Chapter 2
Utilization of Vegetable Oil as Bio-lubricant and Additive

Abstract The environmental and toxicity issues of conventional lubricants as well as their rising cost lead to renewed interest in the development of environmental friendly oils as lubricants and industrial fluids. This chapter provides a review of the fundamental research works carried out on tribotesters to investigate the effectiveness of vegetable oil in suppressing wear and frictional force. Results obtained in these studies are useful to explain the mechanism by which the vegetable oils reduce friction and tool wear in machining (Chap. 3). Intensive review of the previous works shows that vegetable oils have high potential to be used as lubricant and additive to replace conventional lubricants and additives.

Keywords Bio-additives · Bio-lubricant · Vegetable oil lubricant · Tribotesters · Palm oil lubricant

2.1 Exploration for Environmental Friendly Lubricant Additives

Additives are widely used to improve the lubricant performance of base oil. Without additives, even the best base fluids are deficient in some features. The performance of a lubricant depends collectively on the base oil, additives and formulation. Phosphorus, sulphur, zinc dialkyldithiophosphates (ZDDP) are examples of some of the widely used additives. Sulphur-containing additives are probably the earliest known additive compounds in lubricants. In recent decades, it had attracted a considerable amount of research efforts to further explore their potential as effective anti-wear (AW) and extreme pressure (EP) additive (Zhang et al. 1999; Bhattacharya et al. 1995).

Fatty acids, alcohols, amines and esters are some of the AW additives used to produce a molecular film adhering to the surfaces by physical or chemical adsorption (Stachowiak and Batchelor 2005). The lubricant films are built up of orderly and closely packed arrays of molecular layers, with the polar head of the additive

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molecule anchored on the worn surface (Kenbeck and Bunemann 2009). There are also strong dipole interactions between the chains. The effectiveness of the lubricant depends greatly on the tenaciousness of the bond between the polar end group of the molecular chain and the metal surface where it adheres to (Tan et al. 2002).

Sulphur-, chlorine- and phosphorus-containing compounds are commonly used as EP additives to provide protection in EP condition (Canter 2007). These additives would form layers of iron compounds such as sulphides, chlorides and phosphates, respectively, through tribochemical reactions (Hsu and Gates 2005). The mechanism of lubrication which is influenced by these additive elements involves some chemical changes on the surface to form a surface protection film. This film is called boundary lubricating film or a tribofilm. The tribofilm plays a major role in determining the friction and wear of the tribological interaction. The morphology, integrity and mechanical properties of the tribofilms may vary depending on the properties of rubbing material as well as the type of lubricant additives used (Biswas 2000; Kim et al. 2010).

ZDDP was initially used as an antioxidant, but their excellent AW properties were quickly recognised and had been investigated intensively by many researchers. The AW function of ZDDP was attributed to its decomposed products that led to the formation of sacrificial reaction layers on the rubbing surfaces. A variety of ZDDP decomposition mechanisms and the associated chemistry of reaction film had been proposed by many researchers (Mosey et al. 2005; Fuller et al. 1997, 1998; Brancroft et al. 1997; Willermet et al. 1995; Spedding and Watkins 1982). However, the concern for the content of heavy metal zinc and phosphorus as environmental contaminants had resulted in efforts to find more environmentally benign replacements for industrial applications (Cardis et al. 1989). It was stated that even ashless sulphur-containing compounds do not necessarily have good ecotoxicological profiles for environmentally friendly lubricants. Environmentally friendly lubricants must also have high level of biodegradability (Habereder et al. 2009).

