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A comprehensive review of sustainable approaches for synthetic lubricant components

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ABSTRACT

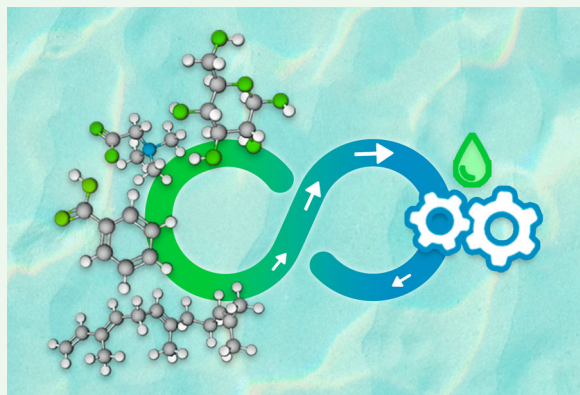
In the last few years, there is a general shift observable toward greener lubrication, fueled amongst others by policy initiatives such as the European Green Deal in consistency with the UN Sustainable Development Goals. At least 70 vol% of a lubricant is composed of a specific base oil, the rest is a variation of additives altering the lubricant properties (enhancing or suppressing existent base oil properties or adding new properties) to be operational for a particular field of application. So, in terms of sustainability, biodegradability, bioaccumulation, and toxicity the type of base oil plays a major role, which makes environmentally harmful petroleum-based lubricant formulations highly problematic for future applications. Hence, this leads to an ever-growing demand of environmentally friendly lubricant alternatives. Within the scope of this review lies the investigation of bio-based, bio-derived, and other sustainable lubricant components that could serve as promising replacements for conventional petroleum-based formulations, in accordance with the principles of green chemistry and tribology. As recycling is embraced by the term sustainability, waste-derived components of non-biological origin are also included in this work. An overview of studies on the tribological performance such as friction and wear properties of these sustainable and benign lubricant components is given.

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Introduction

During the last decade lubricants with low environmental impact have been the subject of research activities and numerous products are available on the market. This is fueled globally due to sustainable development goals (SDGs) to which countries are committed. To give an example, the President of the European Commission, Ursula von der Leyen, has proclaimed the European Green Deal in December 2019 (details see chapter Legal issues) (1).

Measures to implement climate and environmental protection in governmental agendas are given more emphasis. Hence, several foundations have already been laid, accompanied by the propagation of a number of definitions:

What is green chemistry? The concept of green chemistry is defined as 'the design of chemicals and products and processes that reduce or eliminate the use or generation of hazardous substances' by the US Environmental Protection Agency (EPA) (2). Most important in this sense

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is the life cycle of a chemical product, to help with the design of such next-generation products, 12 principles of green chemistry are formulated (3):

- (1) Prevent waste instead of treating or cleaning up in hindsight.
- (2) Maximize atom economy so that few or no atoms are wasted through synthesis.
- (3) Design less hazardous chemical syntheses to pose little or no health risk for humans and the environment.
- (4) Reduce toxicity by a safer chemical product design.
- (5) Solvents and auxiliaries should be safer when needed and avoided when possible.
- (6) Increase energy efficiency to reduce the negative impact on the environment and economy, chemical syntheses should be conducted at room temperature and ambient pressure.
- (7) Use renewable raw materials and feedstocks.
- (8) Minimize or completely avoid unnecessary derivatization products.
- (9) Use catalysts instead of stoichiometric reagents.
- (10) Design chemical substances for degradation at the end of their function.
- (11) Implement real-time, in-process monitoring, and control to prevent hazardous product formation.
- (12) Minimize the potential risk of accidents such as fires, explosions, releases, etc. by choosing safer chemistry.

How green chemistry is implemented in today's economy and the resulting regulations are addressed in detail in chapter Legal issues.

What is green tribology? Tribology – the science of interacting surfaces in relative motion – in general, comprises friction, wear reduction, and lubrication improvement, with the intention to save energy and materials (4). The term green tribology implicates 'the science and technology of the tribological aspects of ecological balance and of environmental and biological impacts' and combines the areas of green engineering and green chemistry. The areas of green tribology incorporate (i) biomimetic surfaces, (ii) environmentally friendly and biodegradable lubrication, and (iii) tribology of renewable sources. The performance of a tribological system plays the most important role in terms of efficiency improvement including prevention of heat pollution, energy dissipation, and protection of material by controlling friction and wear (5). Green tribology becomes increasingly important when heading toward a low-carbon economy and dealing with issues such as environmental pollution, climate crisis, and global energy shortage (4). In this work, we focus on

sustainable lubrication as a contributing factor toward greener tribology.

What is sustainable lubrication? The term 'sustainable' lubrication is not yet universally defined, but it incorporates amongst others 'biodegradable,' 'environmentally friendly,' and 'environmentally acceptable' (bio)lubricants. For a lubricant to be considered as environmentally friendly, it must fulfill the following key criteria to satisfy EPA standards and EU Ecolabel limitations: 1. biodegradability according to OECD 301 and ASTM tests D5864 and D6731 (easily biodegradable), 2. renewability according to ASTM D6866 (carbon content is less than 5 years old and products must have 25% of renewable content) implying a general low carbon footprint, and 3. toxicity according to OECD 201-203 (acute toxicity) and OECD 210-211 (chronic toxicity) tests, or in other words being non-toxic to humans and the environment. Besides, it must be non-bioaccumulative (also see Vessel Incidental Discharge Act) and producible in large quantities in sustainable production. The ideas range from vegetable oils over microbial biofilms and biopolymers to hydration lubrication. Moreover, amongst others the use of minimum quantities (e.g. avoid spills due to specific bottle shape), recyclable packaging, and excluded or limited substances (e.g. for lubricant additives) are addressed within the EU Ecolabel criteria (4,6).

Within this work lubricant components such as base oils and additives of bio-based or bio-derived renewable feedstocks producible in an industrially relevant scale are chosen, which should contribute to less toxicity, an enhanced biodegradation, and promote sustainability of the final product, in line with the green chemistry principles. As a supplementary topic recycled feedstocks are addressed, as they fit within the scope of reducing the environmental impact (circular economy) and maintaining an ecological balance (less primary resource use). Furthermore, a special focus is laid on the tribological performance of formulations proven for industrial applications, while sheer bio-tribological applications are only mentioned briefly and not expanded on.

Legal issues

The year 2021 marked the beginning of the action plan set by the European Green Deal, which works toward a more sustainable economy of the European Union (EU), according to Regulation (EU) No 2018/1999 of the European Parliament and of the Council. In addition, Regulation (EEC) No 880/1992 of the Council of the European Communities describes the European Ecolabel for Lubricants that supports the achievement of those essential climate goals until 2030 (7).

Several national and regional ecolabels, such as the Blue Angel (Germany) or the Nordic Swan (Denmark, Sweden, Norway, Finland, and Iceland) have been replaced and/or harmonized with the European Ecolabel (8). Essentially, it requires (1) the absence of dangerous materials, referring to the European Union Dangerous Preparations Directive; (2) the passing of toxicity tests (OECD 201, 202, 210, 211); (3) biodegradability; (4) low bioaccumulation; (5) a renewable content; and (6) the restriction of certain substances. European Ecolabel Lubricants shall guarantee high technical performance while replacing petroleum-based base oils with alternatives to achieve an energy portion of 32.5% from renewable sources until 2030. By 2050, the long-term strategy of the EU includes the goals to be climate neutral, having an economy with net-zero greenhouse gas emissions, and pursuing efforts to limit global warming below 1.5°C (global average temperature). These goals are in line with the COP21, the climate conference from December 2015 in Paris, and the European Green Deal (7,9).

In the USA, the definition of EAL (Environmentally Acceptable Lubricant) published by the EPA (Environmental Protection Agency) is dominant (9). In comparison to the European Ecolabel, the EPA only includes bioaccumulation, biodegradability, and aquatic toxicity and is only valid for marine applications.

Regarding waste management, Directive (EU) 2018/851, being an amendment to Directive 2008/98/EC, promotes the idea of a circular economy by planning improved waste management requirements and guidance including the whole life cycle of products. Goals of the European Circular Economy sustainability roadmap include a recycling rate of 55% of municipal waste by 2025 and a separate collection of hazardous household waste by 2022 and bio-waste by 2023. Additionally, the Directive is advising Member States to either prevent waste generation in general or to implement a suitable long-term recovery strategy for both re-use and recycling (10).

For US water waste management, the Vessel Incidental Discharge Act (VIDA), formerly named VGP (Vessel General Permit), aims to reduce the environmental impact of discharges (e.g. ballast water), proposing a CWA (Clean Water Act). The CWA regulates discharges of pollutants into the waters of the US and further aims to regulate the quality standards for surface waters (11).

Regulations for commercial lubricants and lubricant components

Only a small number of bio-based lubricants explored at lab and pilot scale will make it to commercial

production. After establishing commercialization, the bio-based lubricant must gain and maintain a market share. The easiest way for a bio-based product to accomplish the market entrance is when it is directly substituting a fossil-based product with matching price and quality and using the same infrastructure (12).

For lubricants and lubricant components, there are lists available classifying their use either as safe for humans and the environment or as potentially harmful. One of those lists is provided by FDA as the 21 CFR § 178.3570 'Lubricants with incidental food contact and limitations of H1 food-grade lubricant components (base stocks and additives), not updated since 1977.' Suitable base stocks and additives for biolubricant formulation might also be found in the White Book™ – Nonfood Compounds Listing Directory of NSF (National Science Foundation), listing countless lubricant components by brand names that are compliant with food safety regulations. Another contemporary (regularly updated) and a comprehensive list is supplied by the European Commission as Lubricant Substance Classification list (LuSC-list) mentioning various substances and base oil brands along with their EEL (EU Ecolabel Lubricant) biodegradability and aquatic toxicity, reaching over various sugars to different glycerides, oil extractives, and fatty acids. Only substances with 100% classification A (ultimately aerobically biodegradable) EEL biodegradation and 100% classification D (non-toxic) EEL aquatic toxicity are approved for the list. Other lists such as the OSPAR List of Chemicals for Priority Action (LCPA), first adopted in 2004, are comprising priority chemicals that pose a concern to the marine environment such as (organo-)metallic compounds, organohalogens, pesti- or biocides, phenols, and others. The list is announced to be updated in 2022 (13–16).

