



## An overview of research on biolubricants in Malaysia and Japan for tribological applications

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KEYWORD	ABSTRACT
Biolubricants Friction Wear Biodegradability Sustainability	Significant research and development is being done to substitute petroleum based lubricants with biolubricants derived from vegetable oils having similar physicochemical properties and tribological characteristics. This paper presents an overview of biolubricants in Malaysia and Japan for tribological applications. Research and development trends on biolubricants are described under the following categories: biolubricants as basestock, biolubricants in mixtures and biolubricants with additives. Excellent lubricant performance is obtained by biolubricant blends with selected additives. Biolubricants having similar performance to petroleum based lubricants, reduce dependence on nonrenewable resources, and increases markets industrial applications.

### 1.0 REVIEWS ON BIOLUBRICANTS

Increasing attention towards sustainability and environmental issues has driven substituting mineral oils with biolubricants. Research and development is being done on biolubricants for effective lubrication with superior tribological performance under wide range of operating conditions and contact geometries. Reviews on the available literature to explore the potential of biolubricants have been published. Research works related to review on biolubricants are presented in Table 1. A critical review has been made on vegetable-based lubricant additives with

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specific properties, manufacturing processes, advantages and applications (Maleque et al., 2003). A study on the source, properties, advantages, disadvantages, potential of vegetable oil-based biolubricants as alternative lubricants for automobile applications, the world biolubricant market and its future prospects are discussed (Mobarak et al., 2014). Currently, there is a significant interest in developing biolubricants derived from organic sources as additives to drilling fluids. Reviews on the potential of biolubricants derived from vegetable oils exceeding the performance of hydrocarbon and mineral lubricants are presented (Kania et al., 2015). Polyol esters of vegetable oils demonstrate suitability of biolubricants for drilling applications. An overview of the development in the thermochemical conversion methods of plant oils to biolubricants is discussed (Yunus and Luo, 2017). An accurate evaluation of cost-effective technology is crucial for the future of the plant oil-based lubricants. Syahir et al. (2017) highlighted the potential of biolubricants for broad range of applications based upon the published researches over the past decade. The correlation between molecular structures, physicochemical properties and lubrication performance of natural oil were reviewed which is essential for lubricant development and selection. Zainal et al. (2018) presented a detailed treatment on biolubricants, the various vegetable oils used as the feedstocks, the processes used for chemical modification of vegetable oils, the lubrication properties, as well as the various additives used to improve the properties of biolubricants.

Biolubricants hold great potential for environmental conservation and economic development as an alternative lubricant. This study presents an overview of research on biolubricants in Malaysia and Japan for tribological applications. The various works on formulation of biolubricants as basestock, biolubricants in mixtures and biolubricants with additives are highlighted.

Table 1: Reviews on biolubricants.

<b>Description of reviews on biolubricants</b>	<b>Authors</b>
Vegetable-based lubricant additives with specific properties and application	Maleque et al. (2003)
Potential of a biolubricant based on vegetable oil as an alternative lubricant	Mobarak et al. (2014)
Potential of biolubricants in drilling fluids	Kania et al. (2015)
Development in the thermochemical conversion methods	Yunus and Luo (2017)
Potential of biolubricants for broad range of applications	Syahir et al. (2017)
Improvement in properties of bio-based lubricants	Zainal et al. (2018)

## 2.0 RESEARCH ON BIOLUBRICANTS IN MALAYSIA FOR TRIBOLOGICAL APPLICATIONS

Abdollah et al. (2016) discussed the current tribology research trends in Malaysia and highlighted green tribology (biolubricants) research and development pioneered in Malaysia from the early 1980's. Malaysian Tribology Society continues to cooperate in promoting research activities in tribology. In Malaysia, palm oil has the potential to be used an industrial biodegradable vegetable oil. Palm oil in Malaysia, with high production rate will be able to meet the demand for vegetable-based lubricants (Syahrullail et al., 2011).

## 2.1 Biolubricants as basestock

Research works related to biolubricants in Malaysia are presented in Table 2. Hydraulic system performance using palm oil as hydraulic fluid as a potential useful substitute for mineral based energy transport media is investigated (Wan Nik et al., 2007). Ghani et al. (2015) presented the performance of both inorganic commercial lubricants and environmental friendly organic palm oil based lubricants in high speed machining of FCD700 cast iron using carbide tool common in manufacturing and automotive industries. It is suggested that attention should be given to the additive elements to improve the performance of the palm oil-based lubricant in order to prolong the tool life. Investigations on coating and surface modifications using bio-based lubricant synthesized from palm oil methyl ester, pentaerythritol, and trimethylolpropane were discussed (Zulkifli et al., 2016). TMP and PE ester provided maximum load capacity under extreme pressure condition. PE showed the lowest CoF of around 0.025 compared to ordinary lubricant at around 0.07 under mixed lubrication conditions. Razak et al. (2015) conducted a tribological study in hip implants to elucidate the performance of palm lubrication between the femoral head and the acetabular cup. The use of palm olein, palm kernel oil, and palm fatty acid distillate as synthetic lubricants for human joints has shown tremendous potential. Al Mahmud et al. (2015) investigated tribological properties of ta-C diamond-like carbon (DLC) coating under DLC-steel contact condition using three vegetable-based oils (palm, sunflower, and coconut). Sunflower oil exhibited better tribological characteristics than coconut oil as a lubricant. A stable transfer layer formed in the cases of sunflower and palm oil lubrication. However, high wear rate and full delamination of the coated plate under coconut oil lubrication did not show a stable transfer layer.