The environmental and toxicity issues of conventional lubricants as well as their rising cost related to a global shortage and their poor biodegradability led to renewed interest in the development of environmental friendly lubricants. Environmental legislation by OSHA and other international regulation authorities discourage the use of mineral oil-based lubricant and environmental-harmful additives. There has been increasing demand for green lubricants and lubricant additives in recent years. Vegetable oils are viable and good alternative resources because of their environmental friendly, non-toxic and readily biodegradable nature. The majority of bio-lubricants are based on esters. There are natural esters which are triglycerides of vegetable oils. Oleochemical esters of fatty acids such as diesters, polyolesters and complex esters are derived from sunflower, rapeseed, palm oil and coconut. Triglycerides of vegetable oils are more polar than petroleum-based oils, thus they have a higher affinity to metal (Suarez et al. 2010). Owing to this character, vegetable oils and their derivatives are suitable for lubrication applications. Conversely, their low thermo-oxidation stability, primarily due to the presence of bis-allylic protons is the main limitation (Fox and Stachowiak 2007; Becker and Knorr 1996). They also have poor corrosion resistance (Ohkawa et al. 1995). Some studies have also shown that most vegetable oils undergo cloudiness, precipitation, poor flow, and solidification upon long-term exposure to cold temperature (Rhee et al. 1995; Kassfeldt and Goran 1997). Erhan et al. (2006) have demonstrated that thermo-oxidative stability and cold flow property can be improved using a combination of proper blending of chemical additives, diluent and high-oleic vegetable oils. Another major obstacle is the cost of the bio-lubricants. A bio-lubricant costs somewhere between 30–40 % more compared to a conventional lubricant. Lubricant formulations for more environmentally benign are, therefore, being developed based on their benefits and limitations.

Oils with more polar groups (like carboxylic acids and esters) possess more sites to react and adsorb with metal surfaces to provide boundary lubrication effects (Stachowiak and Batchelor 2005). A lubricating film with strong bonding to the surface and adequate cohesive interaction among lubricant molecules can effectively reduce the friction and the amount of wear. To maintain a low friction and wear, the lubricating film has to withstand extremes of temperature variations, shear degradation and maintain excellent boundary lubricating properties through strong physical and chemical adsorption with the metal.

The additive molecules dissolved in the oil are attracted to the surfaces by adsorption forces governed by their polarity (Sharma et al. 2009; Kalin et al. 2006). It was found that the friction reduction effect increased with larger amount of adsorptive polar group of the additive in the base oil (Tohyama et al. 2009; Kurth et al. 2007; Adhvaryu et al. 2004). In fact, the polarity of both the base fluids and the additives is very important because each component of the mixture is competing for the metal surfaces reaction. A polar additive that would normally adsorb and desorb reversibly from a metal surface in a non-polar base fluid might have a much lower concentration on the surface than in a formulation that contains a high concentration of polar base fluid such as ester or vegetable oil (Rudnick 2009). Works by Suarez et al. (2010) shows that the wear performance of ZDDP additives contained in polar base fluid is better than that of ZDDP blended in a non-polar base fluid as smaller wear track width and larger load carrying capacity features were observed on the wear track produced in the former lubricant. Hsu et al. (1988) suggested that the first and the foremost of the dynamic and sequential competition in a solution that contains more polar groups is the preferential adsorption of the most polar molecules onto the surface at a particular temperature. However, the increase in the amount of polar compounds in the oil could reduce the adsorption of additives on the metal surface due to competitive adsorption, whereby the efficiency of AW additives could thus decrease (Studt 1989) and it could also provide corrosive effects (Hsu and Gates 2005; Jimenez and Bermudez 2007).

2.2 Wear and Friction Reduction by Vegetable Oil as Bio-lubricant and Additive

Vegetable oils are viable and good alternative resources because of their environmental friendly, non-toxic and readily biodegradable nature. The triacylglycerol structure with long fatty acid chains and presence of polar groups in the vegetable oils make them amphiphilic in character, therefore allowing them to be an excellent choice as lubricants and functional fluids. These triacylglycerol molecules in vegetable oils orient themselves with the polar end at the solid surface making a closed packed monomolecular or multimolecular layer resulting in a surface film that provides desirable qualities in a lubricant (Rudnick 2009). Other advantages include very low volatility due to the high molecular weight of the triglyceride molecule and excellent viscosity properties. Table 2.1 shows several type of vegetable-based lubricants developed for industry applications.

