The reaction temperature, methanol/oil molar ratio, the amount and the type of catalyst, and the mixing intensity are the important variables during the reaction as theyinfluence the reaction. The FAME obtained is found to be suitable for the second transesterification [135]. Although it has been claimed that the use of an additional organic solvent was useful in controlling the activity of water and microbial contaminations, the absence of solvent enhances higher substrate and product concentrations, improves safety, etc. The reaction can be classified as an acid-catalyzed or based-catalyzed depending on the type of catalyst used [136,137]. As reported by several researchers in the second transesterification reaction, alcohols with different structuresare employed to obtain different viscosity values andalso mentioned thatthe selection of an appropriate catalyst is necessary for the effectiveness of the reaction [2]. The equation for the second transesterification of FAME to bio lubricant is shown in Scheme 2. Even the nature of the alcohol used during the reaction characterizes the complexity of the final ester obtained**.** During this reaction, the methanol released contributes to the better yield of biolubricant.

Table 1 Distinct applications of several vegetable oil for industrial applications and health benefits[3,7,42,47,50-64]

Vegetable oil	Health benefits	Industrial application	Ref.
Castor oil	Used for medicinal purpose, fertilizers, used for the manufacture of bullet-proof glasses, contact lenses, lipsticks, metal soaps, polyurethanes, etc	Gear lubricants, greases, special engine and high rotation reactors lubricants, high resistance plastics	[7, 47]
Soybean oil	Production of traditional foods (soymilk, soy sauce, soy paste, etc.), ingredients for food, phytic acid, tocopherols, lectins present in soybean oil acts as antioxidant, anticarcinogenic, antioxidant, Dietary fiber present in soybean oil acts as anti-hypertensive, improves digestive tract function, prevents colon cancer,etc.	Biodiesel fuel, lubricants, metal casting/working, pesticides, printing inks, plasticizers,paints, coatings, soaps, shampoos, detergents, disinfectants, hydraulic oil, biodiesel	[50,51]
Palmarosa oil (green oil)	Food processing, cosmetics, etc.	Biodiesel fuel	[52]
Canola oil	Used for the treatment of cardiovascular disease, cancer, obesity, etc.	Metal working fluids, hydraulic oils, penetrating oils, tractor transmission fluids, food grade lubes, chain bar lubes	[3, 53]
Eucalyptus oils	Sources of essential oils, etc.	Production of wood or pulp for the paper industry, etc.	[54]
Rapeseed oil	It has textural properties of food, used in the treatment of cardiovascular diseases, obesity, hypertension, etc.	Air compressor-farm equipment,chain saw bar lubricants, biodegradable greases, raw material for biodiesel, base for engine oil, hydraulic fluids and inks, Lamp oil, Soap making, Plastic manufacturing etc.	[53,55]
Coconut oil	Use for edible purpose, medicine (anti-cancer), food industries as a confectionery fat in ice-cream.It act as an antiviral, antifungal and antibacterial, main component of infant milk powders due to its easy digestibility, etc.	Gas engine oils, have many industrial uses in the pharmaceuticals, cosmetics, plastics, rubber substitutes, synthetic resins etc. Coconut oil has also been found useful for mixing with diesel. Methyl esters of coconut oil fatty acids is also being used as lubricants and biodiesel in aviation industry, etc.	[56-57]
Olive oil	Prevent platelet aggregation, possess anti-inflammatory, anti-tumor and anti-microbial properties, lowers immunological parameters, lipid and glucose metabolism, prevent oxidative stress, control blood pressure, reduce the risk of neurodegenerative diseases, etc.	Automotive lubricants/engine oil	[58-59]
Palm oil	Food and medicine purpose,	Rolling lubricant, grease, high performance base fluids for biodegradable lubricants, used in soap industry.	[42,61]
Safflower oil	It is used to treat neuropathy, chicken box sores, numbness and tingling, boosting skin health, contro; muscle contractions, helped in weight loss, improve hair growth, boast immune system.	Light-coloured paints, diesel fuel, resins, enamels	[42, 61]
Sunflower oil	Used as edible oil, it act as antioxidant during storage and minimizes the auto-oxidation of fatty acids due to the presence of alpha tocopherols, etc.	Grease, diesel fuel substitutes (biodiesel), lubricants, plasticizers, stabilizers	[42]
Cuphea oil	Foliar spray	Cosmetics, motor oil	[42, 62]
Tallow oil	Used in the manufacturing of antibiotics and pharmaceuticals	Steam cylinder oils, soaps, cosmetics, lubricants, plastics	[42, 63]
Groundnut oil	Edible purpose, antioxidant due to the presence of tocopherol,	Food industry applications	[64]