Table 2: Research on biolubricants as basestock in Malaysia.

<b>Biolubricants as basestock</b>	<b>Authors</b>
Vegetable palm oil degrades faster than mineral oil, and thus viscosity increases significantly with aging condition	Wan Nik et al. (2007)
Palm oil based lubricant creates an adhesive layer of lubricant on the metal surface that provides better surface finish	Ghani et al. (2015)
Saturation level of vegetable oils is an important factor for tribological characteristics of tetrahedral DLC coating	Al Mahmud et al. (2015)
PE and TMP esters had comparable characteristics to the commercial lubricant in terms of coefficient of friction	Zulkifli et al. (2016)
Palm oil can optimize rate of friction and wear on metal acetabular cup and allow for stable MoM hip implants	Razak et al. (2015)
Transfer layer formation by ta-C DLC helps to reduce friction and protect the surface from severe wear	Al Mahmud et al. (2015)
RBD palm stearin has satisfactory performance in reducing the extrusion load compared to additive-free paraffinic mineral oil	Syahrullail et al. (2011)
RBD PS showed friction coefficient reduction compared with additive-free PMO	Tiong et al. (2012)

RBD palm olein has lower coefficient of friction compared to mineral oil	Ing et al. (2012)
Jatropha oil, RBD palm olein and PFAD showed potential for development as an industrial lubricant.	Syahrullail et al. (2013a)
PFAD exhibited better performance in terms of friction while jatropha oils showed better performance in terms of wear	Golshokouh et al. (2012)
Lower wear resistance is related to a higher COF for double fractionated palm oil (DFPO)	Noorawzi and Samion (2016)
Transesterification process of palm oil using TMP ester reduces oxidation stability	Zulkifli et al. (2014a)
CI TMP ester fatty acid creates a boundary film and thus increases lubricity	Habibullah et al. (2015)
Crude jatropha oil (CJO) oxidized by autoxidation mechanism increases viscosity of the oil	Lubis et al. (2015)
Epoxidized jatropha oil (EJO) showed lower friction coefficient and wear properties compared to crude jatropha oil (CJO)	Lubis et al. (2017)

Syahrullail et al. (2011) evaluated viability of RBD palm stearin (Refined, Bleached and Deodorized Palm Stearin, a refined type of palm oil) when used as a lubricant in forward strain extrusion process. RBD palm stearin is the solid fraction obtained by fractionation of palm oil after crystallization at controlled temperature. RBD palm stearin can reduce extrusion load and can produce a product with good surface finish. Tiong et al. (2012) also investigated tribological performance of refined, bleached and deodorised (RBD) palm stearin (PS). The existence of fatty acids in additive-free refined, bleached and deodorised (RBD) palm stearin (PS) effectively increased the presence of lubricant film that resulted in low coefficient of friction compared to additive-free paraffinic mineral oil (PMO). Ing et al. (2012) investigated friction and wear performance of refined, bleached and deodorized (RBD) palm olein. RBD palm olein has showed lower coefficient of friction compared to paraffinic mineral oil. Syahrullail et al. (2013a) investigated performance of stamping oil, hydraulic oil, jatropha oil, RBD palm olein and palm fatty acid distillate under extreme pressure conditions. Jatropha oil showed the smallest coefficient of friction and wear scar diameter, followed by RBD palm olein, and palm fatty acid distillate (PFAD). Stamping oil showed lowest coefficient of friction and wear scar diameter due to presence of anti-wear additives. Golshokouh et al. (2012) compared the performance of palm fatty acid distillate (PFAD) and jatropha oil with mineral oils. PFAD and Jatropha oils showed lower friction torque compared to hydraulic and engine mineral oils. Jatropha oil has a better condition and more stability against wear. Noorawzi and Samion (2016) investigated friction and wear characteristics of double fractionated palm oil (DFPO) as a biolubricant. Coefficient of friction (COF) of double fractionated palm oil (DFPO) increased at higher sliding speed and load.

