



SUSTAINABILITY WITHOUT SACRIFICING PERFORMANCE: WHERE BIO-BASED GREASES ACTUALLY WORK

To reduce water and soil pollution, bio-greases have resurfaced as an alternative to petroleum derived lubricants. Although plant and animal derived lubricants have been used for several thousand years, petroleum products have dominated the market in the past century. This review evaluates different formulations of bio-based greases and the applications in which they are most effective. Bio-greases can be derived from a variety of feedstocks, including edible and nonedible plants, algae, and recycled waste oils. The use of innovative biodegradable additives and chemical modifications enable bio-greases to match and even exceed the performance of conventional greases. However, bio-greases rarely meet performance standards without altering their physiochemical properties, necessitating the innovation of environmentally benign and cost-effective additives and modifications. In addition to their biodegradability and the growing use of non-toxic additives, bio-greases also offer reduced carbon dioxide emissions throughout their lifecycles, producing about 22 fewer kilograms of carbon dioxide per unit of grease compared to an equivalent quantity of conventional lubricant.

Introduction

As petroleum resources become increasingly scarce, and more expensive, engineers have been challenged with reintroducing bio-based materials to the market. Greases reduce friction and wear between moving components like metal joints, shafts, and bearings. Greases are important for reducing energy consumption and carbon dioxide emissions because improved lubrication prolongs the service life of machinery [1]. Mineral oil-based greases currently make up nearly 90% of global demand for lubricating greases [2]. Bio-greases were used before fossil-derived products grew to dominate the market for oils, greases, and more. Animal fats, olive oil, and other bio-based materials were used for lubricating greases until the latter half of the 19th century when petroleum-based materials overtook the market [3]. While bio-greases only make up 1-5% of the market for lubricating greases today [3], they are regaining their popularity due to increasing concern about the environmental disadvantages of petroleum-based products.

Lubricating greases are a solid/semi-fluid mixture of fluid lubricant, thickener, and additives (Figure 1). Fluid lubricants are the main component and are responsible for the actual lubrication [1]. The fluid lubricant can be petroleum (mineral) oil, synthetic oil, or vegetable oil. Greases contain up to 80% of lubricating fluid, or base oil [1]. The base oil influences the pumping and flowability of the grease. Most commercially available greases are composed of mineral oil blended with a soap thickener. The thickener forms a semi-fluid structure when combined with base oils and additives, holding the lubricant until it is dispersed [1]. The two main classifications of greases are those with soap and non-soap-based thickeners. More than 90%

of greases globally contain soap-based thickeners [1]. Finally, additives have three main functions. Some additives are used to improve intrinsic characteristics of the base oil, like viscosity and pour point [1]. Antioxidants are lubricant protective substances. Lastly, some additives give new properties and protect metal surfaces. This last category includes detergents, dispersants, friction modifiers, anti-wear/extreme pressure additives, and rust and corrosion inhibitors [1]. These additives are crucial to ensuring the grease will perform sufficiently in any given application.

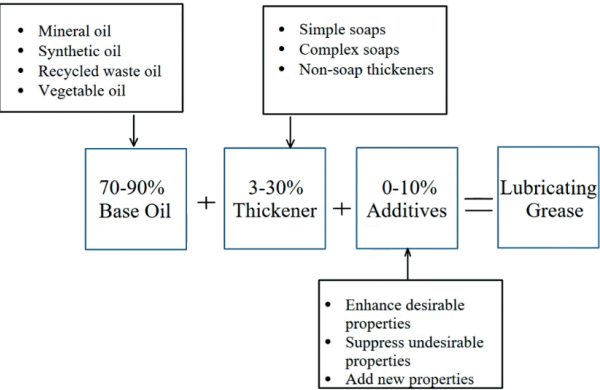


Figure 1. Composition of lubricating greases and the function of each component [1].

Bio-greases are a valuable alternative to conventional lubricating greases because their contents are environmentally benign and their feedstocks more abundant than petroleum. Instead of petroleum, bio-lubricants are derived from biomass like oilseeds,

animal fats, oily residues, and microalgae. Approximately 50% of conventional lubricants end up in the environment [1]. The biodegradable nature of bio-lubricants mitigates the risks associated with the release of lubricating greases into the environment. Bio-oil production begins with pyrolysis, which is the heating of an organic material in the absence of oxygen. Biomass pyrolysis is typically conducted at or above 500 °C to sufficiently deconstruct strong biopolymers [1]. Instead of combusting, the biomass thermally decomposes into combustible gases and biochar. Most of these gases can be condensed into bio-oil, a combustible liquid. The remaining permanent gases form syngas, which can be combusted to provide heat for pyrolysis [3]. The resulting liquid product is a fully bio-based oil.