Table 2. Physicochemical properties of vegetable oils with their respective lubricants[3,88,106-114]

Lubricant	Viscosity 40ºC (cSt)	Viscosity 100ºC (cSt)	Viscosity Index	Pour point (ºC)	Flash point (ºC)	Ref.
Olive oil	39.62	8.24	190	-3	318	[3]
Rapeseed oil	45.60	10.07	216	-12	240	[88]
Jatropha oil	35.4	7.9	205	-6	186	[106]
Jatropha/TMP	43.9	8.71	180	-6	325	[107]
Soybean/alcohols	10.3-432.7	3.0-34.4	45-195	-	-	
Olive/PE	63.08	12.00	190	-24	-	
Palm oil	52.4	10.2	186	-5	-	[108]
Palm/TMP	47.1	9.0	176	-2	355	[109]
Rapeseed/alcohols	7.8-38.2	2.7-8.4	205-224	-31.3 to -18	-	[110]
Soybean oil	28.86	7.55	246	-9	325	[111]
Sunflower oil	40.05	8.65	206	-12	252	
Sunflower/octanol	7.93	2.74	226	-3	-	[112]
Castor oil	220.6	19.72	220	-27	250	[113]
Castor/TMP	20.96	4.47	127	-	-	[114]

Table 3 Typical fatty acid composition (%) of various vegetable oil[3, 26, 27, 41, 68, 81, 84-91]

Vegetable oil	Fatty Acids					Ref.
	Palmitic (C16:0)	Stearic (C18:0)	Oleic (C18:1)	Linoleic (C18:2)	Linolenic (C18:3)	
Neem oil	18	18	45	18-20	0.5	[3]
Sunflower oil	7	5	20-25	63-68	0.2	[26]
Linseed oil	5	3	22	17	52	[27]
Moringa oil	5.50	5.70	73.20	1.00	-	[41]
Tobacco oil	3.57	3.95	11.92	73.43	0.83	[68]
Cottonseed oil	20-25	2.7	18.30	40-55	0.6	[81]
Soybean oil	10.50	3.80	23.70	54.50	6.30	[84]
Coconut oil	8.40	2.6	6.4	1.60	0.1	
Corn oil	12.10	2.30	30.90	53.30	1.10	
Canola oil	4.35	2.00	59.40	21.15	10.35	
Palm oil	41.5	2.70	40.6	11.90	0.30	[85]
Canola oil	3	3	60	30	7	[86]
Jatropha curcas L oil	15.6	9.7	40.8	32.1	-	[87]
Rapeseed oil	9.80	1.60	18.4	16.8	6.50	[88]
Castor oil	-	2-3	3-5	3-5	80-90	[89]
Olive oil	7.30	2.70	60.70	4.40	0.50	[90]
Madhucaindica oil	17.8	14.0	46.3	17.9	-	[91]

Biolubricant through this process and their characteristics are shown in Table 7. Thus the use of excess alcohol leads to low oil conversion [129]. Overall, the influence of the temperature, catalyst concentration, and alcohols was considered for the optimization of biolubrication [46].

