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Synthesis of Bio-lubricants from Waste Cooking Oil: A Review

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Abstract: The Market of Lubricants is largely dominated by Mineral Oils based feedstock which causes environmental degradation. This provides a necessity to search for alternatives of Biodegradable nature. Waste Cooking Oil with certain chemical modifications can be utilised for Lubricating purposes. In this regard, the primary contribution of this work lies in the summarisation of the manufacturing process of bio based Lubricants from Waste cooking oil. The steps include pre-treatment of used oil to remove certain undesirable compounds formed during frying, followed by chemical modification by subjecting to Transesterification, Oligomerization, or Epoxidation reactions to enhance the Thermo-Oxidative properties. This study shall help to identify the need of further research in the area for implementation of the strategy at commercial level.

Keywords: Waste cooking oil, Vegetable Oil, Bio-Lubricants, Eco-friendly, Chemical Modifications

I. INTRODUCTION

Lubricants work towards reduction in wear between two surfaces using a substance that can carry load in between of the two relatively moving surfaces. The action of Lubrication not only reduces wear, but also helps in corrosion resistance, in oxidative resistance, and in insulation [1]. The most common base stock for the production of lubricating oils is mineral oils, which are obtained from the petroleum sources. These mineral oils are obtained during refining of crude oil and hence they are readily available and are economically cheaper [2]. However, the concerned problems with petroleum based feedstock are environmental pollution and depletion of petroleum reserves. This has led to a search for an alternative for the lubrication purpose. Bio based lubricants, or simply called Biolubricants, have been found to be an effective substitute as these use vegetable oils as the feedstock, which are renewable resources [3].

Biolubricants are environmentally friendly, non-toxic to humans and other living things, possesses high biodegradability, and have excellent lubrication performance at a minimal waste disposal [4]. These have a neutral CO balance and readily decompose in nature due to presence of esters, which are found to be degraded aerobically as well as anaerobically [5].

While Vegetable oils are a greener feedstock for lubricant production, they bring along some difficulties. The main challenge in utilizing Vegetable oils as a potential feedstock for Bio-Lubricants production, apart from their low oxidative and thermal stability, is their availability. Many countries, such as India, are already scarce in Vegetable Oil and import from foreign countries the major quantity of vegetable oil in order to fulfil the demand needs. This makes the scientific community look at further alternatives.

To cater the needs of the future and lessen the burden on the environment, optimisation of the resources by employing waste for the production of utilities is of major interest for many scientists [6, 7]. Waste Cooking Oil stands out amongst the waste based materials which can be used for the synthesis of multiple products [8]. Waste Cooking Oils are generated in the process of frying food with edible oil and cannot be utilized again without refining because of toxic components produced during frying operation [9]. Because frying food is frequently used everywhere around the globe in large quantities, and because of growth in population worldwide, Waste cooking oils are generated in large quantities on a daily basis [10]. The global waste cooking oil production is estimated between 20% and 32% of total consumption of vegetable oil with the main producers being hospitality and household sectors [11]. These used cooking oils can be utilized for the production of Bio based lubricants which can not only reduce the burden on mineral oil based lubricants but also provides a safe method for recycling of waste cooking oil [12]. This review focuses on the methods of synthesis of biolubricants from waste cooking oil.

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II. BASIC PROPERTIES OF WASTE COOKING OIL

During the frying operation, major physical as well as chemical changes take place in the vegetable oil. These changes are the results of three different types of reactions, which are oxidative, hydrolytic, and thermolytic reactions, and the extent of these changes vary from oil to oil [13]. Due to these reactions, the properties of the vegetable oil get altered and results in a different set of properties for waste cooking oil. Table I describes some of the properties of waste cooking oil as available in various literatures.