There are three main base oils for bio-lubricants: bio-esters derived from plant oils, polyalphaolefins, and recycled waste oils. Edible oils have dominated the landscape for base oil feedstocks, but their competition with the agriculture industry has catalyzed a shift towards non-edible feedstocks. Vegetable oil-based products are more price competitive than biodegradable synthetic esters, but they often do not perform sufficiently without modification [4]. Algal oils are an attractive alternative due to their high lipid productivity and the possibility of growing algae in non-arable land with wastewater streams [5]. Polyalphaolefins, or PAOs, can be synthesized from renewable olefin feedstocks, plant-derived alpha olefins, and aim to replicate group IV synthetic hydrocarbon characteristics with a smaller carbon footprint [6]. Waste cooking oil has also grown in popularity as a feedstock, and studies have demonstrated that waste-derived esters can achieve a tribological performance comparable to virgin oils when appropriately modified with additives [5]. While all three categories have exhibited sufficient tribological behavior, bio-greases remain a small fraction of all lubricating greases consumed, in part due to their high cost. Bio-greases can be produced more economically if the consumption of bio-greases increases [4]. It is therefore critical to develop environmentally clean, cost-effective solutions for widespread adoption of bio-greases.

Characteristics of Bio-greases

Bio-esters

Fats are esters with saturated or nearly saturated hydrocarbon chains, which are solid at room temperature. Oils are also esters but have a degree of unsaturation, making them liquid at room temperature. Bio-esters are esters derived from vegetable oils or chemically modified triglycerides (complex esters, estolides) (Figure 2). Vegetable oils are a first-generation feedstock for

bio-esters, and they are currently the most common bio-ester feedstock globally. Common base oil feedstocks in the US include soybeans, canola, rapeseeds, corn, and castor beans [3]. Similarly, palm oil is dominant in Asia, while rapeseed and canola oil are popular in Europe [7]. However, there is global interest in non-edible feedstocks, including non-edible oilseeds, like Karanja in India, and algae. The triglycerides in bio-esters are responsible for the discrepancies in the properties of bio-esters and mineral oils [2]. Bio-esters often have high viscosity indices and form robust films that decrease friction and wear [5]. Esters are polar, which improves adhesion to metal surfaces and enhances initial water washout resistance compared to non-polar oils [8]. Esters may also improve thickener polarity and soap network formation, sometimes requiring less soap than their mineral oil counterparts to achieve equivalent consistency [5]. Castor oil is currently a popular base oil because of its high viscosity, excellent polarity, and naturally occurring hydroxyl group that contributes to its oxidative stability and superior film-forming capability [7]. Bio-esters differ most significantly from mineral oils in their higher viscosity indices and stronger interactions with metal surfaces due to their triglyceride structures.



Figure 2. Chemical structure of a generic ester (left), where R and R' are hydrocarbon chains, and a triglyceride fat (right) with two stearic acid chains on the top and middle and one oleic acid chain on the bottom [3].

The triglyceride profile of the feedstock for bio-esters can dictate the properties of the resulting base oil. A study by T. Panchal, D. Chauhan, M. Thomas, and J. Patel demonstrated that as the chain length of a Karanja oil ester increased, it was more likely to disperse within the lithium soap and form a dense matrix [2]. Among three variations of the Karanja oil ester, the oil with the longest hydrocarbon chain length outperformed the other two in weld capacity and shear stability [2]. This suggests that esters with longer hydrocarbon chains have superior anti-wear and anti-friction properties than those with shorter chains because they can better withstand shear stress. Despite the advantages in anti-wear and anti-friction offered by bio-esters, they can be chemically unstable.

Esters are vulnerable to chemical instability at high temperatures. The double bonds in unsaturated fatty acids are prone to peroxidation, which leads to rapid degradation at high temperatures [9]. Therefore, esters oxidize more readily than saturated hydrocarbons because esters and any unsaturation in the fatty acid chains can react with oxygen, especially at high temperatures [10]. Monounsaturated fatty acids have moderate lubricity and oxidative resistance, while polyunsaturated fatty acids have superior fluidity but are more vulnerable to oxidation [5]. In addition to providing superior oxidative stability, greater hydrogen content also improves the cold flow properties of a bio-ester compared to its unsaturated counterpart [3]. Additionally, esters have weak hydrostatic stability, and ester bonds break down in the presence of heat and water. Water hydrolyzes ester bonds, producing acids and alcohols, increasing acidity and degrading the grease [11]. Bio-ester greases can initially resist water better than very nonpolar oils, but long-term water exposure at elevated temperatures can degrade them. Lastly, some esters interact with seal materials (swelling/softening), necessitating careful elastomer selection or additives [10]. Generally, additives are necessary to overcome these challenges.