Vegetable oils may not suitable to be used as lubricants in their natural form due to their poor thermo-oxidation stability, low temperature behaviour and other tribochemical degrading processes that occur under severe conditions of temperature, pressure shear stress and environment (Fox and Stachowiak 2007). However, they can be used effectively as additives, in particular to improve the polarity behaviour of non-polar base fluid solutions, which would contribute to better tribological performance. In the past several researchers have investigated the effectiveness of methyl ester as additive in diesel. Sulek et al. (2010) found that the presence of fatty

Table 2.1 Several type of vegetable-based lubricants developed for industry applications (Shashidhara and Jayaram 2010)

Type of oil	Application
Canola oil	Hydraulic oils, tractor transmission fluids, metalworking fluids, food grade lubes, penetrating oils, chain bar lubes
Castor oil	Gear lubricants, greases
Coconut oil	Gas engine oils
Olive oil	Automotive lubricants
Palm oil	Rolling lubricant,-steel industry, grease
Rapeseed oil	Chain saw bar lubricants, air compressor-farm equipment, Biodegradable greases
Safflower oil	Light-coloured paints, diesel fuel, resins, enamels
Linseed oil	Coating, paints, lacquers, varnishes, stains
Soybean oil	Lubricants, biodiesel fuel, metal casting/working, printing inks, paints, coatings, soaps, shampoos, detergents, pesticides, disinfectants, plasticisers, hydraulic oil
Jojoba oil	Grease, cosmetic industry, lubricant applications
Crambe oil	Grease, intermediate chemicals, surfactants
Sunflower oil	Grease, diesel fuel substitutes
Cuphea oil	Cosmetics and motor oil
Tallow oil	Steam cylinder oils, soaps, cosmetics, lubricants, plastics

acid methyl ester derived from rapeseed oil in diesel fuel resulted in 20 % decrease in friction and twofold decrease in wear. Similarly, Sukjit and Dearn (2011) demonstrated that adding as little as 5 % of fatty acid methyl ester derived from rapeseed in diesel fuel could result in reduction in the wear scar diameter by 40 %.

Malaysia is often viewed as a country that evolved from dependence on tin and rubber to export-oriented manufacturing dominated by electronics assembly, but the commodity that made the country to the technological frontier is palm oil. Palm oil is now a major pillar of Malaysia's industrialization and it holds a considerable lead in global markets. To ensure a sustainable growth of palm oil industry in the country and remains competitive in the global market, palm oil industry in Malaysia in recent years has been shifting to palm oil product diversification from a conventional commercial cultivation as its main export focus until more years to come. Research and development effort, therefore, became more critical, in particular, to explore and develop new palm oil-based products for higher value added in the palm oil chain. Recently, it has been promoted as a biofuel feedstock in compression ignition engines (diesel engines). Palm oil methyl ester has an ester functional group which is a classic example of additive used for lubrication (Canter 2007). Characterization palm oil methyl ester can be carried out using fourier transform infrared spectroscopy (Liew et al. 2014). Palm oil methyl ester was produced from crude palm oil through transesterification process, whereby the triglyceride of palm oil was reacted with an alcohol in the presence of a catalyst as represented by general equation in Fig. 2.1. R_1, R_2 and R_3 represent the hydrocarbon chains of the fatty acid of the triglyceride. This reaction yielded esters and glycerol, which are then separated, in which glycerol being removed as by-product. The palm oil methyl ester was characterised using fourier transform infrared spectroscopy (FTIR). The FTIR spectra shows 1,750 and 1,150 cm^{-1} peaks (Fig. 2.2) that correspond to C=O and C–O esters (Taufiq-Yap et al. 2011).