Zulkifli et al. (2014a) outlined the extreme pressure and temperature characteristics of a palm oil ester-based (POME, palm oil methyl ester) biolubricant synthesised with trimethylolpropane (TMP) and with alkaline as a catalyst. TMP esters improve COF by up to 15% compared to paraffinic oil. Tribological properties in terms of COF and WSD of Jatropha are better compared

to palm due to higher content of triester in *Jatropha*. Lubricant film strength formed at surface contact zone increases due to triester branching structure and increasing polar functional groups, thereby reducing friction and wear. Habibullah et al. (2015) investigated tribological performance of calophyllum inophyllum (CI) based trimethylolpropane (TMP) ester as an energy efficient, sustainable and biodegradable lubricant. The trimethylolpropane (TMP) ester produced from calophyllum inophyllum (CI) oil, has higher density, higher viscosity, higher viscosity index (VI) and lower coefficient of friction compared to paraffin mineral oil and commercial lubricant. Lubis et al. (2015) studied oxidation and thermal degradation of crude *jatropha* oil (CJO). The composition of the tribo-layer formed during sliding of steel in the presence of crude *jatropha* oil (CJO) and epoxidized *jatropha* oil (EJO) under boundary lubricant application was investigated (Lubis et al., 2017). Crude and epoxidized *jatropha* oil (CJO and EJO) oxidation led to oil chemisorption causing formation of protective layers on the surfaces.

## 2.2 Biolubricants in mixtures

Research works related to biolubricant mixtures in Malaysia are presented in Table 3. Masjuki and Maleque (1997) investigated palm oil methyl ester (POME) blends in diesel engine oil to assess the anti-wear characteristics in elastohydrodynamic lubrication. The addition of 5 wt.% POME to the engine oil decreases coefficient of friction and improves smoothness of wear scar surfaces and thus acts as an anti-wear lubricant additive. In the early 1990s, Palm Oil Research Institute of Malaysia (presently known as Malaysian Palm Oil Board) successfully converted crude palm oil (CPO) to palm oil methyl ester (POME) through transesterification (Ing et al., 2012). The pour point behaviour and thermal stability can be ameliorated by transesterification. Maleque et al. (2000) studied tribological performance of 5% palm oil methyl ester (POME) blend in commercial lubricant. Wear rates of 5% POME lubricant are lower, at lower loads and temperatures, whereas at higher loads and temperatures the wear rates are higher. Maleque and Masjuki (2002) investigated tribological performance of palm oil methyl ester (POME) blended with mineral oil. Wear properties are strongly influenced by the use of lubricants and their additives. The presence of POME as an additive in lubricating oil causes changes in the material surface characteristics by the formation of protective boundary films which results in increased wear resistance of the mating surfaces.

Syahrullail et al. (2013b) tested lubricating properties of palm fatty acid distillate (PFAD) mixed in commercial metal forming oil. The coefficient of friction reached its lowest value of 0.054 using a mixture of 20 % total mass of palm oil in the mineral oil. Performance of mineral oil could be enhanced by mixing it with vegetable oil using proper compositions. Most vegetable oils contain methyl ester, monoglycerides, diglycerides, free fatty acids, and triglycerides that improve its lubricating properties. Triglycerides of vegetable oils with long polar fatty acid chains provide high strength lubricant films on metallic surfaces. Refined, bleached, and deodorized palm olein blended with mineral oils was investigated employing image processing techniques to explore the optimum blend that provides enhanced tribological characteristics (Hassan et al., 2016). RBD palm olein and mineral engine oil blend (E53.11/RB 46.89) offers lower coefficient of friction (COF) and smaller wear scar diameter (WSD) compared to the mineral engine oil and neat palm olein. The tribological performance of mineral oil mixed with refined bleached deodorised (RBD) palm stearin has been studied (Zuan et al. (2017)). Coefficient of friction (COF) of 100% mineral oil has the highest value compared to mineral oil mixed with RBD palm stearin and 100% RBD palm stearin. Wear scar diameter (WSD) of 100% mineral oil has the lowest value. Tribological performance of lubricant of refined, bleached and deodorised (RBD) palm olein and

semi-synthetic engine oil were tested (Aiman and Syahrullail, 2017). Engine oil mixed with RBD palm olein (EO80-PO20) show reduction in coefficient of friction (0.071) compared to pure engine oil (0.087). Wear scar diameter of engine oil mixed with RBD palm olein (EO80-PO20) show 40% improvement in performance compared to RBD palm olein.

Table 3: Research on biolubricants in mixtures in Malaysia.

<b>Biolubricants in mixtures</b>	<b>Authors</b>
POME as an additive in diesel engine oil improved the anti-wear characteristics	Masjuki and Maleque (1997)
Friction behavior of POME as an additive in commercial lubricant indicates boundary lubrication regime	Maleque et al. (2000)
POME blend with mineral oil has remarkable influence on tribological characteristics of surfaces in contact	Maleque and Masjuki (2002)
Mixture of palm fatty acid distillate (PFAD) in commercial metal forming oil (CMFO) has potential to reduce coefficient of friction	Syahrullail et al. (2013b)
RBD palm olein blend with mineral engine oil can be an effective alternative biolubricant due to good lubrication performance	Hassan et al. (2016)
Mineral oil mixed with RBD palm stearin reduces coefficient of friction	Zuan et al. (2017)
Performance improvement in mixing RBD palm olein with semi-synthetic engine oil	Aiman and Syahrullail (2017)
An optimum addition of Jatropha oil in mineral base oil for automotive application is recommended	Shahabuddin et al. (2013)
Palm oil-based TMP ester in ordinary lubricant reduces coefficient of friction in boundary and hydrodynamic lubrication	Zulkifli et al. (2013a)
TMP ester blended with paraffin oil provide better surface protection compared to paraffin oil	Zulkifli et al. (2014b)
TMP and PE lubricants derived from POME have comparable tribological characteristics to that of commercial lubricant	Zulkifli et al. (2016)