Base-catalyzed transesterification: In transesterification reaction, a homogeneous base catalyst is found to be more efficient with a higher reaction rate and less corrosion than the acid catalyst [138,139]. However, the catalyst must be substantially anhydrous because the presence of water leads to saponification forming soap and hydrolyse

Table 4. Saturated and unsaturated fatty acid compositions (%) present in vegetable oil[41, 55, 59, 68, 84, 92-96]

Vegetable oil	Compositions of fatty acids (%)		Ref.
	Unsaturated	Saturated	
Calabash	78.13	20.6	[41]
Moringa oil	79.7	5.70	
Sunflower	87.3	12.3	[55]
Jatropha curcas	91.7	7.0	
Neem	64.5	36	[59]
Tobacco seed	86.18	13.52	[68]
Corn	85.42	14.57	[84]
Soybean	84.75	15.18	
Canola	92.75	7.03	
Coconut	8.1	92.78	
Palm kernel	16.9	83	
Olive	82.45	17.53	
Rapeseed	97.6	2.4	[92]
Corn	98.09	1.89	[93]
Palm	39.45	60.55	[94]
Castor oil	96.4	0.9	[95]
Cotton seed	76	25	[96]

Table 5: VariousChemical modifications of vegetable oil, its advantages and disadvantages [59]

Chemical reaction applied for modification	Advantages	Disadvantages
Esterification/transesterification	▪ Enhances resistance to thermo-oxidative degradation ▪ Performs at low temperatures	Needs raw material rich in oleic acid and elevated reaction temperatures
Estolide formation	▪ Enhances resistance to thermos-oxidative degradation ▪ Operates at low reaction temperatures ▪ Accommodates various vegetable oils	Elevated manufacturing expenses
Epoxidation	▪ Enhances slipperiness ▪ Boosts resistance to heat-induced oxidation ▪ Operates at a reduced reaction temperature	Raises pour point while reducing viscosity index
Selective hydrogenation	▪ Decreases the level of double bonds ▪ Enhances resistance to oxidation	Isomerization reactions involving cis- and trans-acids exhibit increased reactivity at elevated temperatures

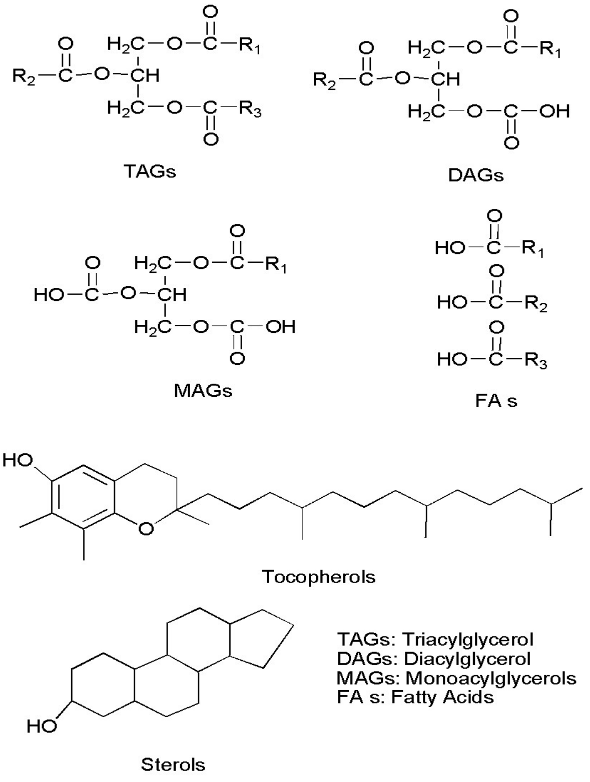


Fig. 1 Schematic representations of major and minor components of vegetable oils

Table 6. Synthesis and chemical modification of biolubricant from various vegetable oil through transesterification[60, 93,115-121]