The European Commission lists 37 producers of 138 fully formulated lubricants (by January 2022) in the LuSC list complying with the criteria of the EU Ecolabel according to Commission Decision (EU) 2018/1702. These criteria cover no addition or formation of hazardous substances or substances of very high concern (≤ 0.010 wt.% in the final product), requirements for technical performance and packaging, and strict regulations on aquatic toxicity, bioaccumulation, and biodegradability. Latter demands, that > 90 wt.% of hydraulic and closed gear lubricants, and metalworking fluids, > 75 wt.% of stern tube, open gears, and two-stroke lubricants, and > 95 % of chainsaw, and other total loss lubricants and greases are readily aerobically biodegradable. A minimum of 25% bio-based carbon content (corresponding to EN 16807) is mandatory in the final product (17,18).

Recent research activities

In view of the large amount of research on biolubricants, this systematic review mainly focuses on developments within a time frame of the last seven years (2015–2022) particularly in the field of biologically benign lubricant components for base oil formulations (as in either bio-originated, or bio-derived) from synthetic or modified organic structures, as well as their tribological properties, friction, and wear behavior, and possible field of applications as lubricants. These abovementioned key issues are complemented by the following restriction criteria:

- Inclusion criteria: benign or biomimetic structures, industrial tribology, tribological findings (concerning friction and wear), oil-based lubrication, water-based lubrication, accessibility in industrial quantities.
- Exclusion criteria: publicized research findings older than 2015, without tribological tests, limited to water lubrication applications, limited to human/bio-tribology.

Only those vegetable oil (VO)-derived lubricant components are discussed, that are not primarily in competition with biodiesel production or the food industry, thus will not comply with UN global malnutrition programs as they may serve as food source. Therefore, only non-edible and/or waste-based feedstocks are investigated here but not the entire area of VOs. Furthermore, this work is not enlarging upon the processing steps (synthesis, catalysis, ...) of the mentioned lubricant components and thus, not discussing their environmental impact caused by production processes.

Vegetable oil-derived lubricants

Fatty acid-based components: estolide esters

Estolides are esters synthesized from fatty acids by homopolymerization, creating secondary ester linkages of one fatty acid to the alkyl backbone of another fatty acid. They are mostly produced from vegetable oils but can be synthesized from animal fats or fatty wastes too. Synthetic estolides are classified into (i) glyceride-based, (ii) ester-based, and (iii) fatty acid-based. Estolides are further characterized by the degree of polymerization and the estolide number (EN), which describes the extent of oligomerization ($EN = n + 1$). They show unique physical properties due to different structures and synthesis pathways, convincing with excellent biodegradability, hydrolytic stability, natural detergency, low volatility, high viscosity, and improved oxidation stability and cold flow

properties, which promotes their use as lubricating fluids (19).

The higher viscosity indices (VI) of estolides result in excellent wear protection behavior. Furthermore, due to a high degree of saturation, estolides maintain a very good oxidative stability comparable to other high-end synthetics, for example, when determined by rotating pressurized vessel oxidation test (RPVOT) in (19). Because of the branched structure of estolides, also short chain lengths (C2-C10) show good pour point ranges from -12 to -30°C , awakening the interest to use estolides also as thickening agents. Large hydrophobic branches on both sides of the estolides create a stearic barrier, resulting in very good hydrolytic stability comparable to those of polyalphaolefin (PAO) base oils (19). Naturally occurring estolides are found in various plant seed oils, secretions from the glandular hair of a caterpillar, as well as in human meibomian glands (within the eyelid) as wax esters (19–21).

The structure of estolides and, consequently, the physical properties such as pour point, cloud point, and viscosity, can be affected by the use of catalysts and synthetic conditions. This enables a targeted adaption based on different fields of applications, such as personal care, automotive, industrial, and marine lubricant base oils (21).

To give some examples of tribological use of estolides, castor oil-derived estolides are investigated with similar properties compared to the already established enhanced lubricity of triacylglycerol (TAG)-based estolides from *Orychophragmus violaceus* (Ov) seed oil. Friction and wear tests are conducted on a macroscale pin-on-disk (steel/steel) tribometer, in a reciprocating motion. The lubricants are tested in the boundary lubrication regime with a maximum contact pressure of 1.5 GPa. Uncapped natural OvTAG estolides (hydroxy fatty acids at the terminal end of the estolide branch chain) show a lower coefficient of friction (COF) at 25°C , whereas capped OvTAG estolides (non-hydroxy fatty acids at the terminal end) show a lower COF at 100°C . Mixing uncapped and capped OvTAG estolides leads to an even lower COF, suggesting that both types have a positive impact on tribological performance. Synthetic castor estolides produce less wear volume on the wear scar for surface and ball track width compared to natural castor oil surface/ball track width (22).

Since the use of vegetable oil (VO) as a biolubricant is limited by poor cold flow properties and oxidative stability, manipulation of the chemical structure of VO overcomes these limitations. The modification of vegetable oil-based oleic acid and further conversion to estolide esters and amides via lauric acid-capped estolides is studied along with tribological tests on a four-ball

tester (steel). The anti-wear properties evaluated by wear scar diameter (WSD) are comparable or even better for all estolide ester and amide samples in contrast to commercial lubricants. The lowest wear with the smallest WSD could be established by octyl estolide ester, followed by estolide amides (23).

Fermentation-based components: farnesene

Farnesene ($C_{15}H_{24}$), a sesquiterpene, is an unsaturated hydrocarbon, that holds great potential as a future lubricant feedstock for base oils since it can be biotechnologically produced from any kind of fermentable sugar feedstock using adapted microbial cells by metabolic engineering. Co-oligomerization of hydrogenated β -farnesene with (petroleum derived) linear alpha olefins, LAOs, (>90% saturates, VI > 120, sulfur < 0.03%) leads to a base oil (farnesene-derived base oil, FDBO) with 30 carbon atoms, that is ready to compete with Group III mineral oils and Group IV PAOs as renewable, low toxic and biodegradable alternative. Furthermore, FDBO applies to the criteria of the EU Ecolabel and the US EPA VGP (Vessel General Permit) (6).

In recent approaches it is demonstrated that farnesene can be produced from sources not competing with the food industry, for example, as α -farnesene from engineered yeast such as *Yarrowia lipolytica* using waste lipid feedstock (24) or *Saccharomyces cerevisiae* (25) and as β -farnesene from bacteria (26). Farnesene could already be successfully produced on an industrial scale from sugar feedstock using genetically modified *S. cerevisiae* yeast strains (6). Moreover, engineered cyanobacteria can be used for α -farnesene production utilizing CO_2 as feedstock (27).

So far, the oligomerization step with the LAOs is the limiting factor for FDBOs biodegradability and environmental friendliness (which is still above those of PAOs), but squalane, a C30-saturated hydrocarbon, might provide the solution. Squalane, a 4 cSt base oil, naturally found in shark livers and olive oil, can be obtained by the dimerization of two farnesene molecules and exhibits superior biodegradability (86% for farnesene+farnesene-derived FDBO, 74% for LAO+farnesene-derived FDBO, 48% for LAO-derived PAO) (6). Squalane is often used in studies as model fluid e.g. as lubricant base oil investigating molecular dynamics with certain additives (28), in EHL traction prediction for rolling bearing applications (29), or for density and viscosity estimation for high-pressure industrial equipment applications (30). Furthermore, it is tested as a lubricant base oil e.g. in mixtures of polyisoprene for gear-mixed lubrication (31). However, general tribological studies of farnesene-derived lubricant base oils are still rare. Bio-based PAO is discussed in detail in chapter PAO.

Carbohydrate polymers

The trend is leading toward plant-based substitutes for petroleum products, due to the renewability aspect contributing to sustainable industry development in line with SDGs. However, this does not necessarily represent also a low environmental impact pointing out, for example, deforestation for palm oil production. Therefore, these feedstocks are excluded in this review. The discussed substitutes include materials originating from biomass such as (hemi-)cellulose, chitosan, and lignin (see chapter Lignin and lignin-derived lubricant components), and non-polymeric materials such as phospholipids and triglycerides. Biomass means organic matter composed of carbon, hydrogen, oxygen, and nitrogen, often having a similar chemical structure as fossil-derived products (32, 33). Carbohydrate-based lubricants are naturally found in the skeletal joints of animals and humans (34) but can also be produced by plants (35), bacteria (36), and fungi (37).

Polysaccharides and glycans

The Pacific Northwest National Laboratory (PNNL) and the National Renewable Energy Laboratory (NREL) supply lists for 'Top Value-Added Chemicals from Biomass,' referring to the top feasible biological or chemically converted pathways for chemical building blocks from biomass sugars, synthesis gas (Vol 1, released 2004), and lignin (Vol 2, released 2007). More than 300 potential compounds from sugar pathways were screened and the top 12 candidates are specified (38,39).

The amphiphilic nature of cellulose makes it interesting for the use as lubricant or additive, since it is interacting with both polar material surface and a non-polar lubricant. Compared to cellulose, which is a homopolysaccharide only built from glucose monomers, hemicellulose is a heteropolymer of five different saccharides. Cellulose can be a precursor for glucose, hydroxymethyl furfural (HMF), levulinic acid, or formic acid (32). Hemicellulose is used for the production of furfural (Top 30 biomass-derived compounds (38)), which provides a stock for 2-methyltetrahydrofuran (MTHF), a potential fuel additive. (40) Cellulose is often used as bio-based lubricant additive, in castor oil or other vegetable oils e.g. as thickener in the form of epoxidized cellulose pulp (ECP) (41), methylcellulose and cellulose pulp, as well as chitin (42), cellulose acetate butyrate (CAB) in acetyl tributyl citrate (ATBC) (43), or as friction-reducing and anti-wear additive in the form of fibrillated or crystalline nanocellulose (44). Also, the potential of dielectric constant variation to control friction behavior with different concentrations of nanocellulose particles is

studied (45). Others discuss the potential biolubricant base oil production from lignocellulose-derived 5-hydroxymethylfurfural (HMF) (46), or 2-alkylfurans and ketones (47). Even superlubricity (COF < 0.004) could be achieved with hydroxyethyl cellulose (HEC) in water for 0.25–2 wt.% and applied loads of 5–9 N on quartz glass in a rotary micro-tribometer (48).