Various blends of jatropha oil mixed with mineral oil base lubricant (SAE 40) were used to evaluate friction and wear characteristics under boundary lubrication conditions (Shahabuddin et al., 2013). Wear rate with addition of 10% jatropha oil to base lubricant (mineral oil) is similar to base lubricant and thus can help to reduce the global demand of petroleum based lubricants substantially. Zulkifli et al. (2013a) evaluated tribological characteristics of palm oil based trimethylolpropane (TMP) ester in ordinary lubricant under boundary and hydrodynamic lubrication. TMP ester produced from palm oil is biodegradable and has high lubricity properties, such as a higher flash point temperature and viscosity index. Mechanical efficiency of machinery

component increases with decreasing coefficient of friction. For boundary lubrication, addition of 3% of TMP ester in ordinary lubricant (OL) decreases the maximum wear scar diameter (WSD) and reduces coefficient of friction (COF) up to 30%. For hydrodynamic lubrication, addition of 7% of TMP reduces the friction up to 50%. TMP ester has an affinity to surface asperities, which reduces wear between sliding contacts and is a potential substitute for petroleum-based lubricants in automotive engines. Zulkifli et al. (2014b) investigated extreme pressure characteristics of a palm oil based trimethylolpropane (TMP) ester blended with paraffin oil. Biolubricant properties and lubrication regime have strong influence on coefficient of friction and wear scar diameter. Zulkifli et al. (2016) studied lubricity properties of trimethylolpropane (TMP) and pentaerythritol ester (PE) produced from palm oil methyl ester as a source of biolubricant. The effects of different sources of esters in different lubrication regimes are investigated. TMP and PE ester retained maximum load bearing capacity compared to paraffin under extreme pressure conditions. PE showed lowest COF at 0.025 compared to ordinary lubricant at 0.07 under mixed lubrication conditions.

### 2.3 Biolubricants with additives

Research works related to biolubricants with additives in Malaysia are presented in Table 4. Ahmed et al. (2014) studied a blend of soybean oil, mineral oil, and additives to in order to develop a biolubricant having viscosity fitted to a commercialized industrial lubricant (ISO VG 68). A blend having a mixture of 52.70 wt % soybean oil, 40.55 wt % mineral oil, and 6.75 wt % additives produced optimum results for viscosity fitting. Sapawe et al. (2016) investigated ways in which oxidation can be contained by combination of palm oil with a phenolic antioxidant (homogeneous mix of palm oil and tertiary-butyl hydroquinone). Palm oil with tertiary-butyl hydroquinone provides better protection at the metal contacting surfaces due to increased lubricity. Palm oil with antioxidant can control oxidation and reduce wear scars by retaining boundary lubricant film resulting from fatty acid molecules. Farhanah and Syahrullail (2016) evaluated the lubrication performance of refined, bleached and deodorized (RBD) palm stearin (PS) with Zinc dialkyl-dithiophosphate (ZDDP) additive as potential alternative lubricant. The addition of 5wt% ZDDP to RBD palm stearin showed a better friction-reducing performance than that of commercial mineral oil.

Tribological properties of palm oil-based TMP (trimethylolpropane) ester biolubricant added with TiO<sub>2</sub> nanoparticles used as additives (Zulkifli et al., 2013b) were evaluated. TMP ester added with nanoparticle reduced COF up to 15% at high load and also improved WSD especially at low load. Nanoparticle additives in lubricant improve tribological properties by formation of protective layer at the contacted interface. Tribological properties of the palm oil biolubricant modified with titanium oxide (TiO<sub>2</sub>) nanoparticles as additives were investigated (Shaari et al., 2015). Palm oil with titanium oxide (TiO<sub>2</sub>) nanoparticles produced the lowest coefficient of friction (COF) and wear scar diameter (WSD). Improvements in the anti-wear (AW) and extreme pressure (EP) ability of chemically modified palm oil (CMPO) were evaluated for conformal and non-conformal contact geometries (Gulzar et al., 2015). CMPO biolubricants synthesized with molybdenum disulfide (MoS<sub>2</sub>) nanoparticles exhibited better anti-wear (AW) and extreme pressure (EP) properties than copper (II) oxide (CuO) nanoparticles. The addition of oleic acid as surfactant aided nanoparticle suspension and also reduced wear. Gulzar et al. (2017) evaluated the dispersion stability and tribological characteristics of nano-TiO<sub>2</sub>/SiO<sub>2</sub> as an additive in a biobased lubricant. Nano-TiO<sub>2</sub>/SiO<sub>2</sub> exhibited appreciable dispersion at a concentration of 0.75 wt% in the absence of a surfactant. Stable dispersion was also effective in reducing friction and

wear compared to palm TMP ester. Palm TMP ester enriched with 0.75 wt% nano-TiO<sub>2</sub>/SiO<sub>2</sub> also exhibited improved tribological characteristics under EP conditions and piston ring-cylinder liner conjunction.