TABLE II BASIC PROPERTIES OF WASTE COOKING OIL [14-20]

Acid value (mg KOH/g)	Density (g/cm3)	Viscosity (mm2/s)	Water content (wt. %)	Saponification value (mg KOH/g)	Iodine number (g I2/100g)	Peroxide value (meq/kg)	Reference
3.6	0.91	4.2	1.9	207	83	23.1	[14]
5.8	0.92	40.2	0.43	-	-	-	[15]
3	0.897	49.84	-	-	102.3	-	[16]
5.3	0.937	190.2	1.1	204.3	104.3	5.6	[17]
1.32	0.924	36.4	0.42	188.2	141.5	-	[18]
5.06	0.923	39.6	-	-	-	-	[19]
15.4	0.913	-	0.18	-	-	14.5	[20]

The typical changes in oil during frying include increase in acid value, viscosity, saponification value, peroxide value, and water content. The oil becomes off odor and dark in color. Some of these properties are undesirable and need to be taken care of before utilizing the waste cooking oil for further reactions for use as biolubricants. Table IIII summarises the characteristics of waste cooking oil that are desirable and undesirable for the purpose of use in lubrication.

TABLE IVI CHARACTERISTICS OF WASTE COOKING OIL

Desired Characteristics	Undesired Characteristics
Highly Viscous	High Acid Value
High Viscosity Index	Dark Color
High Lubricity	High Oxidized and Polymerized Fatty Acid
Low Volatility	Some Undesirable compounds
Good Biodegradability	High Water Content
High Flash Point	Solid Impurities Present
More Economical than Fresh Vegetable Oil	Poor Thermal Stability
	Poor Oxidative Stability
	Poor Low Temperature Properties

Each of these undesirable characteristics can be treated by either physical or chemical methods. The solid impurities or suspended solids can be removed by filtration of the waste cooking oil. The filtration can be achieved through polymeric or metallic membranes, conventional cellulose fibres, or activate filtration methods [7]. The repeated water washing method called Water degumming helps in removal of water soluble salt impurities from the crude material. The volatile compounds and water can be removed by distillation of waste cooking oil or by suitable absorption or adsorption techniques [21]. The dark color of the oils can be improved by bleaching. The high acid value is the result of high free fatty acids content. These acids, if left untreated, affect the reactions having alkali catalysts, by forming soap instead of the desirable products and thus the neutralization of these fatty acids must be done beforehand. The poor thermal, oxidative, and low temperature properties of the waste cooking oil can be enhanced by chemical modification methods, resulting in products which can be used as Biolubricants.

III.CHEMICAL MODIFICATIONS

In a triglyceride molecule, there are two positions which can be subjected to modification- at the carboxyl group around the Ester Moiety or β-hydrogen position, and at the fatty acid chain around the olefin group present in the chain. A typical structure of triglyceride is presented in figure 1.

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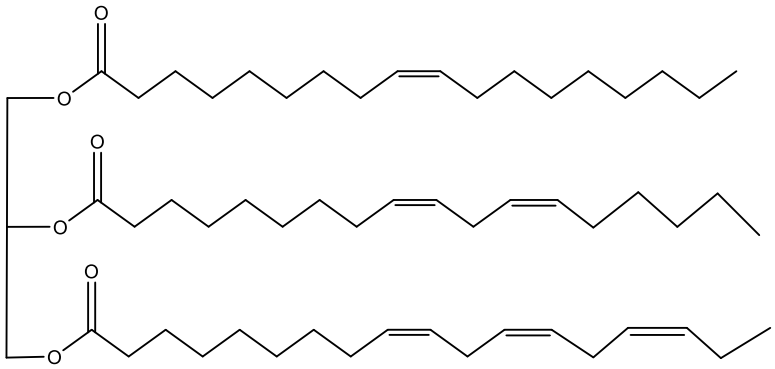

Fig. 1 A Typical Triglyceride Molecule

For carrying out the modification, various methods have been studied and employed over past decades. Out of these, the most prominent are: the rearrangement of the acyl group through esterification or transesterification reaction, the modification of the acyl moiety by forming oligomers of the fatty acids called as estolides, and the modification of unsaturation by epoxidation reaction[22]. These three modification methods are discussed in the following section.

A. *Transesterification*

Transesterification is the reaction of triglyceride, present in fats or oils, with an alcohol to form esters of fatty acid (commonly known as Fatty Acid Methyl Esters or FAME) and yielding glycerol as by product [21]. The reaction scheme of Transesterification is presented in figure 2.