When it comes to lubricating greases, products based on nanocellulose oleogels produced from castor oil are compared to common lithium-based lubricant greases for industrial application. Rheological and linear viscoelastic behavior in a parallel plate-plate setup with a shear rate from 10^{-2} to 10^2 s⁻¹ and 0.08 to 100 rad/s dynamic shear frequency sweep range show the comparable performance of 1.4 wt.% oleogels and the synthetic lithium soap, even though latter contained 8 wt.% of thickener (49).

The influence on thermal, rheological, and tribological effects of ionic and non-ionic gelating agents (originating from cellulose derivatives) in water-polyethylene glycol (PEG) mixtures as base fluids are of interest as the effect of starch polymers on the lubricity, where the ionic gelator showed lower friction and wear than to the non-ionic gelator (50). The impact on friction by the addition of carboxymethylated, hyaluronic acid-based polymers or fatty acids such as stearic acid to methylcellulose compounds result in a decrease of friction-induced wear using a ball-on-disk tribology setup (51,52).

Amongst the polysaccharides fructose and chitosan have to be mentioned as promising lubricants components. Similar to the mentioned tribo-experiments with HEC, superlubricity is also achieved with fructose dissolved in ethylene glycol and 1,3-propanediol in weight ratios from 0.1 to 0.5. At loads between 3 and 6 N in a steel-steel ball-on-disk setup, COF ranging as low as 0.004–0.01 (at 0.5 wt.%) are measured (53).

Chitosan is a polysaccharide consisting of acetylated and deacetylated glucosamine-units, collected by deacetylation of chitin from shells of crustaceans and fungal cell walls, making it the second most abundant natural polysaccharide right after cellulose (33). However, chitosan-derived multifunctional lubricant additives utilized as antioxidant, anti-friction, anti-wear, and anti-corrosion additive, have potential to be an environmentally friendly bio-based alternative to existing non-biodegradable additives such as zinc dialkyldithiophosphate (ZDDP). Chitosan and its derivatives are also promising bio-replacement for vegetable oils as thickeners and modifiers in greases and oleogels (54). Addition of 4% sugar-derived N-octadecyl-D-gluconamides (NOG) gelator to polyethylene glycol (PEG) base oil shows an improvement of

the thermal stability and reduction of friction and wear (55).

Similarly, saccharides do not only serve as promising green additives but also provide potential as environmentally friendly lubricant base oil, for example, xylose from sugarcane bagasse can be effectively used as feedstock for oleaginous yeast to produce single cell oil (SCO) (56). Comprehensive overviews of carbohydrate-based biolubricants from algae and bacteria focusing on extracellular polysaccharides, such as agar, carrageenan, and alginic acids are available (35).

Glycans are complex carbohydrates, linking monosaccharide units through glycosidic bonds, forming linear or branched polymers. They can be either free oligo- or poly-saccharides or bound to proteins (e.g. glycoproteins) and lipids (e.g. glycolipids) (57), and can be found in animals, plants, and fungi (37). As highly functional glycoproteins, so-called mucins, they play a significant part in the field of implant and cartilage wear reduction or prevention, contact lenses, cosmetics, oral salivary (58), synovial fluid (hyaluronan, a glycosaminoglycan) (59), or reducing friction between cornea and eyelid as tear fluid (60,61). In bio-medical application lubricin, a mucin-like glycoprotein lubricin-like synthetic polymers are important boundary lubricants, and naturally occurring alongside hyaluronic acid in the synovial fluid like mucin, lubricin can bind to surfaces and trap water close to the surface, enhancing gliding and reducing friction at the same time (62). Outside the field of bio-tribology, base oils from glycans are not yet studied for their industrial tribological relevance.

Lignin and lignin-derived lubricant components

Lignin is a complex heterogeneous polymer, naturally found in wood, of cross-linked phenolic moieties, mainly p-coumaryl, coniferyl, and sinapyl alcohols, varying in concentration based on the plant source (63). The structure of lignin is diverse, affected by the source of wood and recovery methods (64). Due to the many functional sites within its structure, lignin has a wide range of applications, for example, its amphiphilic nature makes it a good anti-corrosion agent (33). Based on various pathways, modifications on aliphatic and aromatic hydroxy groups of lignin are possible, and different applications for lignin acquired from different sources are known (65).

Various feedstocks are discussed in the literature, ranging from kraft lignin functionalized with laccase SilA from *Streptomyces ipomoeae* (66), lignin-enriched fractions from sugarcane bagasse waste (67) or NCO-functionalized residual lignin fractions from eucalypt and pine wood in castor oil (68). Recently, a new method for low energy-consuming electrochemical

depolymerization method of lignin by using levulinic acid as a sustainable bio-based component for the solvent system is reported, [Figure 1](#) shows the insoluble fraction of kraft lignin ([69](#)).

Lignin is mainly studied as additive in lubricants, for example, as a thickener in the form of epoxide-functionalized alkali lignins ([70](#)), and as friction and wear improver ([71–73](#)). It has been found that the broader the distribution of lignin's macromolecules and the higher the concentration of lignin's hydroxy groups, the better the lubricant properties when using it as a biodegradable additive with polyethylene glycol ([64](#)). Organosolv lignin has an outstanding adhesion character on metal surfaces as well as excellent film strength and is a good non-corrosive green lubricant additive alternative ([74](#)).

Blending with polyethylene glycol (PEG) results in enhanced thermal stability, while wear loss could be reduced up to 93.8% compared to pure PEG when mixed with lignin ([63](#)), or by 89% when mixed with ethylene glycol (EG) ([72](#)). Recent research also implies the usage of lignin as a biolubricant base oil by converting lignin-derived monomers and aldehyde into branched benzene and branched cyclic lubricant base oils ([75](#)). In the last five years, lignin becomes increasingly interesting as a green component in lubricant formulations, but tribological data for lignin-based base oils are still scarce.

Hydrogels

A hydrogel is a macromolecular polymer, which can absorb large amounts of fluids within its interstitial space while remaining insoluble in water due to

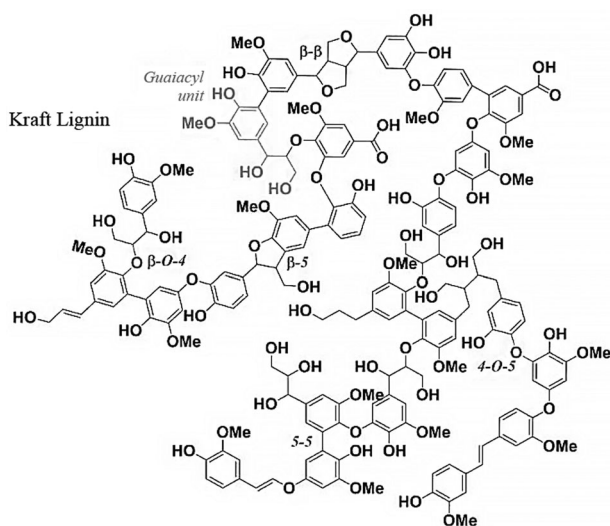


Figure 1. Structural characteristic of kraft lignin from industrial spent pulping liquor. With permission from ([69](#)).

present physical or chemical cross-linking. Biomimicry refers to biological methods and systems found in nature, typically environmentally friendly, that is emulated by modern engineering and technology design. Biomimetic lubrication is often used in the sense of water-based lubrication as in bio-tribology ([5](#)). In general, hydrogels are used as biomimetic lubricating materials in biomedical applications such as drug delivery ([76](#)) and tissue engineering, for example, for articular cartilage repair, since they show good biocompatibility, viscoelasticity, permeability, and therefore, biomimetic properties, but lack in friction and wear properties ([77, 78](#)). They can be synthesized from plant-derived materials including cellulose, hemicellulose, lignin, polysaccharides (starch, pectin, gum), and proteins ([76](#)).

Recent biomedical tribological examinations are conducted with hydrogel in the form of cross-linked alginate-polyacrylamide in invasive joint implants ([79](#)), being agarose-hyaluronan based ([80](#)), as anisotropic sugarcane composite replacing biological tissues ([81](#)), as material for contact lenses ([82–84](#)), or basil-based lubricant gels with ethanol as an additive in catheters and gastric tubes ([85](#)).

Besides that, the tribological properties of gel-like, glycol-based lubricants for application in, for example, water-based bearings or high load conditions are of interest. The lubricating gels are functionalized with nitrogen and phosphorus small molecules in mixtures of water-glycol and water-diethylene glycol, forming a 3D network structure with self-assembly functions. These gels show good anti-wear and friction properties with low wear volume losses at 400 N compared to the base fluids without gels, especially for the 4% gels. Also, for the increasing frequency at 400 N, the 4% gels could maintain a lower friction coefficient throughout the experiment time of 30 min. ([86](#)). Furthermore, sugar-based low-molecular-weight gelator hydrogels are studied as environmentally friendly lubricants ([87](#)).

Polyalcohols

About 75% of polyalcohols (polyols) belong to the group of polyether polyols, whereas the remaining 25% belong to the group of polyesters and other polyols ([88](#)). The representatives of the polyol group range from the simplest ethylene glycol over diethylene glycol, triethylene glycol, and tetraethylene glycol to polyethylene glycol ([89](#)). Renewable polyols can derive from plants-based components such as carbohydrates (monosaccharides, starch), proteins, and essential oil extracts. In addition, polyols can be recollectored from agro-residue or waste from households and the food processing industry, to not compete with possible food sources in the first place ([90](#)). In the lubricating industry, neopentyl polyol

esters composed of a central quaternary carbon atom with attached primary alcohol methylol groups are usually used as base oils for synthetic lubricant production. This relates especially to neopentyl glycol (NPG), pentaerythritol (PE), and trimethylolpropane (TMP) (6).