Renewable PAOs

A PAO molecule has a branched, saturated structure that links multiple alpha-olefin monomers (Figure 4). PAOs have low pour points and good cold start properties, so they are advantageous in cold climates [12]. Their narrow molecular weight distribution provides uniform rheology and minimal shear thinning, so PAOs offer excellent shear stability [12]. Some PAOs do not match thermal oxidative resilience of fully synthetic PAOs [6]. PAOs have inherently low solvency, which can reduce additive effectiveness unless they are blended with more polar fluids like esters [13]. Very low pour points and high viscosity indices give

PAOs good cold start fluidity [6]. Like bio-esters, renewable PAOs can approach or match the oxidative stability of conventional synthetics with additives, but they are expensive and harder to scale than petrochemical-derived PAOs because raw materials are limited [6]. However, renewable PAOs are not the dominant production route compared to petrochemical PAOs. Despite the barriers to producing environmentally advantageous PAO-based greases, their production is important because they often outperform bio-esters in thermo-oxidative stability and water resistance.

A PAO molecule has no double bonds, creating a stable oil with excellent thermal and oxidative stability and good low temperature performance. PAOs are highly saturated, giving them excellent intrinsic oxidation resistance [13]. PAO-based greases tend to have stronger long-term oxidation stability than typical ester-rich bio-greases, especially above 100 °C. Additionally, PAOs are hydrolytically stable because they lack ester functionalities [14]. PAOs are inherently nonpolar and less adhesive, reducing water washout resistance unless polar additives or thickeners are used. Likewise, PAOs are chemically water stable, but they need formulation work to withstand high moisture. These properties are advantageous in high-temperature applications. Synthetic polymer base oils are often used in low temperature applications [15], but renewable PAOs often fail to meet this requirement without modification.

Waste oil feedstocks

Reprocessed waste oils or re-refined stocks are a viable sustainable feedstock for bio-greases after decontamination and chemical modification. Waste oils can come from used cooking oil and animal tallow. The use of waste oil diverts streams from disposal and can make grease base stocks with comparable viscosity and friction/wear performance to traditional feedstocks after proper processing [16]. Waste oil feed stocks offer lower raw material cost compared to virgin synthetics or bio-feedstocks due to reusing waste, which makes them an appealing environmentally conscious choice compared to bio-esters and renewable PAOs [16]. Some greases from treated waste oils have oil bleed, separation, and wear performance like commercial greases [17]. The quality of recycled base stocks depends on the contaminants in the source, additive remnants, and residual breakdown products, all of which can impact oxidative stability and consistency. Proper processing of the recycled stock is therefore critical in producing a grease that performs predictably.

Properly refined recycled stocks can have good baseline performance and impressive sustainability value, but they often require formulator input, like additives and/or thickener tuning, to reach high-performance grease standards. While waste oil feedstocks are economically attractive, they require significant processing. Some recycled oils can match oxidation performance of conventional base oils after refining, which may be costly [17]. The oxidative stability of the resulting oil depends on the feedstock and purification process. Any residual surfactants, degraded additives, or oxidation products not removed in processing may reduce water tolerance. Dewaxing and additives are often necessary for waste oil feedstocks to match the cold flow properties of PAOs and bio-esters [17]. In addition to needing additives and modifications like bio-esters and renewable PAOs, greases derived from recycled waste also require the treatment and removal of existing contaminants.

A Comparison of Bio-grease Feedstocks: Bio-esters, PAOs, and Waste

Bio-esters derived from vegetable oils are currently most abundant. Vegetable oils are characterized as non-flammable liquids with good lubricity and viscosity indices as well as a superior film strength and affinity to metal surfaces [2]. Many bio-esters have excellent low-temp characteristics, often better than mineral oil [18]. However, vegetable oils lack thermo-oxidative and hydrolytic stability compared to mineral oils, limiting their potential applications [2]. Vegetable triglycerides degrade quicker under high heat compared to PAOs or mineral oil, so they need antioxidant additives or chemical modification to be more stable [5]. Bio-esters have excellent lubricity, high viscosity indices, and advantageous environmental compatibility.

Their boundaries include thermal and oxidation stability and cold flow, which can be mitigated by functionalization, such as epoxidation or synthetic esters, or additives. Triglyceride oils are relatively inexpensive but require additives to prevent oxidation and hydrolyzation. Unless chemically modified, esters often have lower intrinsic thermo-oxidative stability than PAOs [19]. Unmodified natural oils, or triglyceride esters, may crystallize or gel at low temperatures, requiring chemical modification to reduce their pour point. thermal degradation. Overall, bio-esters are advantageous in eco-sensitive applications but require antioxidants for high temperatures.

Renewable PAOs are good for cold flow, shear stability, and predictable rheology. Their performance ceiling is in extreme heat or extended service, as formulation synergy with ester co-solvents and antioxidants are often necessary to enhance the function of PAOs. PAOs excel in cold-start and shear stability but fall behind conventional synthetics under long, hot, heavy loads without synergistic blends.

Waste oil feedstocks are economically attractive because they utilize recycled waste stocks. However, their rheological properties and thermo-oxidative stability are dependent on the source of waste oil and proper processing. Therefore, it is difficult to characterize the properties of waste oil feedstocks. The strengths and limitations of bio-esters, renewable PAOs, and waste oils are summarized in Table 1.