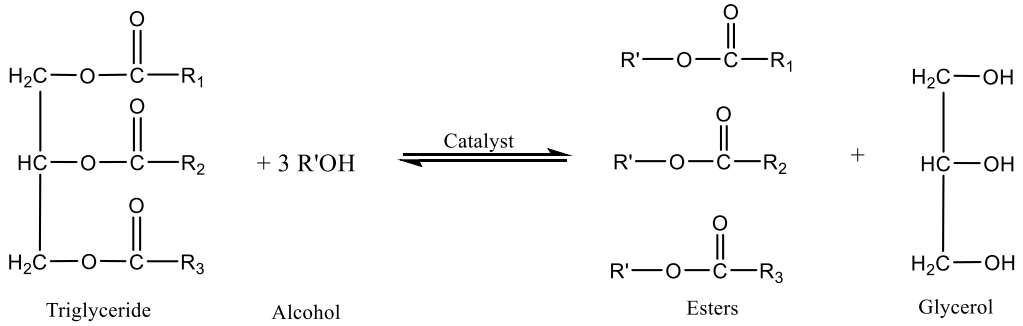

Fig. 2 Reaction Scheme of Transesterification Reaction

Stoichiometrically, one mole of triglyceride reacts with three moles of alcohol to produce three moles of Fatty Acid Methyl Esters and one mole of glycerol; however, in practice, alcohol is taken in excess to shift the reaction towards completion. This reaction is carried at atmospheric pressure and at a temperature of around 50–75 °C which is within the boiling point of methanol [23]. The reaction is generally accomplished by an acid, alkali, or enzyme catalyst which increases the speed of the reaction. The catalyst can be homogeneous and/or heterogeneous in nature [24]. The most widely used homogenous acid catalyst includes sulphuric acid and hydrochloric acid, and homogenous base catalyst includes hydroxides and alkoxides of sodium and potassium [25].

Due to the high free fatty acid content in Waste cooking oil, homogeneous alkaline catalysis causes soap formation. If a homogeneous acid catalyst is used, the problem of soap formation gets eliminated but reaction is much slower and causes other problems such as corrosion of equipment. If only homogeneous catalysts are to be employed, a two-step esterification process can be carried which first uses acid catalysts to produce fatty acid methyl esters and then uses alkaline catalysts to finish the reaction [26].

Homogeneous Catalysts display high reaction activity and are low in cost, but have lengthy and difficult recovery processes from the product due to their high solubility. In order to mitigate the problem, heterogeneous catalysts can be used which make product recovery easier [27]. Heterogeneous catalysis, such as Calcium oxide (CaO), has several benefits which include eco friendly disposal, recyclability, and ease in separation of catalyst [28]. The drawbacks of heterogeneous catalysts include slower reaction rate and less efficiency than homogenous catalysts, but heterogeneous catalysts can be designed as per the use to have enhanced properties [29]. Due to insolubility of oil in alcohol, the mass

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transfer rate gets lowered and results in low yield. The use of co-solvents, generally of medium polarity, can eliminate the problem of solubility and increase the mass transfer and hence the reaction rate and yield [30].

B. *Oligomerization*

Oligomerization of the fatty acids results in formation of chemical compounds called Estolides. Estolides show promising results as base stock for lubrication due to having superior low temperature properties, excellent cold temperature properties, and readily biodegradability [31]. Estolides offer great flexibility in designing as per the targeted physical properties. The estolides of varying physical properties, such as viscosity, density, or pour point, can be synthesized by varying the chemical structure of fatty acid chain or degree of Oligomerization [32].

Based on the chemical structure of the starting fatty acids, the estolides can be formed either by olefin addition reaction or by hydroxyl condensation reaction for fatty acids having hydroxyl groups. In olefin addition reaction, the carboxylic acid group is added to the unsaturation of another fatty acid. In hydroxyl condensation reaction, the hydroxyl group reacts with the carboxylic acid group of another fatty acid [33]. The reaction scheme of a typical estolide synthesis reaction by olefin addition mechanism is presented in figure 3.