Research on the usability of polyalcohols as sustainable lubricants mainly focusses on their tribological behavior. PE ester, NPG ester, and a formulated lubricant are compared to a commercially available non-bio-based one. The formulated lubricant is made from PE and palm oil methyl ester with extreme pressure, anti-wear, and corrosion inhibitor additives. The coefficient of friction is determined in a four-ball steel/steel tribometer, in mixed and boundary lubrication regime from 50°C to 100°C. Concerning friction and wear, the formulated lubricant gives the best results compared to PE base oil (91). Similarly, the tribological behavior of PE, NPG, and trimethylolpropane (TMP) as vegetable oil-based polyol ester lubricants originating from waste mango seed kernel oil is investigated with a four-ball tribometer according to ASTM D4172. PE ester shows a lower coefficient of friction and a smaller wear scar diameter related to its higher viscosity compared to NPG ester and TMP ester (92). In another approach, TMP is used to enzymatically esterify the microbial lipid of the yeast *Rhodotorula glutinis* to produce a biolubricant with excellent low temperature, and good friction (COF < 0.1) and wear properties (93). Palm oil-based polyol is investigated as a green lubricant concerning its viscosity, viscosity index, flash point, coefficient of friction, and wear properties. The tribological tests are run on a four-ball tribometer (steel/steel) showing a higher COF for the polyol sample (COF = 0.071), but a decreased wear scar (WSD = 624 μm) compared to palm-oil (COF = 0.064, WSD = 660 μm) (94). Tribological investigation on a ball-on-disk tribometer, rolling bearing tests, and wear tests with a combination of glycerol and sorbitan monooleate as base oil with additives such as chitosan, glyceryl monostearate, and calcium phosphate lead to a decrease in the COF. For the combination of high-oleic sunflower oil (HOSO) with glycerol and cellulose ether as additive this effect could not be observed (95). Glycerol monooleate can efficiently reduce the COF as a 5 wt.% additive in PAO in boundary lubrication (96). Glycerol in mixtures with propanediol shows superlubricity performance with a coefficient of friction below 0.01 and 723–871 MPa Hertzian pressure for steel tribo-pairs in a ball-on-disk setup (97). Additionally, polyols such as 1-decanol are studied in the elastohydrodynamic lubrication regime, contact conditions that are found inside bearings and gears. The coefficient of friction is evaluated in a ball-on-disk setup (steel-steel, steel-glass, and

glass-glass), a load between 1 and 50 N with Hertzian pressure from 0.1 to 1.1 GPa and macroscale superlubricity is observed for all surfaces (98).

Ionic liquids

Ionic liquids (ILs) are composed of a positively charged bulky organic cation, and an organic or inorganic anion (99). The room temperature ionic liquids are liquid at ambient conditions due to poorly coordinated asymmetric moieties and are most suitable for base oil lubricant design. The ILs generally exhibit high thermo-oxidative stability, non-flammability, and strong affinity to metallic surfaces, which predetermines them as promising lubricant candidates especially for highly demanding applications. ILs can be also recovered in high yields (>95%) which fulfills the circular economy demands to limit resource consumption, waste, and pollution (100). Their physicochemical properties can be tuned due to immense cation and anion structural variability. These characteristics make ILs a promising class of substances for the development of potentially high-performing and eco-friendly lubricants. As the structural variability of ILs is immense, the determination of structure–activity relationships is crucial to pre-design ILs with low environmental impact.

Structural recommendations from toxicity and biodegradability perspective

Since ionic liquids are composed of diverse chemistries and the ions are often of non-biological sources, the toxicity of these mixtures is of special concern. In general, a quick classification is mostly not possible and therefore, this topic is covered with particular care in this chapter. The IL toxicity mainly depends on the nature of cation and its alkyl chain length. The longer the side chain the higher the toxicity, due to increased lipophilicity enabling cell membrane disruption (101). The antimicrobial toxicity increases especially for C₁₂–C₁₄ chain lengths but polar functional groups, for example, ester, can reduce the toxicity of long side chains (102). The aromatic imidazolium and pyridinium-containing ILs have tendency to inhibit enzyme activities (103). The non-aromatic cations, such as quaternary ammonium exhibit lower toxicity compared to aromatic moieties (100). Morpholinium and dicationic IL moieties are also featuring low toxicity effects due to reduced lipophilicity (104). Especially polyfluorinated anions BF₄⁻, PF₆⁻, and favored [(CF₃SO₂)₂N]⁻ are more toxic compared to, for example, Br⁻ and Cl⁻ halides due to increasing overall IL hydrophobicity leading to cell membrane damage (105). In general, the influence of the anion moieties on toxicity is less predictable than that of cations (106).

Biodegradability aspect is crucial to avoid issues related to bioaccumulation or high mobility. Unfortunately, the opposing trend to toxicity recommendations exists, with preference for longer alkyl chain lengths. The short-chain imidazolium cations, such as C_1 – C_4 have been reported to exhibit poor biodegradability [107]. Functionalization of ILs with e.g. ester-, carboxyl- or terminal hydroxyl-group significantly enhances biodegradability due to the possibility of enzymatic attacks (104). The cyclic side chains are causing the drop in the enzymatic activity and cause lower biodegradability (100). Also, branched side chains are more resistant to biodegradability (107). Aromatic functional groups and symmetric side chains of the same length cause bulkier or symmetric moieties and are not biodegradable, while polar functional groups improve the biodegradability of at least short side chains. The imidazolium-based ILs undergo very limited biodegradation as well as dicationic moieties in contrast to ammonium and pyrrolidinium undergoing full mineralization (104). The inorganic anions do not contribute directly as a carbon source to biodegradation, as they are already in their mineralized form. Anions with high carbon content, such as octyl sulphate, feature excellent biodegradability due to being modeled on sodium dodecyl sulphate used as a positive control in biodegradation tests (107). The fluorinated anions with a large portion of highly stable C–F bonds are not biodegradable while carboxylic acids with linear C_4 – C_{10} chains are generally biodegradable but shorter chains are toxic (104).

Thus, it is difficult to reconcile the opposing structural demands from toxicity and biodegradability perspective. However, some overlap of recommended structural features can be identified to comply with both requirements. The eco-friendly IL design should focus on non-aromatic cations with non-halogenated counter anion and the side chains should be kept below C_{12} length, while functionalization by polar groups (carboxy-, ester-, terminal hydroxyl-group) is preferential, as summarized in Figure 2.

As the cation and anion have distinctive structural features, their toxicity and biodegradation as well as mobility and bioaccumulation in environment can significantly vary. Thus, it is not sufficient to design ILs with only one moiety (cation or anion) derived from, for example, naturally occurring sources to ensure their environmental benignity, while neglecting the impact of the counter ion (104). Unfortunately, this trend is seen in the majority of publications from tribological field claiming the green IL formulation, so these will be omitted herein. Apart from deliberate IL structure modeling bound to the need of extensive toxicity and biodegradability studies, is the utilization of naturally

occurring bio-derived cation and anion moieties. Hence, tribological performance will be described only for ILs with both bio-based moieties.

Tribological performance of ILs with bio-based moieties

From tested choline-based ILs with amino acid anions, the choline L-phenylalanine [ChPhe] achieve the lowest wear volume and 46% wear reduction compared to 1-hexyl-3-methylimidazolium bis(trifluoromethane sulfonyl)imide [C_{6mim}][NTf₂] reference while leading to a thicker film exhibiting more effective anti-wear properties although displaying the highest COF (108,109). Excellent anti-wear and friction reduction properties are reported for choline ILs with aspartic [Ch][Asp] and glutamic acid [Ch][Glu] anions (109,110). Promising anti-corrosion properties and 70% lower friction than for paraffin-based reference oil are shown by choline L-proline [Ch][Pro] under full film elastohydrodynamic lubrication (EHL) regime (111).

Additionally, superlubricity (COF < 0.01) can be achieved by [Ch][Pro] in glycerol aqueous solution applied between 1-3 wt.%, while 3 wt.% concentration displayed the lowest COF (112). Addition of lignin into [Ch][Pro] results in low friction and good wear protection of both DLC and steel surfaces (73). Also, choline monocarboxylate acids exhibit excellent lubrication properties similar to amino acid-based counterparts even for difficult to lubricate copper and aluminum-based surfaces (113). Furthermore, choline with ricinoleic fatty acid anion, found in castor seed oil (44), can reduce COF in comparison to plain glycerol solution (114).

Further, ILs based on naturally occurring quaternary ammonium, being also identified as preferential cation for ILs with low environmental impact (see Structural recommendations from toxicity and biodegradability perspective), are designed with fatty acid counter anions. Fatty acid anions hexanoate ($C_{6:0}$), octadecanoate ($C_{18:0}$), and octadec-9-enoate ($C_{18:1}$) with ammonium-based (N_{8881}) cation are investigated for their tribological and corrosion performance on steel-steel, steel-aluminum alloy, steel-bronze, steel-cast iron, and steel-tungsten carbide surfaces. After 21 days of corrosion testing, only steel-bronze shows corrosion activity for [N_{8881}][$C_{18:0}$] and [N_{8881}][$C_{18:1}$] ILs. The three ILs show comparable friction behavior on the same counter-pair materials, the COF of 0.06 and lowest wear is measured on tungsten carbide (115). When ammonium (N_{1444}) octanoate ($C_{8:0}$) is compared to fluorinated L-F104 imide it exhibited lower friction and much less wear at loads of 200 N for steel/steel and 100 N for steel/copper and steel/aluminum contacts (116). In another study, oleate ($C_{18:1}$)