Oil/Fatty acid methyl ester/ Fatty acid	Alcohol used	Oil/Fatty acid :alcohol ratio	Catalyst used	Reaction condition	Yield (%)	Ref.
Palm oil methyl ester	Trimethylol propane	3.9:1	$NaOCH_3$	20mbar, T=120°C, >1h	98	[60]
Oleic acid	Hexadecanol	6.25:7.5	Sulfated zirconia	140°C, 4h, 300 rpm	81.7	[93]
Rapeseed oil methyl ester	Trimethylol propane	17.1:5.3	$NaOCH_3$	110ºC,8h,reduced pressure 3.3kPa	99	[115]
Palm oil methyl ester	Trimethylol propane	10:1	$NaOCH_3$	110°C, 1-1.5 mbar	98	[116]
Jathropha oil	Trimethylol propane	4:1	H_2SO_4 (2%)	150°C, 3h	98.6	[117]
Jathropha oil	Ethylene glycol	3.5:1	$NaOCH_3$	120°C, 2.5h	-	[118]
Sunflower oil	n-Propanol	1:15	Heteropoly-acidssupported by Clay (K-10)	170ºC, 8h	72	[119]
Sunflower oil	n- Octanol	1:15	Heteropoly-acids supported by Clay (K-10)	170°C, 8h	78	
Karanja oil methyl ester	Hexanol	1:1	$NaOCH_3$(3%)	Under vacuum boiling temp	94.5	[120]
Karanja oil methyl ester	Octanol	1:1	$NaOCH_3$ (3%)	Under vacuum boiling temp	93.1	
Karanja oil methyl ester	Neo-pentyl glycol	1:0.5	$NaOCH_3$ (3%)	Under vacuum boiling temp	95	
Linoleic acid	1-Octanol	6.25:7.5	Sulfated zirconia	140°C, 4h, 300 rpm	84.6	[121]
Stearic acid	1-Octanol	6.25:7.5	Sulfated zirconia	140°C, 4h, 300rpm	93.9	
Oleic acid	1-Octanol	6.25:7.5	Sulfated zirconia	140°C, 4h, 300rpm	98.6	

Table 7. Bio-lubricant synthesized through esterification/transesterification process, their reaction conditions and characteristics [9]

Resultant Bio-lubricant	Reactants used	Reaction conditions	Oxidative/ thermal stability	Yield (%)
TMP triesters	Palm ME, TMP and SodiumMethoxide catalysts	140°C, 25 mbar, 25min, oscillatory flow reactor at 1.5 Hz with 20 mm amplitude	355°C Degradation temp.	94.6
TMP triesters	Jatropha, TMP and SodiumMethoxide catalysts	150°C, 10mbar, 3 h	-	>80
TMP triester	Canola biodiesel ME, TMP andSodium methoxide catalysts	110°C,1 mbar, 5 h	Induction time: 0.74 h	90.9
TMP triester	Castor biodiesel, TMP andDibutyltindilaurate catalysts	170°C, 0.01 bar	RPVOT: 43 min (Butylated hydroxytoluene added)	89.7
n-alcohol-esters	Soybean oil, various alcohols andSulfatedzirconia catalysts	140°C, 4 h	-	>80
FA-n-octyl esters	Sunflower oil, octanol and Fe-Zn doublemetal cyanide (DMC) complexes catalysts	170°C, 8 h	23 min (RBOT)	98
TMP triester	Castor biodiesel, TMP and Sodium methoxide catalysts	120°C, 0.01 bar	RPVOT: 150 min (Butylated hydroxytoluene added)	-
TMP triesters	High oleic palm ME, TMP andSodium methoxide catalysts	120-150°C, 0,3 mbar, 45 min	-	-
TMP triesters	Jatropha ME, TMP and Sodium methoxide catalysts	150°C, 55 min	325°C Degradation temp.	-