Kiu et al. (2017) investigated tribological properties of graphene nanosheets (GN), carbon nanotubes (CNT), graphene oxide (GO) as lubricant additives in vegetable oil (VO). Graphene nanosheets (GN) with 50ppm concentration addition in vegetable oil showed the lowest coefficient of friction and wear scar diameter compared to carbon nanotube (CNT) and graphene oxide (GO). Carbon based nanoparticles have gained much interest as lubricant additive due to their remarkable mechanical, chemical and electrical field properties. Talib et al (2017a) explored the lubrication properties of modified jatropha oils (MJOs) mixed with hexagonal boron nitride (hBN) nanoparticle additives. Modified jatropha oils (MJOs) were developed jatropha methyl ester and TMP. MJO5 with an addition of 0.05 wt% of hBN nanoparticles was found to reduce the friction and wear up to 76% and 24%, respectively in comparison with synthetic ester (SE). Hexagonal boron nitride (hBN) compounds generated tribochemical reactions under boundary lubrication and showed anti-friction and anti-wear properties. Amiril et al. (2018) evaluated corrosion inhibition behaviours, antifriction and antiwear abilities of fully oil-miscible phosphonium-based ionic liquid (PIL) and ammonium-based ionic liquid (AIL) additives in modified Jatropha oil (MJO) lubricant. MJO with 10 wt % of AIL and MJO with 1 wt % of PIL have shown superior friction reduction, lower worn surface area and excellent surface finish than neat base oil. Presence of polyol esters, fatty acids and polar structure of AIL and PIL in the biodegradable lubricants greatly reduce friction and wear.

Table 4: Research on biolubricants with additives in Malaysia.

<b>Biolubricants with additives</b>	<b>Authors</b>
A blend of Soybean oil, mineral oil, and additives suitable for use in sliding bearing is described	Ahmed et al. (2014)
Palm oil with a phenolic antioxidant provide improved lubrication at higher temperatures	Sapawe et al. (2016)
Performance improvement in lubrication of refined, bleached and deodorized (RBD) palm stearin with the addition of Zinc dialkyl-dithiophosphate (ZDDP) additive	Farhanah and Syahrullail (2016)
TiO <sub>2</sub> nanoparticles added to TMP ester exhibited good friction and wear reduction characteristics	Zulkifli et al. (2013b)
TiO <sub>2</sub> nanoparticle additives improve tribological properties of palm oil biolubricant	Shaari et al. (2015)
Chemically modified biolubricants exhibit excellent oxidation stability and low-temperature flow properties	Gulzar et al. (2015)
Stable dispersion of nano-additives is highly desirable for the effective lubrication performance of nanolubricants	Gulzar et al. (2017)
Vegetable oil (VO) with optimum concentration and most effective nanoparticle type (GN, CNT, GO) were investigated	Kiu et al. (2017)



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High formation of TMP triester in MJOs showed an improvement in tribological characteristics	Talib et al. (2017a)
MJO+AIL 10% and MJO + PIL 1% have great potential to be used as sustainable cutting fluids for metal working applications	Amiril et al. (2018)

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### 3.0 RESEARCH ON BIOLUBRICANTS IN JAPAN FOR TRIBOLOGICAL APPLICATIONS

Kimura et al. (1996) highlighted research activities on tribology in Japan and also presented a review on eco-tribology. The Japanese Society of Tribologists, Tribology Division of the Japan Society of Mechanical Engineers and the Machine Design continue to cooperate in promoting research activities in tribology. Takayanagi (2005) focussed on regulations and trends of biodegradability lubricating oils and future expectations. The environmentally friendly products are developed in various fields as the consciousness to environment increases. The biodegradability lubricating oils continue to compete with traditional petroleum-based oils in the marketplace. Based on the principles of green chemistry, Minami (2017) highlighted the significance biodegradable lubricants. Sasaki (2010) emphasized the challenging target announced by Japanese government in reducing greenhouse gas emissions to 25% by 2020 compared with 1990. The proportion of ecology in Ecotribology rose more than economy against the background of the rise of people's concerns to environmental problems. The role of an environmental performance has grown as an important factor deciding the product's value. Environmentally friendly tribology, eco-tribology, through surface modification technology (for example diamond-like carbon (DLC) coating), is seen to be an effective engineering technology that can contribute to sustainable societies. Diamond-like carbon (DLC) coatings under boundary lubricated conditions offer excellent mechanical and tribological properties and they provide ultra-low friction and superior wear resistance (Tasdemir et al., 2013). A combination of lubricant formulation and counterbody material is a crucial factor for the use of DLC under boundary lubricated conditions. Zinc dialkyldithiophosphate (ZnDTP) anti-wear additives form tribofilm on the surfaces depending on the DLC coating under boundary lubrication conditions (Tasdemir et al., 2014). Research on decreasing the detrimental impact of lubricants on the environment (Sasaki, 2010) now encompasses topics such as halogen-free and biodegradable oils, carbon-neutral vegetable oils, the minimum quantity of lubrication (MQL) and process fluid lubrication (such as water lubrication).