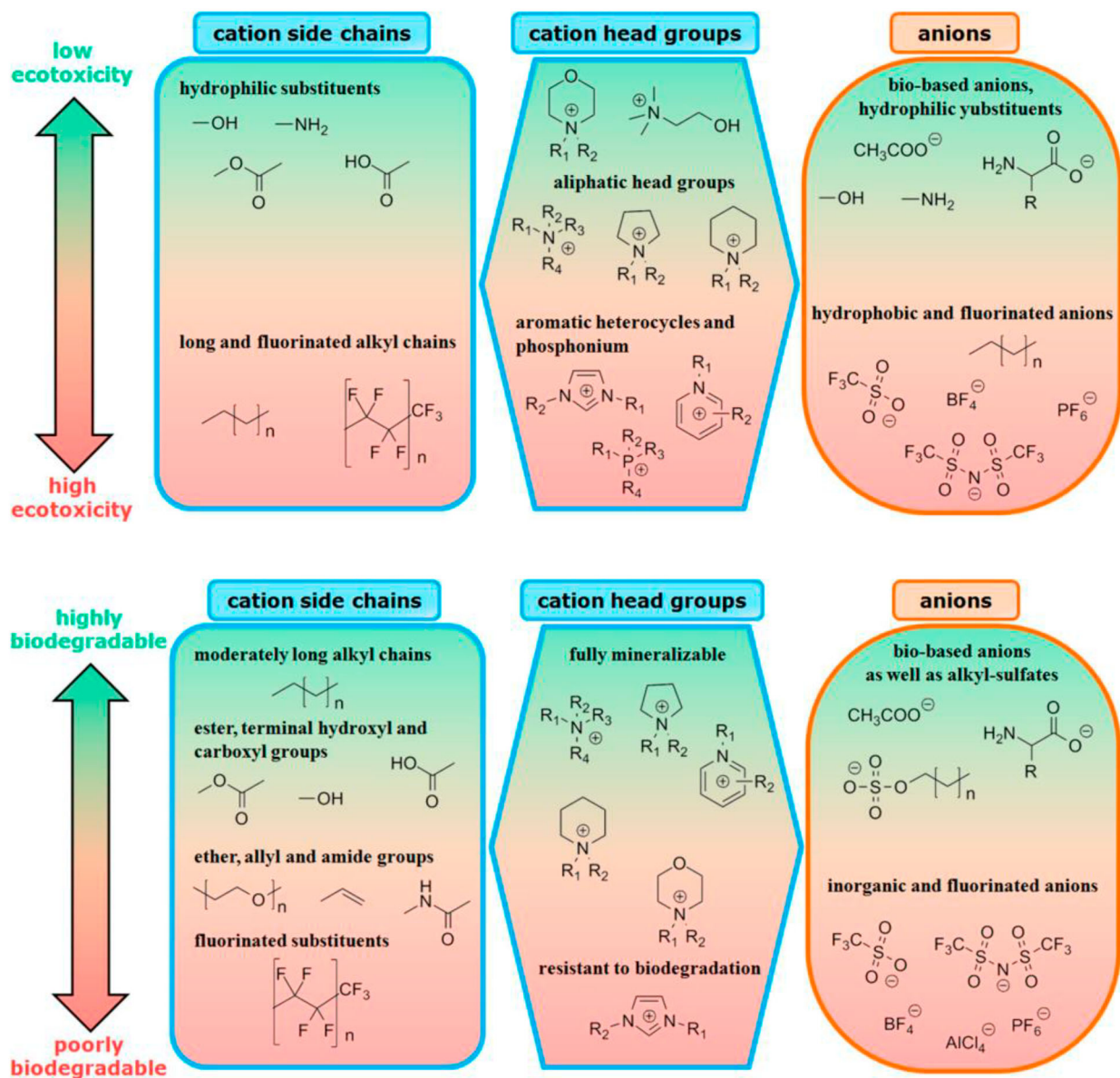


Figure 2. Structural recommendations for IL's design with the lowest environmental impact. With permission from (104).

tetrabutylammonium (N_{4444}) performs as the best anti-wear and friction-reducing anion under boundary lubrication when compared to other tested C_6 , C_8 , and $\text{C}_{18:2}$ anions (117). When studying ILs based on (N_{4444}) cation and C_{10} , C_{12} , C_{14} , C_{16} , and $\text{C}_{18:1}$ fatty acid anions as 1% additives in mineral oil, it is observed that COF slightly increases with alkyl chain length increase but decreases with unsaturation as a result of changes in viscosity and in van der Waals interaction triggered by these structural differences (118). Preliminary studies propose that longer alkyl chains of cations lead to higher viscosity accompanied by an increase in van der Waals interactions but this effect is also influenced by interactions between cation and anion and the anion moieties (119). Thus, when comparing $[\text{N}_{6666}][\text{C}_{8:0}]$ and

$[\text{N}_{6666}][\text{C}_{16:0}]$, the latter has a lower viscosity, a higher viscosity index, and a lower COF. The $[\text{N}_{6666}][\text{C}_{8:0}]$ is shifting from EHL to mixed lubrication regime at higher speeds and temperatures leading to an increase in COF, whereas $[\text{N}_{6666}][\text{C}_{16:0}]$ remains in EHL regime most of the time. Ball-on-disk tribo-tests show slightly better friction performance of $[\text{N}_{6666}][\text{C}_{8:0}]$ over $[\text{N}_{6666}][\text{C}_{16:0}]$ (120). Adding protic long-chain palmitate-based ($\text{C}_{16:0}$) IL in 1 wt.% to protic short-chain citrate IL, both with ammonium-based cation, increases the viscosity and improves the tribological performance (121). Furthermore, excellent anti-wear and good friction reduction properties (COF ~ 0.1) are accomplished with 2% palmitate ammonium IL in water, without any signs of tribo-corrosion (122).

Further potential bio-moieties for IL design, such as glycine betaine present in plants or sweeteners, for example, saccharinate, acesulfamate, or medically proven docusate might be explored. When both cation and anion are naturally based, the ILs have the highest chance to comply with rigorous toxicology and biodegradability criteria. However, as for all bio-derived lubricants, the IL design must ensure not only the non-toxic and biodegradable features but must prove to deliver desired performance under particular application conditions (101,104).

Other natural compounds with tribological potential

The following structures were chosen due to being benign and contributory to the field of sustainable lubrication, most of them are researched as lubricant additives in the field of bio-tribology. Furthermore, they are industrially synthesized in commercial-scale productions with long-standing expertise. While they are not yet investigated as industrial lubricant components, they might be considered as environmentally friendly lubricant additives, for example, in micro-tribology applications (for low load contacts like in micro electromechanical systems, MEMS).

Proteins

Often proteins such as albumin, globulin, mucin, and lectin are studied in the context of human bio-tribology for prosthetics and implants (123,124), joints and cartilages (125,126), oral tribology (dryness, response to food,...) (127–131), contact lenses (132,133), etc. Within a biomimetic approach, bovine serum albumin (BSA) is investigated as a potential additive in concentrations from 0.1 to 0.6 mg/ml in rice bran oil (RBO) for bio-cutting fluid formulations. Tribo-tests are carried out on a four-ball tester (steel-steel), where the coefficient of friction and wear scar diameter values are the lowest for 0.4 mg/ml BSA in RBO. The green cutting fluid could further outperform a commercial cutting fluid based on the coefficient of friction values at 400–1000 rpm disk speeds as well as in weight loss of pin using a pin-on-disk tribometer (iron-steel) with 100 N constant load in a rotating motion (134).

Vitamins

Vitamins are a natural component in various plant-based oils, but their influence on tribological performance especially outside of bio-tribology is hardly investigated. A fully formulated biolubricant from esterified rubber seed oil (E-RSO) is tested with 1.5% α -tocopherol (vitamin E) and 1% L-ascorbic acid (vitamin C) as

antioxidant additives and compared to the plain rubber seed oil and commercial mineral oil (SAE20W40). The values for (bio-)chemical oxygen demand of E-RSO are within the accepted ranges of biodegradability. A combination of the named vitamin-based antioxidants along with butylated hydroxyanisole (BHA) shows better oxidative stability of the fully formulated E-RSO than plain E-RSO and RSO and is comparable to the commercial SAE. Furthermore, the friction and wear properties of fully formulated E-RSO are superior to SAE (135).

Lecithin

Lecithins are glycerophospholipids. Phosphatidyl choline, phosphatidyl inositol, phosphatidyl ethanolamine, and phosphatidic acid form the main phospholipids in lecithin from soya and sunflower. The soybean lecithin is obtained by degumming the extracted oil of the used soybean seeds. Lecithin is non-toxic, well tolerated, and can be entirely metabolized by humans (136). Soy-derived lecithin is used as gel-forming thickener, due to its ability to self-assemble into vesicles in polar solvents like glycerol, formamide, and ethylene glycol, which might be of interest in biolubricant applications (137). Moreover, lecithin is used as an anti-wear additive in hydrotreated mineral oil (10 cSt) and diester, where 0.5 wt.% of lecithin reduces the wear scar diameter by 21 times and the 1.5 wt.% solution by 45%, it has better friction and anti-wear properties than ZDDP (136,138).

α -lipoic acid esters

The α -lipoic acid (5-[1,2]-dithiolan-3-yl-pentanoic acid) occurs naturally in plants and animals. It is a cofactor of several enzymes and possesses antioxidative properties. Derivatives of lipoic acids are potential natural and environmentally friendly alternatives to toxic additives due to their cyclic disulfide moiety. First tribological tests of selected lipoyl esters in synthetic base fluid indicate good anti-wear properties (139).

Benzoic acid

Benzoic acid occurs naturally in many plants. It is reported that the addition of benzoic acid to titanium greases improves anti-wear properties (140). Low friction coefficient and high anti-wear properties are observed for the new category of titanium complex grease using titanium tetrakisopropanolate, 12-hydroxystearic acid, and benzoic acid as additives in naphthenic mineral oil (KN 4010)/polydimethylsiloxane as base oil. (141). Good anti-wear properties and low coefficient of friction are observed for 2 wt.% benzoic acid tested in Jatropa

vegetable oil when compared to mineral oils SAE 5W-30 and SAE 10W-30 (142).

Common synthetics

Unlike mineral oils, which are mainly classified by their performance, synthetic (bio)lubricants can be categorized according to several approaches, for example, according to the feedstock or the synthesis. The most widely used classification is according to their chemical composition (143,144). Due to the wide range of various chemistries, several groups can be defined. However, in the following, only some of the most common synthetic lubes shall be discussed. This includes (a) synthetic esters, (b) synthetic hydrocarbons, particularly polyalphaolefins, and (c) (poly)ethers.

Synthetic esters

In the first decades after the invention of synthetic ester lubricants the focus was primarily on their ability to provide equipment protection advantages under extreme operating conditions, including very low or very high temperatures, and their ability to resist oxidation. In the past decades, the interest shifted more and more toward their ability to minimize the impact on the environment and use them as so-called 'biolubricants' (143). Hereby, the term biolubricant is assigned to lubricants either made from bio-based raw materials or other environmentally benign hydrocarbons (145,146).

These fully synthetic esters may be differentiated into two groups, both of them showing high biodegradability and low toxicity: (1) diacid esters and (2) polyol ester (144,147). While this does not mean ester oils automatically show environmentally benign characteristics when fully formulated, there are certain classifications for additivated synthetic esters for this purpose, for example, the HEES type classification for hydraulic applications (148).