Various investigations had shown that palm oil methyl ester additives improved the lubrication performance of the diesel base oil (Masjuki and Maleque 1996a, 1996b, 1997; Maleque et al. 2000). Masjuki and Maleque (1997) reported that adding 5 vol% of palm oil methyl ester in the base oil lubricant resulted in low wear rate of EN31 steel ball bearing. Palm oil methyl ester, converted from crude palm oil through transesterification, has very low sulphur content (0.002 wt%), and therefore is environmental friendly. Liew et al. (2014) found that in the

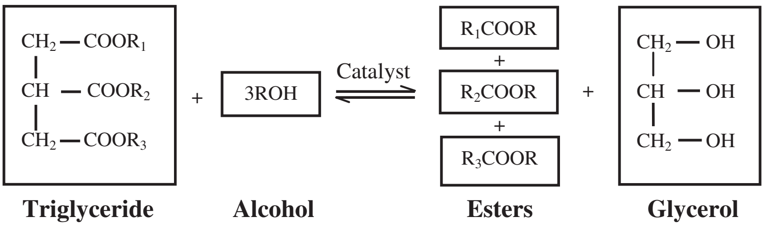

Fig. 2.1 Transesterification reaction for producing esters from oil (triglyceride)

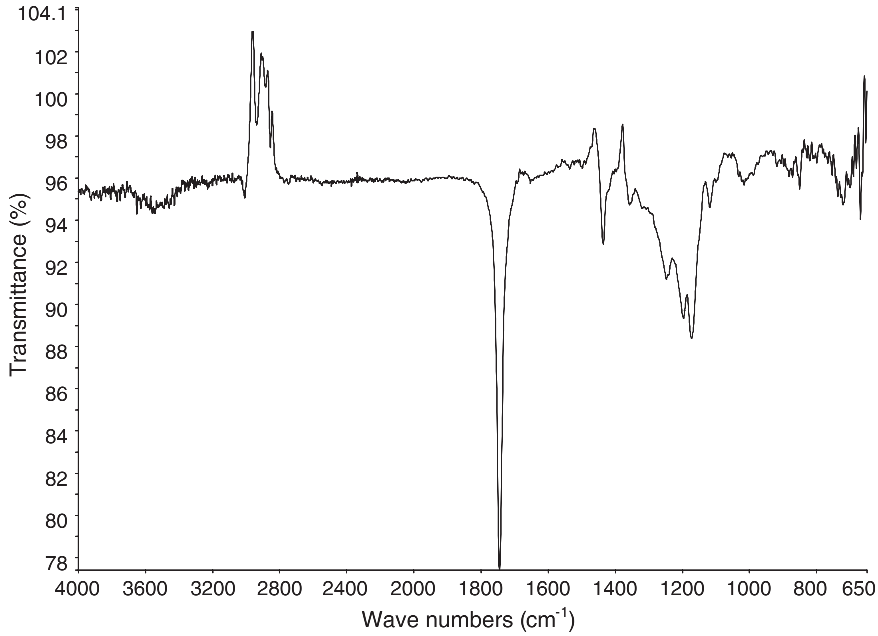

Fig. 2.2 The IR spectrum of palm oil methyl ester (Liew et al. 2014)

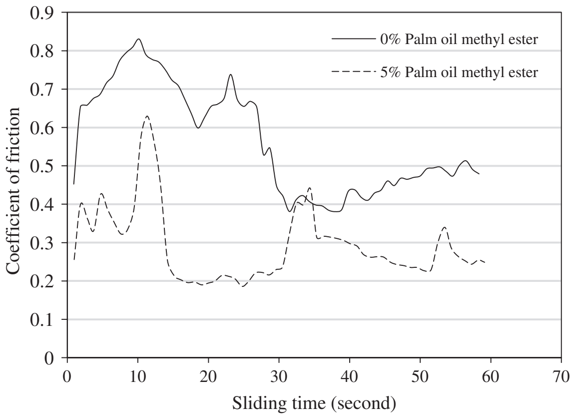

Fig. 2.3 The change in friction coefficient in different lubrication conditions at nominal load of 1,100 N (Liew et al. 2014)

presence of palm oil methyl ester in the mineral oil resulted in a shorter running-in period and lower steady-state frictional coefficient at nominal load of between 600 and 800 N. The difference in the friction coefficient produced in mineral oil with and without palm oil methyl ester became more apparent at loads above 800 N (Figs. 2.3 and 2.4). The performances of lubricants in EP can also be expressed in terms of welding load (Kabuya and Bozet 1995; Singh and Verma 1991). Under mineral oil without palm oil methyl ester, complete welding of the four