Advantages of biolubricants would be much more beneficial for lubricant industries if their tribological properties were improved by additive technologies (Minami et al., 2007). It is necessary not only to expect novel technological development but also to create realistic solutions by extending conventional technologies (Sasaki, 2010). Tribology has supported various technological developments over the years and has also responded promptly to societal demands for decreasing substances from engineering products that would be hazardous to the environment (Sasaki, 2010). As societies aim to become sustainable, tribology needs to be involved and contribute to the solution more than ever before (Sasaki, 2010).

#### 3.1 Biolubricants as basestock

Research works related to biolubricants in Japan are presented in Table 5. Sasahara et al. (1999) studied vegetable oil in oil-mist lubrication. A very small quantity of oil with compressed air to end mill is supplied from a nozzle in oil-mist lubrication system. Tool wear in oil-mist lubrication is small compared to machining without lubrication. Oil-mist supplying position

affects tool wear. Mia et al. (2007) investigated vegetable oils (olive oil, castor oil, mustard oil and coconut oil) in boundary lubrication. Various alternatives to petroleum based lubricants that are explored include synthetic lubricants and vegetable based lubricants. High-pressure tribological properties of vegetable oils that are important for the traction control and the prevention of surface failure under elastohydrodynamic lubrication were measured. Pressure-viscosity coefficient of vegetable oils showed lower values compared with mineral oil and hence can be used in EHL at moderate high-pressure. Viscosity-pressure-temperature relation and density-pressure-temperature that are important for performance of lubricating oil at high-pressure condition were derived from experiments. Vegetable oils typically contain about fatty acids, while mineral oil typically contains saturated aliphatic compounds, namely paraffinic and naphthenic, and small amount of aromatics. Mawatari et al. (2013) investigated first solidification characteristics of rapeseed oil, soybean oil, sunflower oil, palm oil, coconut oil, olive oil, camellia oil, mustard oil, castor oil, rapeseed methylester, and soybean methylester. Viscosity-pressure-temperature relation was derived for each vegetable oil. Ikejo et al. (2010) carried out a pitting test and a scoring test of spur gears lubricated with biodegradable oils. Biodegradable oils have higher pitting resistance than mineral hydraulic oil. Biodegradable oils have higher scoring resistance than the mineral hydraulic oil and the turbine oil.

Table 5: Research on biolubricants as basestock in Japan.

<b>Biolubricants as basestock</b>	<b>Authors</b>
Tool life becomes longest when oil-mist is supplied at the disengage point in milling	Sasahara et al. (1999)
Vegetable oils (olive oil, castor oil, mustard oil and coconut oil) are excellent boundary lubricants	Mia et al. (2007)
Vegetable oils as base oil for lubricants are usually excellent boundary lubricants	Mawatari et al. (2013)
Biodegradable oils effect on the tooth surface failure of spur gears were investigated	Ikejo et al. (2010)
Cutting fluids can reach the precut surface and involves lubrication effect	Kaneeda (2002)
In MQL turning, synthetic biodegradable esters are superior to a vegetable oil and provide the satisfactory cutting performance	Wakabayashi et al. (2003)
A multifunctional lubricant applicable to both machining and other lubricating parts of machine tools was developed	Suda et al. (2004)
MQL cutting demonstrates synthetic esters possess the preferable adsorption ability on to the freshly cut metal surfaces	Wakabayashi et al. (2006)
MQL machining produces similar cutting performance to conventional flood supply machining while using much less fluid	Min et al. (2005)
MJOs exhibit better machining performance in terms of tool wear, surface quality and chip formation compared to SE	Talib et al. (2015)

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MJOs with a high viscosity showed good lubrication properties and enhanced machining performances	Talib et al. (2017b)
Polar structures adhere to the metal surfaces and create a thin film that is effective in reducing friction and wear	Rahim and Sasahara (2017)