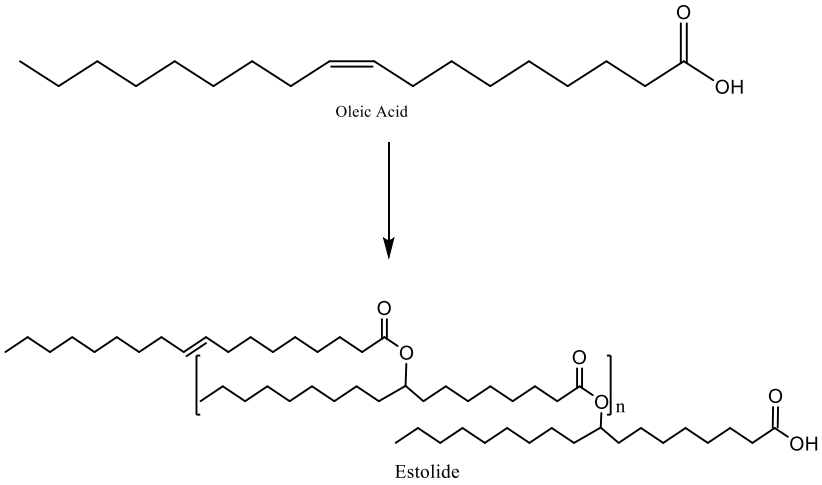

Fig. 3 Reaction Scheme of Oligomerization Reaction

The quantification or the extent of oligomerization in estolides is done by representing estolide number(EN), which shows the average number of the fatty acids added to base fatty acids [34]. The secondary ester linkage formed in the estolides enhances the physical properties of vegetable oils and results in compounds which outperform vegetable oils in many properties, but the low oxidative stability still remains in estolides and hence these often require standard additives [34, 35].

Estolides can be synthesized under acidic conditions with various mineral acids including Sulfuric acid and Perchloric acid. The sulfuric acid based estolides decompose over time due to release of sulfonic acid moieties which decrease the pH of estolide solution. The perchloric acid based synthesis provides a stable estolide, which is light colored [36]. Further up gradation can be made, such as the alcohol portion of the ester moiety can be replaced by branched chain alcohol which further reduces the pour point [37].

Due to the problems, such as strong acid catalysts and undesirable byproducts, which are associated with the conventional chemical route of synthesis of estolides, alternative production methods, based on enzymes, have been developed over time. The lipase enzymes have shown high selectivity, acts under mild process conditions, and are found as a better method than chemical for production of estolides [38].

C. *Epoxidation*

The reaction of synthesis of epoxides, which are cyclic ethers consisting of three elements in the ring, is known as Epoxidation reaction, and the product oil containing the epoxide groups is known as epoxidized oil [39]. The high reactivity of epoxides are attributed to the strongly strained three membered ring which reacts with compounds having active hydrogen and results in elimination of stress by opening of the oxirane rings [40]. As the level of epoxidation increases, the viscosity increases slightly, and the final product is more polar with significantly higher viscosity [41].

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The various vegetable oil epoxidation methods include the conventional epoxidation employing homogeneous catalyst, the heterogeneous catalyst method with catalyst such as ion exchange resins, the chemoenzymatic epoxidation method, and the epoxidation in presence of polyoxometalates [42]. The cleanest process would be to insert the oxygen from air to the site of unsaturation, but the product under here is far from industrial applications [43].

In the conventional process, which is most widely used, the vegetable oils are converted to epoxides with the help of peracids, such as peracetic or performic acids. The preparation can be divided into two stages: at first peracids are developed from corresponding acids and hydrogen peroxide in the presence of acid catalysts which activates the carbonyl carbon of the organic acid; subsequently the formed peroxy group assist in formation of epoxide group by acting as oxygen carrier [44]. The reaction mechanism of the conventional two-step process is as presented in figure 4.