One major aspect when it comes to stability is the crucial knowledge of structure-stability relationships (149). With this knowledge, nearly tailor-made lubricant components for each application can be synthesized. Based on targeted synthesis via long-chain alcohols and acids, the resulting completely saturated synthetic esters show high thermo-oxidative and hydrolytic stability as well as more stable ageing characteristics (145). For example, feedstocks include C6–C13 alcohols (i.e. n-hexanol, n-heptanol, isonal, and decanol), C5–C18 mono acids (i.e. valeric, heptanoic, pergalonic, and oleic acid) with neopentyl polyols, such as pentaerythritol (PE), polyol esters, diacids (i.e. adipic acid, azelaic, sebacic, and dodecanedioic), and various dimer acids (145).

In the production of esters from non-edible animal fats those having less saturated acids improve oxidation

stability as well as better lubricity properties. However, the lower amount of unsaturated acids increases the pour-point. When it comes to applications, where a low ash-content of the base-oil is beneficial, synthetic esters with shorter-chain alcohols are better performing (150).

Besides chemistry concerns, the tribological performance of synthetic esters is a main area of research. Different isomers of isooctyl naphthate-based oils show, compared to isooctyl sebacate as a reference, higher oxidation stability as well as lubricity. It is found that the introduction of a naphthalene ring improves the flash points, stability, and lubrication properties due to the large π -electron conjugate system. Tribotests with an SRV ball-on-disk tribometer show a positive effect in terms of lubrication of the naphthalene ring system, additionally (151).

When the friction performance of synthetic esters is measured with a pin-on-disk tribometer a correlation with the molecular structure of the lubricant is revealed. Esters with branched-chain structures show an improvement in low-friction performance with an increasing number of branches or carbon atoms. For esters with phenyl-groups, 1,3-benzene esters seem to be better performing than orthophthalic esters. Tribological performance of the esters with the chain structures seems to be better than those with the phenyl group when the difference in their viscosities is small (152).

Based on one of the 12 priority chemicals identified by the US Department of Energy for the establishment of a 'green' chemical industry (38), 5-furandicarboxylic acid, an isooctyl furan dicarboxylate (FD) as lubricating oil is synthesized. Tribo-experiments with a 4-ball tribometer (steel ball) and SRV tribometer (steel/steel, steel/copper, steel/aluminum) show the superior performance of Isooctyl-FD to the references isooctyl sebacate and isooctyl adipate in terms of anti-wear and friction behavior. However, viscosity-temperature and low-temperature properties are inferior. These properties should be improved by additives (153).

Synthetic esters do not only act as a base oil but can also be applied as additives. A pentaerythritol rosin ester (PRE) is synthesized from abietic acid, an abundant renewable natural resource, to act as an environmentally friendly multifunctional additive in rapeseed and soybean oil. In blends with 20 wt.% PRE, the onset temperature of oxidation as well as the oxidation induction time are increased significantly, which proves the potential of PRE as a substitute to common additives in eco-friendly bio-based lubricants (154).

In addition to conventional and commonly used surfaces, the performance of biodegradable oils on special

coatings is a topic of high interest, as so far, the tribological behavior is not fully investigated (155).

PAO

Polyalphaolefines (PAO) can be designed with higher biodegradability but since their industrial-scale production is not necessarily based on renewable materials they are not considered as bio-based in general. However, if they fulfill this criterion, they can be counted as an important class of high-performing synthetic environmentally friendly lubricants base oils, in particular in Europe (147).

There are numerous studies comparing the performance characteristics of different synthetic biodegradable lubricants such as esters or PAOs with mineral oils in order to increase the knowledge on similarities and differences in tribological performance. The friction behavior and wear protection ability of these selected base lubricants are compared using a pin-on-disk tribometer. Based on the results it can be concluded that the viscosity has the predominant influence on the friction and wear-protection properties of PAO and mineral oils, contrary to ester oils where these properties are influenced by their polar functional groups. All tested lubricants generally obey the trend of a typical Stribeck curve, and the increase in viscosity decreases the minimum friction coefficient at the valley (between mixed and full-film lubrication regime) of each Stribeck curve (152).

To investigate the effect of the structure on the performance, a bio-based PAO-40 synthesized from complex vegetable oils is compared to a non-functionalized mineral-based commercial PAO-40 synthesized only from alpha olefins. The effect of methyl ester functionalization on physicochemical properties and tribological performance is investigated. A beneficial influence of the methyl ester groups on several parameters is revealed, amongst them a higher density between 40°C and 100°C, higher viscosity index, lower COF, and lower wear scar diameter when using 4-ball tribometer, and a higher elastohydrodynamic (EHD) lubricant film thickness under boundary conditions. However, in terms of oxidation stability evaluated by pressurized DSC and total acid number, the performance of the conventional mineral-based PAO without methyl ester groups is better (156).

In applications with nano-grained stainless steel, which shows a higher susceptibility for oxidative wear due to the increase of grain boundaries, PAO 4 shows an effective improvement of the wear-resistance and friction-reduction compared to dry conditions. Under lubricated conditions, the tribological performance ranges from severe oxidative to mild adhesive wear (157).

One issue related to synthetic lubricants is the compatibility with elastomers. PAO-based synthetic oils with different additives are investigated based on a new method for lubricant-elastomer compatibility. It is revealed that static tests give an indication of possible chemically active additives, but the dynamic test is necessary for the simulation of real application conditions. Test results are dependent not only on the oil but as well as on the additive combination (158).

Ethers

Ether-type synthetic oils show outstanding characteristics, especially for certain special applications where their excellent heat resistance, oxidation, and radiation resistance and low vapor pressures are of importance. This makes them suitable for high-temperature and high-vacuum applications, and for locations subjected to radiation (144,159). This particularly concerns polyphenyl ethers, which are used as lubricants in extreme environments such as nuclear power plants, space satellites, nuclear submarines, food sterilization equipment, radiation chemistry laboratories, and medical imaging systems. Besides liquid lubricants, they are commercially available as greases (160). Ether-based lubricants may be produced from renewable biomass-derived feedstocks, for example, through direct or reductive etherification of pyrolysis oils, plant, and algal oils, or C5–C6 sugars (161).

The lubrication properties and mechanisms of ethers, compared to various other oxygenated compounds, for example, ketone, alcohols, and esters, are investigated in detail to study the effects of the functional group, carbon-chain length, and ambient humidity on the lubricity of the oxygenated compounds. For this purpose, the high-frequency reciprocating rig is used to measure the lubricity, and microscopic observation of disk specimens with a scanning electron microscope is done. The significant effect of the functional group on the enhancement of lubricity is shown. However, the increase in ambient humidity worsens the lubricity of the oxygenated compounds since the water absorbed by the oxygenated compounds tends to compete with the lubricating compounds in the adsorption process and prevents the formation of the lubricating film. This leads in the case of oxygenated compounds with hydroxyl, ester, and ether groups to abrasive wear with corrosion (162).

Waste

There are different types of waste which can be used as feedstock for sustainable lubricant production from the viewpoint of circular economy even though they might not be from a bio-derived source. The following

section gives an overview over the main approaches for the reuse of products that are normally being discarded.

Waste cooking oil (WCO)

Due to the growing world population, the increasing demand for food led to a higher production of kitchen waste. Among other things, WCO and grease pose a problem, as these wastes have a low degradability, commonly enter the environment due to improper disposal and, hence, result in water pollution and sewer clogging. However, they are produced in large quantities in households, restaurants or in the industry. To overcome their disposal problems, scientists work on its conversion into cheap, easily available, and eco-friendly biolubricants. Besides the growing interest in new applications such as value-added product development leading to financial benefits, actions are taken to overcome the mismanagement of WCOs and to meet public policies (163–165). In Europe (EU + UK), almost all the collected WCOs (0.7–1.2 million tons/year) and additionally a high amount of imported ones are used for biodiesel production, being the third most used feedstock with a share of approx. 19% (2.8 million tons), after rapeseed oil (37%) and palm oil (30%) (166).

WCOs consist of triglycerides, which can be chemically modified to enhance their lubrication performance, for example, make them more stable against oxidation. Figure 3 shows possible chemical modification pathways for the triglycerides such as (trans)esterification, estolide formation, epoxidation, and further ring-opening and acetylation reactions to improve the physicochemical properties (163).

Water extraction of contamination of the crude WCO is a common pre-treatment step within WCO recycling methodologies. The condition of this procedure is crucial as it has a great influence on the quality of the resulting oil. Thus, the analytical determination of optimum conditions is of great interest (167). Edible sunflower oils in fresh and used condition as well as after four different recycling treatments with different pH and temperature are compared to find the best procedure for natural regenerated oleic acid bases. Based on their data and available data from literature, they design a prototype recycling machine for WCO with the possibility to tune the resulting oil depending on pH and temperature (168). Moreover, they successfully use multivariate surface responding analysis (SRA) to find optimum conditions for the water treatment with special regard to density and flash point temperature (167).

A promising application of WCO is the production of environmentally friendly greases. It is possible to produce biodegradable greases from used frying oils in

optimal concentrations of 10–15 wt.% in the base oil exhibiting the same quality level as greases from fresh rapeseed oil. However, upscaling or commercializing is difficult as the composition and condition of the used oils highly depend on its source and usage (169). Greases formulated with WCO and spent bleaching earth (solid waste generated from the bleaching process in the palm oil industry) possess promising performance characteristics for future utilization (170).

Additional research is done on the development of new processes for the transformation of waste into value-added products. A new green and efficient method for preparing octylated branched biolubricants from WCO is presented by hydrolyzation to unsaturated fatty acids with subsequent esterification, epoxidation, and nucleophilic reaction. They obtained a product with excellent lubricant properties in terms of a low pour point, high viscosity index, high thermo-oxidative stability, and enhanced lubricity in a high-frequency reciprocating rig (HFRR) (171).

As mentioned, besides chemical and process considerations, the tribological performance of resulting compounds has to be sufficient. Investigating the tribological performance of mixtures of soybean oil and used frying oil proves that these mixtures are promising candidates for eco-friendly lubricants accompanied by low friction coefficients in boundary and/or mixed lubrication regimes. Especially with the addition of additives, their lubrication performance can be further enhanced (164). Introducing the green synthesis of pentanal-derived alkyl ester dioxolanes with used frying oil as feedstock and determination of density, viscosity, TAN, TBN, and IV (iodine value) prove methyl 8-(2-butyl-5-octyl-1,3-dioxolan-4-yl)octanoate as promising biolubricant candidate (172).