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Kaneeda (2002) investigated the extent of lubrication by cutting fluids using oil submerged cutting experiments. Machinability of ductile metal can be dramatically improved by applying oleic acid or extreme pressure oil on the pre-cut surface, due to a reduction in friction between the lamella of the chip. Lubrication by cutting fluids involves lubricant applying effect which can reduce cutting forces by more than 90%. Wakabayashi et al. (2003) investigated cutting performance of synthetic biodegradable esters in minimal quantity lubrication (MQL) machining. Tribological behavior of lubricants in MQL turning were evaluated with the aid of tool surface analysis and the adsorption characteristics. In order to develop multifunctional fluid, synthetic biodegradable polyol ester is combined with an exceptionally small amount of an organo-phosphorus compound which is highly effective extreme pressure additive (Suda et al., 2004). The developed multifunctional fluid has excellent performance in MQL machining, potential in spindle bearing lubrication and sufficient lubricating abilities for both hydraulic and slideway applications compared to commercial lubricants. Wakabayashi et al. (2006) evaluated multifunctional lubrication performance of synthetic polyol esters in minimal quantity lubrication (MQL) machining. Since some of those lubricants are often contaminated with cutting fluids and disposed without adequate separation treatments, it must certainly be convenient to prepare a multifunctional fluid, which is applicable to both machining and lubricating purposes. The adsorption ability is further enhanced by surrounding oxygen may result in effective lubricating film formation (Wakabayashi et al., 2006). Min et al. (2005) investigated adsorption characteristics of MQL media during orthogonal cutting. Adsorption of MQL media is closely related to lubrication performance in machining. An understanding of its tribological behavior and cutting performance of MQL machining is critical to take full advantage and expand its applicability. Oxygen in MQL supply plays a significant role in lubrication. Oxygen and ester reduced cutting forces and also produced better surface quality.

Talib et al. (2015) compared different compositions of refined modified jatropha oils (MJOs) with the crude jatropha oil (CJO) and synthetic ester (SE). MJO with high viscosity outperformed other MJOs during the machining process due to production of a thin film that can withstand at high temperature and reduces the tool wear. Talib et al. (2017b) evaluated machining performances of modified jatropha oil (MJO). Various types of TMP ester developed from jatropha oil are known as modified jatropha oil (MJO). The effectiveness of green solid lubricant namely hBN in MJO was analyzed at various concentrations. Crude jatropha oil (CJO) has been acknowledged as an environmentally benign metalworking fluid (MWF) in the machining industry. However, CJO has poor thermal-oxidative stability that leads to poor lubrication behavior. Biolubricants are primarily composed of triglycerides made up of esters from glycerol molecules with three long chain fatty acids and the polar structures of triglycerides is desirable in boundary lubrication. Moreover, triglycerides provided excellent lubricity, reduced cutting force and specific energy. MJO5 added with 0.05 wt% of hBN particles exhibited the optimum tribological behavior and excellent machining performances. The CJO was improvised through transesterification process at various molar ratios of jatropha methyl ester (JME) to trimethylolpropane (TMP) and through the addition of various concentrations of hexagonal

boron nitride (hBN) particles (Talib et al., 2017b). Rahim and Sasahara (2017) found that the biolubricant made from palm oil significantly improved the drilling performance under the MQL condition. Metalworking fluid acts as cooling and lubrication agent at the cutting zone in the machining process. Manufactures tends to substitute the mineral oil to bio-based oil such as vegetable and synthetic oil. Palm oil consists of high percentage of unsaturated fatty acid in the carbon chain which enhances the formation of high lubrication strength film on the tool-chip or tool-workpiece interface (Rahim and Sasahara, 2017).

### 3.2 Biolubricants in mixtures

Research works related to biolubricant mixtures in Japan are presented in Table 6. Ogura and Seki (1989) formulated rolling oil using a combination of ester and paraffinic mineral oil and this oil saved 14% of energy consumption compared to a conventional oil under standard operating conditions. It is observed that the formulated rolling oil can save energy consumed by a laboratory/production mill compared with conventional rolling oils. Better lubrication is obtained with structures with more paraffinic chains and less branched chains. Aromatic rings give poorer lubrication than paraffinic chains, and naphthenic rings result in the poorest lubrication. Hydrocarbons, paraffins showed the lowest coefficient of friction, while aromatics exhibited a higher coefficient of friction, with naphthenes showing the highest. Masjuki et al. (1999) carried out investigations on a palm and a mineral oil to evaluate wear, friction, viscosity, lubricant degradation and exhaust emissions. Viscosity index (VI) of palm oil based lubricant was higher than that of mineral oil based lubricant. Palm oil based lubricating oil exhibited better performance in terms of wear, and that the mineral oil based lubricating oil exhibited better performance in terms of friction. Muraki et al. (2007) studied tribological characteristics of biodegradable lubricants including rapeseed oil and two synthetic esters. Massive wear was brought about by oil-soluble aluminum soap formation due to the reaction between rapeseed oil and aluminum. Synthetic esters produced the boundary lubrication film, which promoted the formation of silicon oxide layer on the aluminum-silicon surface.

Table 6: Research on biolubricants in mixtures in Japan.