Table 1: Strengths and performance limits of bio-esters, renewable PAOs, and waste oil.

Base Oil	Strengths	Performance Limits
Bio-esters	High lubricity, biodegradable, high viscosity index [2]	Thermal/oxidation limits without modification; cold flow can be an issue [2]
Renewable PAOs	Cold flow, shear/stability, predictable rheology [12]	Cost; extreme heat vs. advanced synthetics [6]
Waste oil	Sustainability, cost [15]	Quality control; extreme condition limits [15]

Chemical Modifications to Vegetable Oils

Vegetable oils are a biodegradable raw material for lubricants but have lower oxidation stability and higher pour point compared to mineral derived base oils. Chemical modifications can overcome these drawbacks, and modified base fluids can approach synthetic performance [18]. Esterification, transesterification, epoxidation, and stolid have all been investigated. Transesterification, followed by esterification, is the leading chemical modification because it offers lower cost and energy consumption and better efficiency than the alternatives [5, 9]. These modifications aim to change the properties of the oil by altering the chemical structure of the oil.

Fatty acid chains and functional groups are often the targets of chemical modifications. Altering fatty acid chains and functional groups of vegetable oils can enhance the viscosity index, oxidative stability, and cold-flow behavior of the oil to meet the needs of industrial applications [5]. Transesterification reacts triglycerides with alcohols in the presence of an acid, base, or catalyst. Transesterification with polyols or alcohols reduces viscosity and enhances lubricity [5]. Esterification of fatty acids produces synthetic esters with high viscosity indices, low volatility, and improved biodegradability [9]. Both reactions produce base stocks with better cold flow compared to the unmodified oil without sacrificing the inherent lubricity of the unmodified oil.

Epoxidation introduces oxirane rings into unsaturated fatty acids, creating reactive sites that can be further modified through ring-opening to produce polyols or estolides. Epoxidation offers enhanced oxidative stability and high temperature resistance, but epoxides are susceptible to low-temperature crystallization [5]. Ring-opening reactions with organic acids produce estolides with superior cold-flow and anti-wear characteristics [5]. Epoxidized derivatives combined with synthetic esters have demonstrated a

promising solution to decreasing the pour point of vegetable oils [5]. Epoxidized oils demonstrate improved oxidative stability and reduced volatility, but they still require further stabilization with antioxidants for long-term durability. For example, recent studies report that epoxidized palm oil estolides blended with zinc oxide nanoparticles performed comparably to PAOs [5]. In addition to transesterification and esterification, there are other less common methods of chemical modifications used to alter the structures of vegetable oils.

Other chemical modifications include enzymatic modification, hydrogenation, and hydroisomerization. Enzymatic modification, especially lipase-catalyzed esterification, enables selective conversion and minimizes by-product formation. This option may help to decrease the overall carbon footprint of bio-grease production [5]. Hydrogenation reduces unsaturation by converting double bonds into saturated bonds, improving the oxidative stability of the oil. Hydrogenated oils have enhanced thermal and oxidative resistance but have a suboptimal pour point. Hydroisomerization can overcome this drawback by altering the carbon chain to improve cold-flow behavior without compromising the oxidative stability [5]. Catalytic hydrogenation along with selective isomerization produces high performance synthetic based oils with balanced viscosity-temperature properties [9]. These options, however, are less cost effective than transesterification and esterification.

Modified vegetable oils are entering the commercial lubricant markets, particularly in hydraulic fluids, metalworking fluids, and gear oils (Table 2). For example, Trimethylolpropane esters are used in aviation turbine oils because of their high flash points and low volatility [5]. Chemical modification is a vital step in bio-grease formation because it generates bio-greases that perform well in industrial applications.

Table 2: Chemical modifications of base oils [5]

Modification	Property Targeted	Industrial Application
Transesterification	Cold-flow, biodegradability	Hydraulic fluids
Epoxidation	Oxidative stability, polarity	Gear oils
Hydrogenation	Thermo-oxidative resistance	Turbine oils
Estolide Formation	Anti-wear, film strength	Engine oils

Thickeners: Soap-based and Polyester

Soap-based thickeners are formed by reacting fatty acids and metal oxides. Thickener formation must be modified for bio-grease production. Conventional grease manufacturing processes use temperatures as high as 220 C, which may oxidize biodegradable base oils [3]. Soaps are formed in situ by adding alkali to a mixture of fatty acids and base oils. Mineral oils are nonpolar and do not react with the alkaline solution, but bio-based oils are esters formed from the same or similar fatty acids as those forming the soap [3]. These fatty acids can react to form a complicated soap matrix that performs unpredictably extreme temperatures, but microwave-based processing can be used to make a more uniform product [3]. In addition to the reactions between fatty acids, the metal used in soap formation can affect the properties of a thickener.