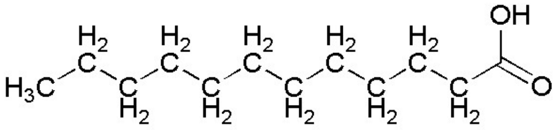


Fig. 2 Schematic representations of a saturated fatty acid

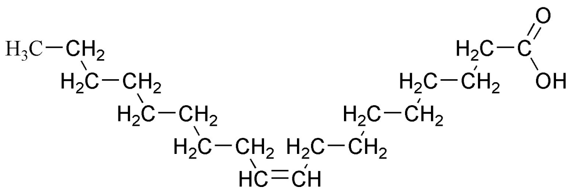


Fig. 3 Schematic representations of a mono-unsaturated fatty acid

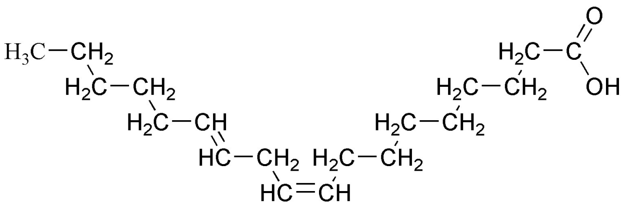


Fig. 4 Schematic representations of a poly-unsaturated fatty acid

ester to form free fatty acid (FFA). Subsequently, the base catalyst will be irreversibly neutralized by FFA and alkaline salt will be formed. Studies have shown that high ester yield can be procured through longer reaction time. Usually, the based-catalysed transesterification reaction requires less than 1 hour to complete the reaction. Further studies have shown that the acid value increases with reaction time when NaOH is used as a catalyst for the methanolysis of soyabean oil [140]. It is also found that esters are formed as an anionic intermediate in the presence of a base catalyst which can dissociate back to the original ester or form a new ester. The widely used basic catalysts are alkaline metal alkoxides, hydroxides, and sodium or potassium carbonates [28]. Alkaline metal alkoxides do not produce water during the reaction so are better catalystsas as compared to hydroxides, and are the most active catalysts with >98% yield within 30 minutes [140]. When water is produced during the reaction, hydrolysis occurs and forms FFA thus increasing the acid value. $NaOCH_3$ when used as a catalyst produces less acid value compared to NaOH [141]. However, alkaline metal alkoxides are not commonly used in a large scale production due to their toxicity, disposal problem and high price [142]. Sodium and potassium hydroxides, carbonates and alkoxides such as methoxide, ethoxide, propoxide and butoxide are generally used alkalis. Sodium hydroxide (NaOH) or Potassium hydroxide (KOH)is used as basic catalysts with methanol or ethanol. Al_2O_3 is also a commonly used catalyst due to its availability and low cost [143]. NanocrystallizedCaO catalyst is also considered as an efficient catalyst with low toxicity, highly available, easy in preparation, and high surface area associated with the small crystallite sizes and defects [144-146]. The increasing order of basic strength of the oxides and hydroxides of group 2 is Mg < Ca <Sr< Ba. Ca-derived bases are found to be the most promising ones as it exhibit low methanol solubility and the least toxicity [147].

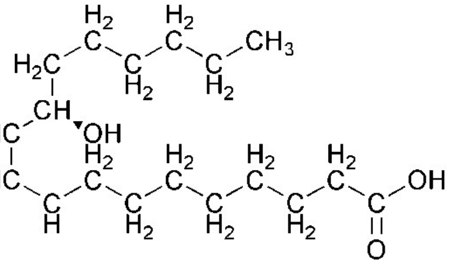
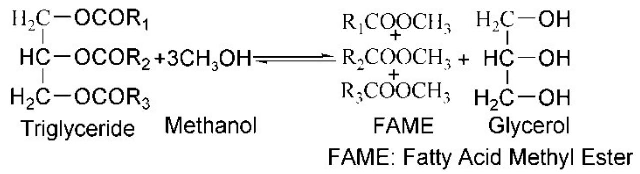
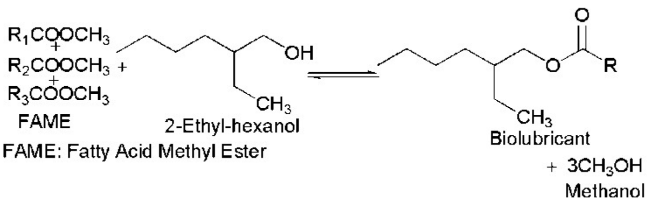