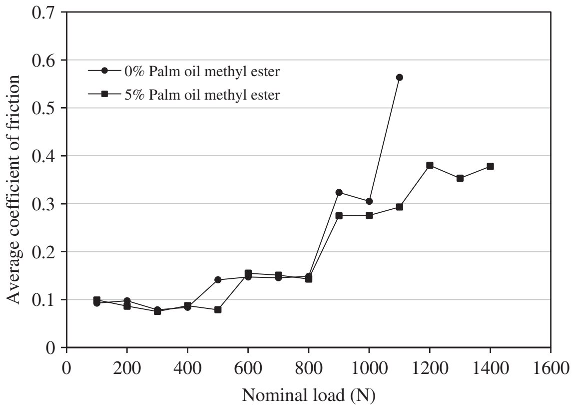

Fig. 2.4 Effect of nominal load and lubrication condition on the average coefficient of friction (Liew et al. 2014)

Table 2.2 Weld load and wear scar diameter for different lubrication condition

Lubrication condition	Weld load (N)	Average diameter scar (mm) produced at the nominal loads of		
		300 N	600 N	800 N
Mineral oil (without palm oil methyl ester)	1,200	0.28	1.97	2.60
Mineral oil (with 5 vol% palm oil methyl ester)	1,450	0.29	1.79	2.30

balls occurred at 1,200 N (Table 2.2). The presence of palm oil methyl ester in oil resulted in a higher critical load of 1,450 N.

Gong et al. (2003) investigated the wear reduction bought about by two kinds of synthetic thiophosphate (tri-*n*-octyl thiophosphate and tri-*n*-octyl tetrathiophosphate) and tricresyl phosphate as additives in rapeseed oil in sliding of steel. Synthetic thiophosphate resulted in lower wear and this could be attributed to the tribochemical reactions between the steel and the thiophosphate, and the formation of a boundary and protective layer on the worn surfaces. A series of long-chain dimercaptothiadiazole derivatives had been tested as AW and EP additives in vegetable oil using a four-ball tester. The long-chain thiadiazole derivatives were capable of improving the EP characteristic of the base colza oil. Thermal films generated from these derivatives are composed of ferrous sulphate and a small amount of adsorbed organic sulphide (Chen et al. 2012). Work by Gao et al. (1999) showed that thiadiazole derivatives in paraffin oil under boundary lubrication at high loads resulted in greater friction reduction and exhibited better AW properties than lubricant containing ZDDP. It was also found that thiadiazole derivatives had better antioxidative and anticorrosive properties than ZPPD.

Synthesis of vegetable oil and thiols could result in the formation of hydroxyl thioether derivatives in the vegetable oil. This process retained the vegetable oil structure and its associated benefits such as high flash point, viscosity index, lubricity and eco-friendly but removed poly-saturation in the fatty acid chain with addition of polar functional groups that significantly improved surface adsorption on metal leading to a reduction in wear and friction coefficient (Sharma et al. 2009). Xu et al. (2014a) demonstrated that catalytic esterification of crude bio-oil derived from spirulina algae resulted in enhanced lubrication performance. The coefficient of friction produced by ethanol blended with bio-oils esterified using potassium fluoride/alumina and potassium fluoride/HZSM-5 zeolite as catalysts was 22 and 10 % lower, respectively, than that produced by crude bio-oil blended with ethanol. The esterified bio-oils produced lower friction coefficient because it resulted in the formation of a better protective tribofilm on the worn surfaces.