Common metals used for grease thickeners include lithium, calcium, potassium, and sodium. The thickener used can influence the properties of the grease. Lithium soaps are known to possess superior washout resistance and excellent high temperature performance, and their performance and longevity are often worth their high cost [9]. While lithium, calcium, potassium, and sodium soaps all offer their respective advantages, complex soaps are used for high-temperature applications [3]. Soap thickeners have been shown to impact the drop point of a grease. The drop point of grease is the temperature at which the thickener is no longer able to maintain the base oil within the thickener matrix. This may be due to the thickener melting or the oil becoming so thin that the surface tension and capillary action are insufficient to hold the oil in the thickener matrix [2]. The drop point of the lubricating

grease is directly proportional to the concentration of thickener. Sodium soaps often have a lower drop point compared to lithium soaps [9]. Increasing thickener from 10% to 30% increased the drop point of sunflower oil, soybean oil, and rapeseed oil by about 20%, 26%, and 6%, respectively [9]. The type of base oil has a significant effect on the drop point of bio-grease, and the drop point of bio-grease can be increased by using a more viscous base oil [9]. Understanding the impact of thickener concentration on drop point is critical for achieving effective high-temperature performance. However, there is interest in developing bio-based thickener systems for a fully biodegradable product.

In addition to soaps, bio-based polyester thickener systems show promising tribological performance. Thickeners with polymeric structures can be beneficial compared to thickeners with low molecular weight structures, so polymeric thickener systems are common in industrial greases [20]. Thickeners with the urea group functions are widely used in industry, but these structures are not biodegradable because urea itself is a degradation product. Urea can, however, be excreted by organisms and reconverted in further biochemical processes, so it does not pose an environmental threat [20]. Thickeners containing urea use modified urea derivatives that lose their solubility in water, preventing them from being converted in biochemical processes and creating persistent substances in the environment. Using ester functional groups in polymeric thickener systems is a viable alternative to develop a fully biodegradable grease [20].

S. Vafaei et al. investigated bio-based greases with nine different polyester-based thickener systems and castor oil base oil. The resulting bio-greases were compared to a petrochemical urea-grease and a bio-grease with polyurea thickener. Three thickener systems were deemed viable alternatives to polyurea-based greases: dodecanedioic acid - 1,4-butanediol (DDS-BD), dodecanedioic acid - 1,3-propanediol (DDS-PrD), and succinic acid - 1,4-butanediol (BS-DS) [20]. DDS-BD exhibited a smaller film thickness than the reference grease, and this discrepancy increased at higher rolling speeds [21]. DDS-PrD demonstrated a film thickness that increased with rolling speed [20]. Its film thickness is comparable to the reference grease at low speeds and outperforms the reference grease at high speeds. Lastly, BS-BD had a film thickness comparable to the reference grease at all tested rolling speeds [20]. Polyester thickeners have lower friction coefficients than the reference, but these coefficients increase more sharply at higher rolling speeds than the reference. DDS-PrD exhibited the lowest coefficient of friction at low rolling speeds compared to DDS-BD and BS-BD, while DDS-BD displayed the lowest coefficient of friction at high rolling speeds [20]. Decreased film thickness in the contact area risks solid contact of metal components. The bio-based and biodegradable polyester greases show suitable tribological properties, but their anti-wear performance and thermal stability should be further investigated before use in industrial applications.

Additives to Enhance Bio-grease Performance

Additives are frequently used in lubricating greases to improve their performance. Antioxidants interrupt radical chain reactions to enhance oil stability under thermal stress, pour point depressants can minimize crystal growth, and viscosity index improvers ensure stable viscosity across a wide temperature range [3]. Although they are crucial for the proper function of greases, conventional additives can also be environmentally toxic. For example, greases for high-pressure applications often include zinc or sulfur additives. Bismuth-additives may be a more benign alternative to heavy metals like antimony, molybdenum, and lead. Although bismuth is the heaviest of these elements, it is less toxic than lead because it does not bioaccumulate [3]. Natural antioxidants, like tocopherols and lignin derivatives, have been explored as alternatives to traditional additives, such as phenolic and aminic compounds [5]. In addition to these swaps, new additives have been developed to offer improved environmental compatibility. Some of these innovations include clays, nanoparticles, ionic liquids, and gums.

Illite

Clay materials, especially layered silicates, are of interest as nontoxic additives because of their high surface area, lamellar structure, and chemical stability. Kaolin, sepiolite, and silicates

are naturally occurring additives that have been used successfully as a lubricating additive [21]. More recently, illite has been investigated as a superior clay additive. Illite is a naturally occurring non-expanding clay mineral composed of stacked layers (Figure 3). The weak interlayer bonding in illite allows for shearing between layers, making illite a possible solid lubricant [10]. Illite offers good thermal stability and low reactivity and is a highly abundant resource, making illite a cost effective and sustainable material. Studies demonstrated that 0.1 weight% illite reduces friction and wear by 53% and 57%, respectively, compared to calcium grease with no added illite [21]. Illite improved the thermal stability of calcium grease but promoted oxidative degradation.