Fig. 5 Schematic representations of a hydroxy fatty acid



Scheme 1: General equation for first transesterification of a triglyceride



Scheme 2. Equation for second transesterification of FAME to biolubricant

Acid-catalyzed transesterification: The acid-catalyzed transesterification approach is deemed less significant in industrial applications compared to the base-catalyzed method because of its sluggish reaction rate. It is reported by several researchers that basic catalysts become inappropriate when working with triglycerides containing high levels of free fatty acids (FFA) as they tend to react with these acids to form soap [148]. Also, acid-catalyzed reactions require high alcohol-to-oil molar ratio, reaction time, and temperature as compared to base catalysis [153]. The commonly used acid catalysts are sulphonicacid, sulphuric acid, hydrochloric acid, and phosphoric acid. Among these acid catalysts, sulphuric acid is the most widely used catalyst as it leads to high yield [141,149-152]. However, it is also reported by some researchers that the reaction rate is very slow with this acid catalyst as well as it requires a high temperature above 100°C to complete the reaction.

Industrial biocatalysts: Lipase has been used as an industrial biocatalyst with good yield. It is also used for esterification and transesterification reactions and has been broadly investigated with high yield [115,154-155]. It is found that the immobilized residue can be regenerated and reused as well as the product extraction is easier using this catalyst [156]. Therefore it is a feasible method for the production of alkyl esters from animal fat or vegetable oil [157]. The transesterification reaction processed by acids, bases, alkoxides, and alkaline metal hydroxide catalysts has a major drawbacks or disadvantages like the problem of emulsification and corrosion are related to acid/base processes. Also, acid-catalyzedreaction proceeds slower than the based-catalyzed reaction, high-quality feedstock with less acid content is necessaryforthe base-catalyzed reaction to prevent the formation of unnecessary saponification of the free fatty acids, the incomplete solubility of oils and fats in alcohol results in the barrier of triglycerides conversion. So, in order to overcome this drawback the use of an ionic liquid-catalyzed transesterification reaction was developed [158-160].

Green catalyst: The ionic liquid is now considered a green catalyst due to its low volatility, ability to stay in liquid form in a wide range of temperatures, and less toxicity. It is also thermally stable, recyclable, and has variousstructures [161]. It is also known as room temperature ionic liquid because of its ability to stay in a liquid state below 100ºC [162]. Therefore, it is considered as a promising catalyst for esterification and transesterification processes [67].

CONCLUSION

The literature reviewed revealed that with increasing environmental pollution and depletion of petroleum resources,vegetable-oil-based lubricant has become a potential alternative to conventional petro-based lubricant. It is found that vegetable oil is suitable oil for deriving biolubricants using appropriate catalyst/s along with some chemical modifications as they exhibit good lubricity, high viscosity index, higher inflammability, better wear performance, increased equipment service life, higher flash point, and lower pour point as compared to other lubricants, and a great degree of biodegrability. The main advantages of biolubricant are its biodegradability and low aquatic toxicity. The environmental compatibility of vegetable oil provides them with an advantage over conventional mineral oils in terms of overall operating cost. The biodegradability of biolubricant is the strongest point in the case of automobile applications. However, the commercial mineral lubricantshave not yet been completely replaced by bio-based lubricants due to the lower production of vegetable oil which led to an increase in price. So, researchers are still at the initial stage with the aim of replacing petro-based lubricant with biolubricant to create a sustainable environment.

Abbreviation

ASTM	American Society for Testing and Materials
DAGs	Diacylglycerol
DMC	Doublemetal Cyanide
FAs	Fatty Acids
FAME	Fatty Acid Methyl Ester
FFA	Free Fatty Acid
GLC	Gas-Liquid Chromatography
IR	Infrared
MAGs	Monoacylglycerol
OECD	Organisation for Economic Co-operation and Development

PAO	Polyalphaolfien
PE	Polyethylene
PGA	Polyalkylene Glycol
PP	Poor Point
RBOT	Rotating Bomb Oxidation Test
RPVOT	Rotating Pressure Vessel Oxidation **Test**
TAG	Triacylglycerides
TMP	Trimethylolpropane
VOC	Volatile Organic Compound

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