Shi et al. (2014) studied the effect of water content in glycerol solution on the viscosity, friction coefficient, wear loss and film thickness under elastrohydrodynamic and boundary lubrication. Despite the viscosity and the film thickness of the glycerol solutions decreased greatly with increasing water content, there was an optimum amount of water in the glycerol solutions which resulted in the lowest friction coefficient. A correlation was not found between friction coefficient and wear volume loss. Under elastrohydrodynamic lubrication, the friction coefficient of rapeseed oil was about three times higher than that of pure glycerol. Under boundary lubrication, the friction coefficient produced by rapeseed oil and pure glycerol was similar. However, a more stable and lower friction coefficient was produced when the glycerol solution consisted of 5–20 wt% of water. While the water content was beneficial for glycerol to decrease the friction coefficient, excessive water content resulted in marked reduction in the viscosity and thus the load carrying capacity of the solution. These works showed that glycerol aqueous solutions have great potential to replace rapeseed oils as environmentally friendly base oils.

Although coconut oil is more stable than many vegetable oils, it is not widely used due to its high congelation temperature. Being a vegetable oil having typical triacylglycerol structure, it has most of the salutary properties of other vegetable oils as lubricants such as high viscosity index, good lubricity, high flash points and low evaporative loss. Although it has similar disadvantages of poor properties at low temperature, it shows much better thermal and oxidative stability owing to its predominantly saturated nature of its fatty acid constituents. Jayadas and Prabhakaran (2006) found that adding 2 wt% of ZDDP in coconut oil resulted in significant wear reduction in the four-ball test. The welding load of the coconut oil containing 2 wt% of ZDDP was higher than that produced by commercial 20W50 Oil.

Quinchia et al. (2014) studied the frictional and lubricating film-forming properties of various type of improved vegetable oils based lubricants (high-oleic sunflower, soybean and castor), using 4 wt% of ethylene–vinyl acetate copolymer and 1 wt% of ethyl cellulose as additives. It was found that castor oil showed the best lubricant properties, when compared to high-oleic sunflower and soybean oil, with very good film-forming properties and excellent friction and wear behaviour. This could be attributed to its hydroxyl functional group that increased both the viscosity and polarity of this vegetable oil. Ethylene–vinyl acetate copolymer exerted a slight effect on the lubricating film-forming properties, reducing the friction and wear mainly in the mixed lubrication region. Ethyl cellulose, on the other hand, was much more effective mainly with castor oil, in improving both mixed and boundary lubrication.

Alves et al. (2013) found that modified vegetable oils such as epoxidised sunflower and soybean oils resulted in lower friction coefficient than the mineral and synthetic oils. However, the presence of CuO and ZnO nanoparticles in the epoxidised vegetable oils resulted in higher friction coefficient and wear. It was postulated that the effect of nanoparticles on the wear and friction coefficient was governed by the nature of the adsorption of the lubricant on the contact surfaces. Adherence of the polar groups of the vegetable oils on the worn surface caused the nanoparticles to roll and hence three-body abrasion to take place, resulting in increased wear. The reduction in the wear and friction coefficient bought about these oxides nanoparticles in mineral and synthetic oils could be attributed to adherence of the nanoparticles and formation of a physical tribofilm on the worn surface. Xu et al. (2014b) reported that emulsified bio-oil (produced form the fast pyrolysis of rice husk) produced the lowest coefficient of friction. This was followed by bio-oil and diesel oil. The diesel and bio-oils produced the lowest and the highest wear, respectively. It was concluded that the emulsified bio-oil produced the best overall results and this was due the presence of various acidic components with polar groups in the emulsified bio-oil. Suarez et al. (2009) demonstrated that adding some soybean oil methyl esters and diesel-like pyrolytic fuel (produced through pyrolysis of soybean oil) enhanced the lubricity of diesel fuels. This result showed the potential use of both bio-fuels as additives for improving the tribological property of fossil fuels. Another work (Xu et al. 2010) found that the lubrication ability of the diesel fuel blended with bio-oil (produced through fast pyrolyzing rice husk) was better than that of the conventional diesel fuel. However, the presence of bio-oil in the diesel fuel resulted in inferior anti-corrosion and anti-wear properties.

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