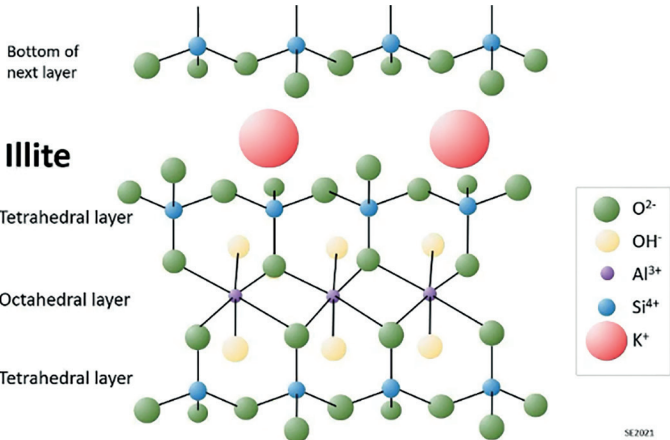


Figure 3. Composition of illite [22].

Nanoparticles and Ionic Liquids

Nanoparticles and ionic liquids have been investigated to improve the anti-friction and anti-wear properties of greases. Nanoparticles form protective films, reduce asperity contact, and improve load-carrying capacity, which ultimately reduces friction and wear. J. R. Patel et al. compare the reduction in friction and wear offered by CuO, ZnO, TiO₂, and graphene nanoparticle additives [5]. Graphene nanoparticles demonstrated the best results, reducing the coefficient of friction by 40% (Figure 4). The least effective nanoparticle, CuO, still offered a promising 25% reduction in friction (Figure 4).

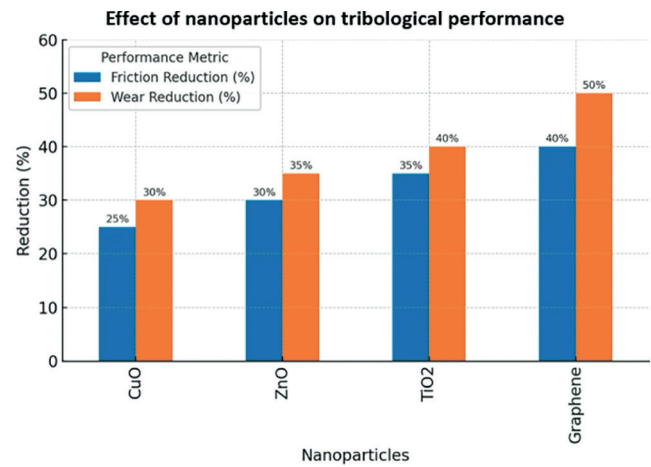


Figure 4. Reduction in friction (blue) and wear (orange) caused by adding CuO, ZnO, TiO₂, and graphene nanoparticles to lubricating grease [5].

Ionic liquids improve friction and wear while enhancing oxidative resistance. However, some ionic liquids can be costly and toxic, so they have yet to be adopted on a large-scale [5]. Synthetic esters and ionic liquid blends have lower coefficients of friction than mineral oils, which make these alternatives more energy efficient (Figure 5). This reduces greenhouse gas emissions and extends equipment life.

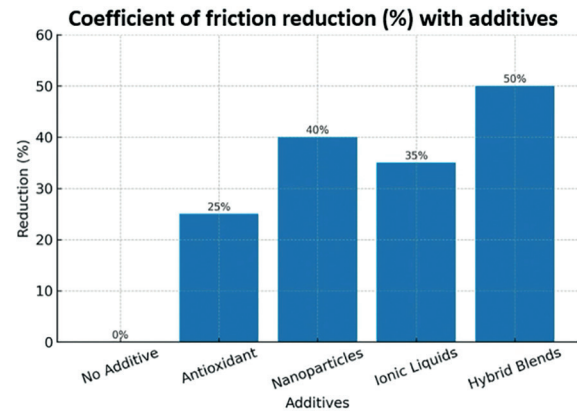


Figure 5. Reduction in friction observed when using various additives [5].