However, there are some issues that need to be overcome for enabling industrial implementation. For example, the supply chain plays a significant role and requires the deployment of effective policies and regulated practices for WCOs recycling and collecting. It has to be ensured that the consumed resources for recycling do not surpass those from the obtained WCOs (e.g. transportation routes). A second challenge of WCOs reuse is their highly heterogeneous nature exhibiting a large variability in physicochemical and sensory properties and significant amounts of various impurities (165).

Waste lubricant oil (WLO)

Lubricants are either consumed during mechanical processing or partly lost due to, for example, evaporation, combustion, but about 50% of the lubricant remains as waste oil (173). About 97% of all lubricants are

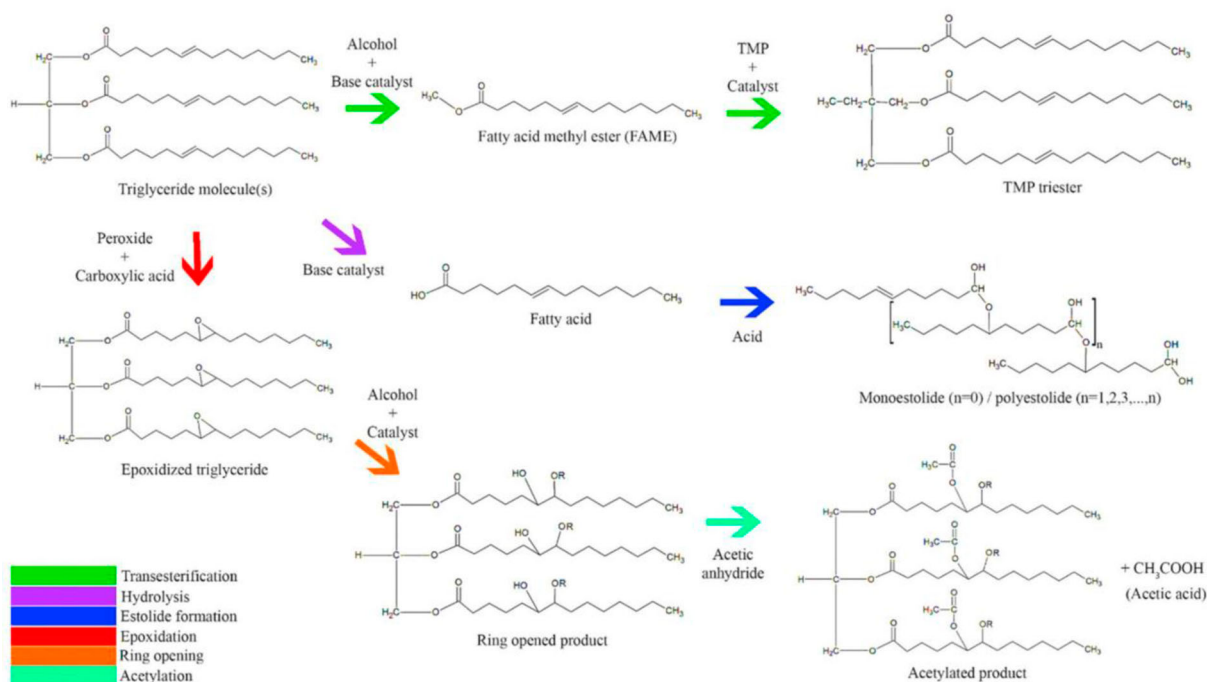


Figure 3. Different chemical reaction pathways for the modification of triglycerides. With permission from (163).

originating from mineral-based oils, being considered as hazardous waste after use (174). WLOs are usually toxic and non-biodegradable, contaminating soil, water, and air, and therefore, causing serious problems for the ecosystem, when not properly disposed (173). Though WLOs are not considered as biofriendly lubricants, they can be recycled instead of just disposing them, which increases their sustainability. This cannot only be seen as a more environmentally friendly approach but can also be applied to WLOs from bio-derived sources in the future.

Depending on their field of application, in Europe, WLOs are laundered, reclaimed, regenerated, burned, mildly or severely reprocessed, and thermally cracked, for energy recovery or re-oil processing purposes. Regeneration of WLOs can lead to lubricating oils with similar properties as the virgin lube base oils when cleaned from contaminants, oxidation products, and additives (173).

Diverse methods are proposed to produce recycled lubricants in the required quality, for example, when discussing the removal of organic compounds, including alkanes, aliphatic amines, alcohols, and aromatics, from waste lubricant oil distillate with microbubble-ozonation with elevated temperature and stable pH (175). A green way to recover base oil from truck engines by applying mechanical stirring and ultrasound is presented as promising recovery yields could be proven, showing an effective removal of Ca, Mg, Na, and Zn and almost 100% elimination of Al, Cr, Fe, and Mo (176).

There are various possibilities in the reuse of WLOs, for example:

- Production of lubricating greases: Rheological and physicochemical properties of greases based on used motor oils are studied while substances interfering with the grease structure are successfully removed resulting in a quality base stock comparable with commercial base oils (177).
- Production of base oils: used mineral-based lubricating oils are recovered by extraction with polar organic solvents. The physicochemical and thermal properties of the obtained oils are further investigated, such as density, viscosity, ash content, acid number, and thermal and oxidative decomposition profiles (178).

However, also life cycle assessments have to be taken into account. Various WLO management scenarios are evaluated for future implementation in Serbia. They investigate the re-refining of used oil for the recovery of base oils, WLO as a substitute for fossil fuels in cement kilns, and combusting WLOs in waste incinerators with energy recovery. It is found that no single WLO application meets all desired requirements. Moreover, they propose four possible WLO management scenarios with an anticipated annual saving of up to 22,100 t CO₂ equivalent and 34,300 t oils equivalent which would contribute to the sustainable industry development (179).

Industrial waste

The demand for recycling of industrial wastes, for example, in terms of recovery of valuable materials like hydrocarbons, that can be upgraded into fuels, base oil, and greases, is not only rising on the governmental side, but also from society (180). In general, waste minimization, disposal, and environmental impact are the most important new factors, which are shaping the market (144).

Research is done on various types of waste processing. For example, food processing side streams from confectioneries and wheat mills are used for their further fermentation with yeast and fungi to produce microbial oil. The various physicochemical properties of these microbial oils are measured, such as pour point, cloud point, acid value, viscosity, and oxidative stability, illustrating their suitability as a sustainable feedstock for biolubricant production (181). The production of lubricants from industrial waste contains high amounts of sulfur when using polyaddition and polycondensation methods (182).

Agro-industrial waste

One drawback of biolubricants from plant-based feedstock is the fact that they might directly compete with agricultural and food production, but the use of agro-industrial waste could solve this problem (183). The following section presents examples of agro-industrial wastes which can be converted into value-added products.

For example, Cardanol, the main component of cashew nutshell liquid (CNSL), consists of phenolic lipids containing mostly unsaturated aliphatic sidechain. Its amphiphilic structure makes it an alternative lubricant, as the polar group adheres to metallic surfaces leading to a monolayer formation effectively reducing friction and wear by reducing the metallic surface contact. The production of CNSL is estimated to be approximately 1×10^6 tons. This makes CNSL when using a green and efficient synthesis, a biodegradable and low-cost lubricant alternative. Tribological tests of Cardanol indicate good performance in the boundary lubrication regime showing COF values below 0.2 (183).

Extracted coffee waste oils are proposed as alternative raw material for biolubricant production as with 8.9 billion kg of coffee produced in 2016 it is worldwide the most popular beverage. To efficiently use the coffee waste, oil is removed and converted to biofuel products (biomass, biogas, etc.). Furthermore, hydrolysis can be used to convert waste coffee oils to fatty acids (184). Oil extracted from waste coffee grounds is tested as lubricant base oil and as a 5% additive in PAO, leading

to significantly reduced COF values compared to pure PAO and showing very promising results as a possible friction modifier additive (185).

Natural wax from one of the most widely cultivated medicinal plants in Gansu Province *Codonopsis pilosula* is investigated. Tribological tests indicate that the natural wax shows excellent lubricity in a load range from 100 to 250 N and a temperature range of 25–150°C, which is comparable to synthetic PAO 8 oil and plant oils, like hydrotreated rapeseed oil (186).

Litsea Cubeba kernels are waste products of essential oil production from litsea cubeba. By applying litsea cubeba kernel oil as feedstock trimethylolpropane fatty acid triester (TFATE) could be synthesized as biolubricant base oil. They found that the obtained TFATE possesses promising properties as an alternative biolubricant feedstock due to its good oxidative stability and low-temperature performance (187).

Lastly, supercritical extraction of waxes from sugarcane waste (leaves, rind, and bagasse) is proposed as an alternative biolubricant but tribological tests are not performed (188).

Waste of animal origin

The lubricity of conventional synthetic lubricants is not only compared to these from agro-industrial waste, mainly of plant-based origin but also to lubes from the waste of animal origin (189). In contrast to plant-based lubricants, lubricants of animal origin are completely water insoluble as they are derived from animal fats and therefore hydrophobic. These animal fats include hard fats (i.e. stearin) and soft fats (i.e. lard) and are structurally composed of triglycerides (glyceryl esters of fatty acids) (189). Possible sources of animal waste are wide-ranging.

Two fresh commercial synthetic cutting oil emulsions against a new metalworking bio-based oil derived from slaughtering waste are assessed. Focus is laid on the impact of the lubricants on the emulsion treatment of metal working wastewater. In these terms, best results are obtained with the emulsion produced with the bio-based lubricant, however, tribological tests are not performed (189).

Bio-paraffin production by special hydrocracking of pre-treated waste lard from processing plant represents an alternative to synthetic lubricants and can be further used in versatile ways. During the treatment of waste lard, only 2–4% of secondary waste is formed and even this waste can be used as feedstock for biogas production (190).

Furthermore, fish oil residue is considered as non-edible waste and a possible source of biolubricant production. The non-edible waste caused by the fish

industry is estimated to 5.2 million tons per year. Lipids extracted from these fish oil wastes, with good acid values and oxidation parameters, are successfully converted into trimethylpropane fatty esters with kinematic viscosities in the ISO VG 22 range (191).