<b>Biolubricants in mixtures</b>	<b>Authors</b>
A high-quality oil was formulated by blending paraffinic mineral oil as base oil with ester as the main additive	Ogura and Seki (1989)
Palm and mineral oil based lubricants were formulated to reduce the wear and friction of the tribological component of engine	Masjuki et al. (1999)
Friction-and-wear characteristics of aluminum-silicon alloy were better using synthetic esters compared to rapeseed oil	Muraki et al. (2007)

### 3.3 Biolubricants with additives

Research works related to biolubricant additives in Japan are presented in Table 7. Tribological properties and mechanism of wear magnification of organic sulphides in esters were examined under boundary conditions (Minami et al., 2007). Organic sulphides magnify wear in unsaturated esters, while antioxidants prevent the magnified wear by organic sulphides, under boundary lubrication conditions. Peroxides, generated during the rubbing process, seem to play a significant role on magnified wear by organic sulphides in vegetable oils. The scoring-load capacities of the vegetable oils (Corn Oil, Soybean oil, Rapeseed oil) with anti-wear additive of

Dibutyl Phosphonate (DBPo) were studied (Nadano et al., 2005). The scoring-load capacities of the vegetable oils increased in following order: turbine oil < corn oil < soybean oil < rapeseed oil.

Table 7: Research on biolubricants with additives in Japan.

<b>Biolubricants with additives</b>	<b>Authors</b>
Boundary film of worn surfaces revealed presence of organic oxides, sulphides (unreacted additive) and sulphoxides	Minami et al. (2007)
Anti-wear additive of Dibutyl Phosphonate (DBPo) in vegetable oils increased the scoring-load capacities of vegetable oils	Nadano et al. (2005)
Proposed new additive technology towards environmentally acceptable lubricants based on molecular design	Nakayama et al. (2008)
Lubricant additives synthesized from cystine (Cys <sub>2</sub> ) reduced friction coefficient of PAOs and synthetic esters	Minami et al. (2010)
Zinc dioctyl dithiophosphate (ZDoDP) additive in palm oil reduces friction coefficient	Alias et al. (2017)

It is desirable to eliminate or reduce certain elements such as heavy metals, phosphorus and sulfur from lubricant compounds to develop environmentally acceptable lubricant systems. Nakayama et al. (2008) developed prototype additives free of both metals and phosphorus, and sulfur from anti-wear additives. The prototype additives possessed tribological properties comparable to conventional additives. A natural product was converted into different novel additives; then they were evaluated in a solution of synthetic fluids under boundary conditions (Nakayama et al., 2008). Novel environmentally adapted lubricant additives were synthesized from cystine (Cys<sub>2</sub>, with two carboxyl groups and two amino groups), an essential amino acid obtained from natural sources (Minami et al., 2010). The tribological properties of the additives in a solution of synthetic oil were evaluated under boundary lubrication conditions. The additives consist of hydrogen, carbon, nitrogen, oxygen, and sulfur; they are free of phosphorus, chlorine, and metals. The diesters of cystine also exhibited comparable antiwear properties to the conventional additive zinc dialkyldithiophosphate (ZnDTP). The disulfide improves tribological properties of boundary film, the polar functional groups is beneficial for the adsorption of molecules on metal surface and the straight hydrocarbon moiety improves friction reducing properties. The use of amino acids as versatile building blocks for the synthesis of environmentally adapted additives was discussed. Alias et al. (2017) observed that coefficient of friction reduces to 0.083 at 2 wt.% of zinc dioctyl dithiophosphate (ZnDoDP) as an additive in palm oil. ZDoDP additive contains phosphorus and sulphur that react with metallic surface, which creates a thin protective layer that acts as a cushion between the surface contacts and hence reduces friction and wear.

## CONCLUSION

Biolubricants have gained importance as alternatives to petroleum based lubricants due to global climate change. Increasing attention to environmental issues has driven the lubricant industry toward the use of biodegradable and environmentally accepted lubricants from renewable sources. Biolubricants from renewable sources have relatively low impact on global

environments. The use of biolubricants is influenced by their availability, renewability, environmentally friendly properties and government regulation on the use of mineral lubricants. The replacement of petroleum based lubricants with similar performance biolubricants derived from vegetable oils reduces dependence on nonrenewable resources, and increases markets for industrial applications.

Base biolubricants offer good lubricity, low volatility, high viscosity index and high flash point and low evaporative loss. Base biolubricants are available, renewable, biodegradable, sustainable, environmental friendly and non-toxic. Furthermore, biolubricants adhere to metal surfaces due to the existence of its polar ester group, and provides better lubrication characteristics. Replacements/mixtures of base biolubricants with mineral oils reduce dependence on nonrenewable oil resources. Focus is on research and development of additives for base biolubricants to reduce wear and friction in the tribological systems. Biolubricants have poor cold flow properties as well as low oxidation, and these shortcomings can be addressed by modifying the biolubricants chemically or incorporating additives. Biolubricants with stable dispersion of formulated additives is highly desirable for enhanced tribological performance.

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