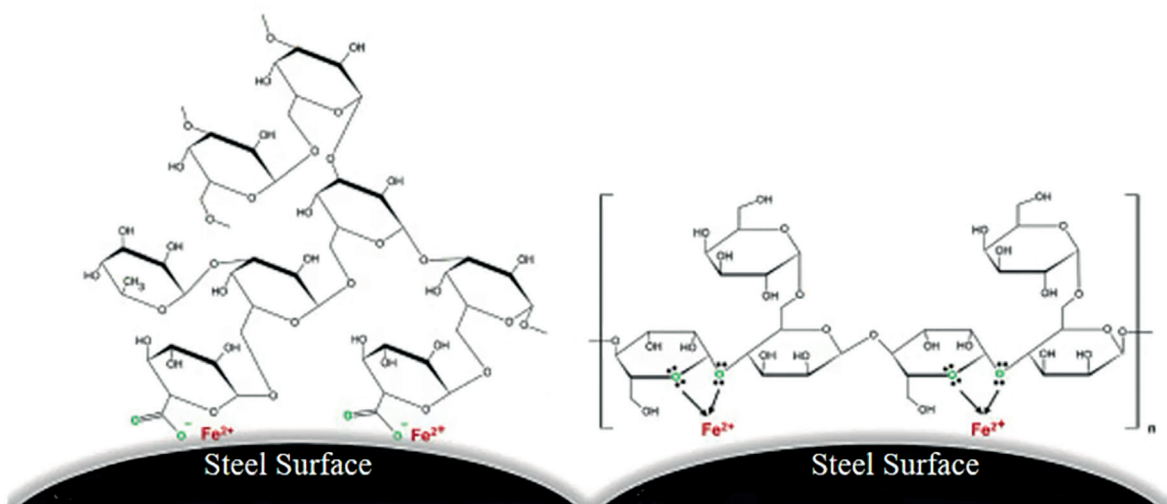


Figure 6. Polymer-layered silicate nanocomposite tribofilm at the lubricant/steel interface formed by the chemisorption of GA (left) or physisorption of GG (right) [23].

Polymers

Polymers are promising lubricant additives, and biopolymers may offer similar advantages as well as biodegradability. Polysaccharide gums like gum acacia (GA) and guar gum (GG) additives can enhance extreme-pressure (EP) performance of bio-greases sourced from vegetable oil and organoclay [23]. Gums have several reactive functional groups that facilitate easy anchoring over metallic surfaces to form protective films. Gums form a polymer-layered silicate nanocomposite tribofilm at the interface, via chemisorption for GA or physisorption for GC, which allows for superior performance (Figure 6). The polar functional groups on the gum interact with metal hydroxide species on the metal surface. For GA, chemisorption occurs through an acid-base interaction between the carboxyl group of the gum and metal hydroxide surface. For GG, physisorption is driven by relatively weak, non-covalent forces between the oxygen-rich functional groups of the gum and the metal surface. These non-covalent forces are primarily hydrogen bonds, electrostatic forces, and Van der Waals forces.

Under heavy loads and high temperatures, the ideal lubricating film separating the contact surfaces must be extremely thin. This film eventually breaks in some spots, leading to surface damage. EP additives enable the rapid formation of a high-melting and easy-to-shear protective film over these vulnerable spots [23]. In a recent study, weld loads (WLs) and pre-weld loads (PWLs) of greases with GA and GG additives were recorded, revealing that both gums have promising potential as EP additives [23]. GA displays better performance enhancement than GG since GG only acts as an effective EP additive at sufficiently high concentrations [23]. Overall, polysaccharide gums are a promising step towards developing bio-based greases with excellent tribological performance.

Applications of Bio-greases

Biodegradable, low toxic greases are good in industrial applications where greases are intentionally purged or replaced frequently. Bio-greases can reduce remediation costs in the event of lubricant spills for municipal and public works applications. Biodegradability and low ecotoxicity are regulatory requirements or operational priorities in these applications [24]. Bio-greases are used in general purpose bearings, medium-speed motors, and light gearboxes and articulations because they perform well when loads, temps, and speeds are moderate [25]. Germany has successfully employed ester-based hydraulic oils in railway switches, and Indian railways have successfully adopted biodegradable greases [5].

Hybrid greases are used in many industrial, transportation, or high-performance settings. Greases used for these high-performance applications often combine bio-based stocks with synthetic PAOs or synthetic esters because pure bio-greases often cannot withstand temperature variations and heavy loads as well as hybrids due to their inferior oxidative stability [25]. Precision machinery, like spindle bearings and robotics, require consistent film strength and low torque across a wide temperature range, which is best achieved by PAO or modified synthetic-bio ester blends rather than unmodified bio-oils [26].

Bio-based greases may outperform mineral-based greases for applications with extreme conditions. Bio-greases may be able to better withstand severe operating conditions and contaminants like dirt, sludge, and water [3]. Hybrid blends of bio-based and synthetic lubricants offer good cost performance, a wide temperature operating range, and good thermo-oxidative stability.

The ideal greases for varying applications and conditions are summarized in Table 3. PAO-dominant base oils offer superior oxidation resistance and stability, but they often need polar modifiers for water resistance, making PAOs ideal for high temperature and long-life applications. Bio-esters can provide good low temperature performance and initial water resistance but require chemical and hydrolytic stability enhancements for high heat and water exposure, so bio-esters are best used in eco-sensitive applications and moderate service conditions. Recycled base stocks are viable if refining additives bring them close to the performance of virgin oils, and they are best used when cost or sustainability are priorities over performance.

Table 3: Ideal lubricating grease for a given application.