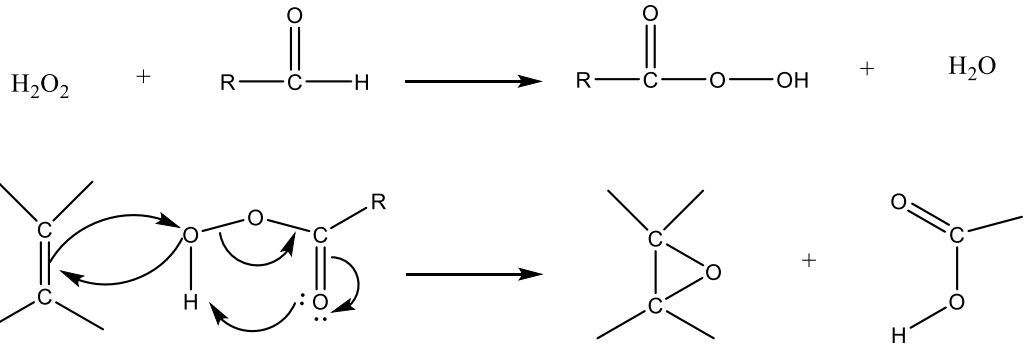

Fig. 4 Reaction Mechanism of Epoxidation Reaction [45]

The drawbacks of conventional chemical processes are that the by-products formed are strongly corrosive, the peracids formed in the first stage are unstable and explosives, and due to the oxirane ring opening the selectivity of epoxides is low [42]. The advantages of the epoxidized vegetable oils are that they are inexpensive and renewable materials which show promising results for use in industrial applications [46]. These oils have a wide portfolio of applications ranging from metal working fluids to lubricating additives, as high temperature lubricants and with ring opening as low temperature lubricants [47].

IV. RESEARCH GAPS AND FUTURE TRENDS

With some chemical modifications in the molecule moiety, waste cooking oil can be used in the synthesis of bio based lubricants. This utilization of waste, which otherwise degrades the environment, into biolubricants generates an alternative to mineral oil based lubricants and is superior to mineral oils in terms of biodegradability, renewability, and toxicity. While this operation of conversion can be employed at large scale, the primary issue arises in the collection of waste cooking oil. It is difficult to collect the used oil in large volume from sources such as kitchens and food processing industries. In addition, in the majority of the general kitchens, people utilize the used oil by mixing up with the fresh oil.

Even if the organized collection of the waste cooking oil can be taken care of, another issue arises in the economy of the product. Due to the poor oxidative and thermal stability of vegetable oils, the modifications which have to be carried on the molecule to enhance these properties add to the costing. Also, before carrying these modifications the oil must be free from such impurities which interfere with the reactions. This demands adequate purification of the oil prior to modifying it. The sum of these stages increase the price of the final commodity, making it no longer comparable to the mineral oil based lubricants which are available at a much cheaper price.

Owing to the fast depletion of the crude reserves, the price of the mineral oil based products is expected to rise in the near future and ultimately limit the reach of the product to systems with high efficiency requirements. Hence, there is an urgent need to seek alternative options. Bio-lubricant from waste oils is a feasible solution, but to account for availability and cost effective methods of purification and modifications, further research is needed in the field.

V. CONCLUSION

Finding an eco friendly approach to decrease the reliance on mineral oil sources can provide a pathway to a greener future. Waste cooking oils can be used in the synthesis of biolubricants and thus provide an alternative to waste disposal. Biolubricants mark high lubricity and biodegradability, and low toxicity. The operation of frying leads to formation of undesirable characteristics, which need to be improved before using the oil for lubrication. The pretreatment of waste

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cooking oils can improve the quality characteristics, while the chemical modifications of the structure improve the physical characteristics of the waste oil. The chemical modification methods include Transesterification reactions to form esters, Oligomerization reactions to form estolides, and Epoxidation reactions to form epoxides. All these result in products having high thermal-oxidative stability and having all the desirable characteristics which are required in a lubricant. However, there are some challenges that need to be addressed. The large scale collection of the waste oil is difficult and the oil contains impurities which need to be removed by employing filtration methods which increase the cost of the product. In order to mitigate the problems of collection of raw material and economy of the product, future research is needed in the area.

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