Application	Best Lubricant Strategy
Environmental compliance (marine, rail, parks)	Bio-grease (renewable esters/EALs) [23]
Food processing + incidental contact	Bio-grease [27]
General industrial moderate conditions	Bio-grease or hybrid blend [25]
High-load, high-temperature, precision or extended life	Hybrid blend or PAO [25]
Extreme cold starts & mobility	PAO [17]

Regulatory Pressure vs. True Lifecycle Impact

A bio-lubricant is any lubricant that biodegrades expediently and is nontoxic for both humans and the environment [10]. There are two categories of bio-lubricants. Environmentally acceptable lubricants (EALs) meet standards for biodegradability, toxicity, and bioaccumulation. Environmentally friendly lubricants (EFLs) are hypothesized to be environmentally benign but are not proven to meet the same standards as EALs. Solid and semi-solid lubricants are not easily liquified and therefore not easily recycled, so biodegradability is a promising alternative to recycling [3]. Biobased lubricants can have substantially lower global warming potential than mineral-derived equivalents [27].

Regulators mandate biodegradability and aquatic safety for EALs, non-bioaccumulative formulations, and biobased content measures. These regulations focus on the end-of-life behavior of the substance. For example, the EPA requires that there is no visible oil sheen downstream from facilities located in or near waterways [3]. Additionally, waterways must contain less than 10 ppm mineral-based oil. It is nearly impossible to eliminate spills and leakage, so environmentally benign materials are a prudent replacement for mineral-based greases [3]. The full carbon

footprint, agricultural impacts of feedstock production, and end-to-end environmental costs of additives are not widely regulated yet. EAL mandates reduce the acute ecological risks associated with lubricant spills, but they do not guarantee that a bio-grease is the lowest lifecycle environmental product available. Current regulations are most effective in marine discharge and forestry, where greases and oils may directly enter the water or soil. These regulations are less effective for mitigating the upstream environmental impacts of lubricating greases because they do not address the carbon emissions from production, impacts of feedstock cultivation, and additive toxicity [28]. Additive, and potentially contaminant, toxicity is important when considering the lifecycle benefits of greases. In addition to harmful additives, greases can acquire metal particulate contaminants during use, necessitating proper disposal [3]. The lifecycle benefits of bio-greases vary widely depending on the feedstock and additives. Bio-based lubricants offer less carbon dioxide emissions in production, use, and disposal [5]. Mineral oil-based lubricants emit about 22 more kilograms of carbon dioxide, compared to an equivalent quantity of bio-based lubricants, over their lifecycle (Figure 7). Overall, bio-based lubricants have a smaller carbon footprint than mineral oils.

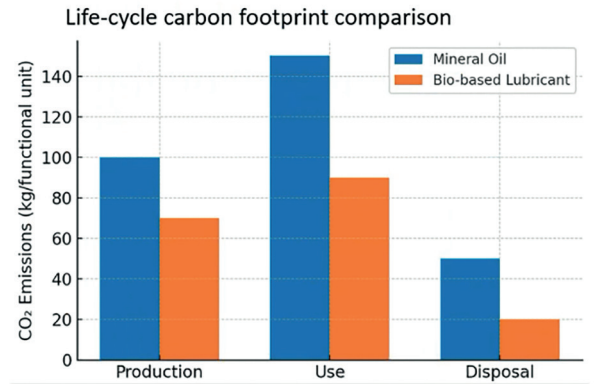


Figure 7. Carbon dioxide emissions (kg/functional unit) for lubricant production, use, and disposal for mineral oils (blue) and bio-based lubricants (orange), respectively [5].

Conclusion

Bio-greases are a prudent replacement for petroleum derived lubricating greases because petroleum resources are scarcer and more expensive than ever. Additionally, biodegradable and environmentally benign alternatives to conventional greases are a crucial step away from polluting our water and soil resources. Bio-greases can be derived from edible and nonedible seed oils, PAOs, and recycled waste oils. While vegetable oils are currently the dominant feedstock, there is a global focus on shifting away from edible feedstocks to avoid competition with the agricultural industry. Although greases from all three feedstocks can offer challenges in thermo-oxidative stability, shear stability, and cold flow properties, each feedstock also presents its own advantage. With appropriate additives and chemical modifications, bio-greases can meet or even exceed the performance of conventional greases. Environmentally benign additives and thickeners have been shown to effectively enhance the performance of bio-greases. Bio-greases are viable replacements for petrochemical lubricating greases. Bio-esters, especially those from vegetable oil feedstocks, are the most widely understood and most popular source of bio-lubricants today. Research on less understood, and potentially more costly, sources is crucial because of the performance and environmental advantages offered by renewable PAOs and recycled waste oil, respectively, compared to bio-esters. Finally, it is imperative that regulations weigh the lifecycle impact of bio-greases and incentivize the adoption of bio-based alternatives to petrochemical-derived lubricating greases.

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