The tribological properties of unmodified and modified synthesized beef tallow grease are studied. The effects of modifications, such as antioxidant additives, anti-wear additives, waste PET polymer compound, and thermally processed graphite additive, are studied on the synthesized beef tallow. Tribological tests are carried out using a four-ball geometry proving that synthesized beef tallow modified with 20% of PET waste and anti-wear additives shows enhanced friction and wear properties. Additionally, adding thermally processed graphite to the grease compound shows improved tribological performance (192).

The physical and chemical properties of esterified housefly larva lipids are studied for the application as biolubricants. Tribological tests are carried out according to ASTM D4172-94(2010) for measuring the wear scar diameter (WSD) of the gained 2-ethylhexyl fatty acid esters indicating good anti-wear properties of the ester. Assessment of typical oil parameters proves promising chemical properties of the esters for biolubricant base oil utilization (193).

Lubricant additives derived from poultry chicken feathers are showing good antioxidant characteristics and anti-corrosion performance in polyol as the base oil, but only moderate anti-wear potential which using a four-ball tester (194).

Plastic waste

More than 75% of annually produced plastic materials are discarded as single-use plastics, ending up in landfills, polluting the environment through uncontrolled discharge, or incinerated for electricity production (195). Conventional plastics are non-biodegradable and accumulative in the environment, posing a serious threat to any species at a global scale. Thus, the conversion of plastic waste into value-added products, such as lubricants, is of high interest in order to enforce circular economy principles and to support sustainable industry developments. Some process conversion strategies and the resulting products are discussed in the following section.

Focusing on the chemical upcycling of polyethylene by hydrogenolysis via Pt/SrTiO₃-catalyst conversion leads to a break-up of long hydrocarbon chains into narrowly distributed oligomeric chains, and therefore, to high-quality liquid products, such as lubricants and waxes (195).

Plastic waste is treated and converted into lubricant additives via copolymerization for engine oil enhancement. More specific, the change in physicochemical properties of three synthetic esters created from polypropylene, high-density polyethylene (HDPE), and low-density polyethylene (LDPE) is studied, when added to different mineral base oils. Polypropylene ester as a lubricant additive shows the best impact on viscosity index and pour point (196).

Recycled polypropylene and lubricant oil are used for synthetic bitumen formulation, as an alternative to the common disposal of these materials. Rheological tests, such as dynamic oscillatory, creep-recovery, and shear steady tests, are carried out in parallel-plate geometry with a frequency range of 0.01–100 rad/s, 1 Pa constant stress (linear viscoelastic region), and temperatures between 25°C and 150°C (197).

Alternative high-quality liquid lubricants (HQL) originating from pre- and post-consumer polyolefin waste are investigated regarding their tribological behavior. They found that HQLs exhibited similar performance to synthetic base oils (e.g. PAOs) in terms of wear scar volume and even a superior performance compared to Group III mineral oils. Moreover, they discovered synergistic effects on friction and wear by blending the upcycled plastic lubricant with synthetic mineral oils. Life cycle assessments and techno-economic analyses prove that this novel technology represents a cost-saving method to reduce the detrimental environmental impact of plastic waste (198).

A comparative study on the hydroconversion of polyolefin plastic waste to fuels, lubricants, and waxes by hydrogenolysis and hydrocracking of model alkanes to obtain basic knowledge for engineering better catalysts and processes is carried out. A direct comparison of results from different laboratories is highly challenging due to aspects like complex and varying feedstock, impurities, additives, and various processing issues (199).

A method for the selective conversion of polyolefins originating from single-use plastics to fuels is presented, with a process that could be adapted for the conversion of different common plastic wastes as well as composite plastics to fuels and light lubricants (200).

A new route for the chemical recycling of polyethylene from plastic waste to α,ω -divinyl-functionalized oligomers (Figure 4) via a sequence of bromination, dehydrobromination, and olefin metathesis reactions is proposed. By techno-economic assessments, it could be proven that this process could be applied on an industrial scale for upcycling plastic waste into value-added chemicals which can be utilized as feedstock for lubricant synthesis (201).

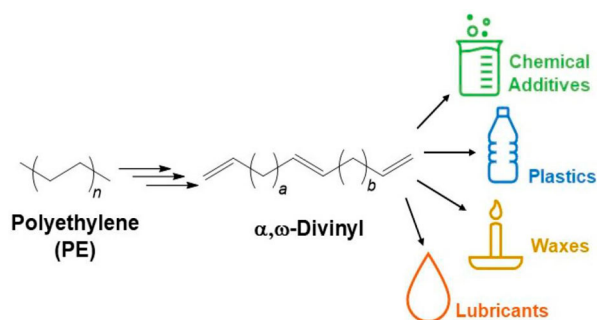


Figure 4. Conversion scheme of polyethylene to α,ω -divinyl-oligomers and possible applications. With permission from (207).

Miscellaneous

Besides the above-mentioned sources to produce sustainable lubricants, there are some special niche feedstocks which can successfully be converted into value-added products.

Kitchen chimney dump lard recycling into biolubricant is investigated along with its rheological and tribological properties. These measurements are focusing on viscosity, wear, and friction behavior, showing no change in specific wear rate (202).

The production of long-chain fatty acids like erucic acid (C22:1) from crude glycerol and waste cooking oil by a recombinant yeast strain is researched. Erucic acid can be further used for lubricant, surfactant, or biodiesel production (203).

In another approach, the synthesis of graphite oxide from waste carbon source (e.g. paper waste) is demonstrated. Using graphite oxide as nano-additive in synthetic engine oil has a beneficial effect on the coefficient of friction and wear properties. Tribological characterization is carried out with synthetic base oil with and without the addition of graphite oxide on a four-ball tester (steel, high carbon, and high chromium alloy) (204).

Converting waste tires into pyrolytic oil is not only providing a possibility for waste tire recycling but also results in an improved oil suitable as base oil for lubricant applications, meeting the standard requirements for commercial lubricant oils (205).

Characterization and condition monitoring

One of the most important characteristics of a lubricant, no matter if manufactured from renewable/natural or of fossil origin, is the maintenance of the desired performance properties (206). Exposure to elevated temperatures, leading to oxidation reactions, and contaminations from environment or machine parts are only some of the stress factors causing degradation (207). Based on the intrinsic properties and the

structure synthetic lubes, for example, esters, can show advantages over mineral-based ones, in particular regarding high-temperature and oxidation stability (143). When it comes to bio-based synthetic lubes, this property may be affected by the presence of a higher degree of double bonds or free fatty acids. Thus, the stability of these products is of high concern and continuous condition monitoring to maintain and prolong the performance is key, which also facilitates proactive measures (additive addition) (145). For this reason, appropriate techniques have to be used to determine the impact of these structural variations on the thermo-oxidative stability and performance of the lubricants.

While routine analytics such as determination of viscosity, determination of water content, or determination of neutralization number can be done accordingly to the standards for mineral oils (206), the oxidation stability methods are often not fully suitable for bio-based synthetic oils and have to be modified, e.g. some tests cannot differentiate between bio-based oils with different oxidation stability (147). Methods inspired by food chemistry or the bio-fuel industry (147,208) are one promising approach, supported by analytical methods which focus not only on the determination of sum parameters, such as oxidation. Detailed analytical characterization of synthetic or bio-based (complex mixtures) lubricants has to deal with the wide variety of chemistries, from synthesized hydrocarbons like polyalphaolefins via esters to polysaccharide or protein-based lubricants. However, the well-defined molecular structure due to synthesis process enables in-depth characterization and, consequently, elaboration of the correlation between structure and thermo-oxidative stability as well as performance and property prediction in different ageing conditions, which is a challenge for bio-based lubricants consisting of complex mixtures (209).

By means of artificial alteration and in-depth analysis, for example, with gas chromatography coupled to mass spectrometry (GC-MS) the impact of the structure of the lubricant-oil components on the alteration behavior can be monitored. This combination of techniques is used to simulate the ongoing reactions under well-defined, reproducible, and variable conditions and to correlate the identified degradation products with the tribological performance (151,208,210,211). Other chromatography techniques, such as liquid chromatography combined with high-resolution MS, enable to study the degradation of synthetic biodegradable lubricants and the used additives even in more extent (212). However, this approach is not sufficient for a comprehensive elucidation of the main pathways of oxidative degradation

and identifying involved molecule types as well as oxidative degradation product formation over time. When it comes to oxygen-containing lubricants, such as synthetic esters, the clear identification of the oxidation reactions is even more challenging.

The use of stable isotopic tracer within an artificial alteration set-up under oxygen pressure and GC-MS enabled to obtain detailed information on the oxidation products of oxygen-containing components. In detail, the approach enables the unambiguous attribution of oxygen atoms in the degradation products and particularly hints to the mechanism of the formation of the degradation products. Hence, the involvement of specific molecule structures in the degradation process can be assigned (149,213).

Conclusions

Challenges and outlook

Heterogeneity of the biological or waste feedstock, even in the same category, compared to petroleum-based feedstock is very challenging for industrialization attempts. Based on the findings outlined above, the following summary on challenges of sustainability for lubricant formulations and the lubricant industry can be drawn:

- Need for thorough characterization and where necessary separation (compounds decreasing stability, corrosive ones, those limiting operational window, for example, waxes, etc.)
- Need for toxicity and biodegradability assessment, not only of the individual constituents but also of the whole part of the feedstock (mixed toxicity effects, for example, synergism) used in the final lubricant formulation
- To have industrial relevance and to be economically viable, it is preferential that the feedstock is compatible with the existing industrial infrastructure, especially for the transition state toward a sustainable industry by using renewable sources to replace crude oil (easiest introduction for molecules resembling the petroleum feedstock, e.g. alkenes)
- When thinking in terms of green chemistry and sustainable lubrication also production pathways and materials (and availability of those), for example, for synthesis, catalysis, etc. must be considered and regulated
- Advanced surface and material design (as a concept of green tribology) will further challenge energy consumption
- Proper disposal and/or recycling of fully formulated lubricant must be ensured → everything is toxic in

large amounts and to certain organisms → biodegradability ensures it does not persist and cause long-term toxicity and it avoids the issue of bioaccumulation. Biodegradability is dependent on the type of organism and ambient conditions.

Not only the feedstock and lubricant product assessment but also the whole lubricant life cycle analyses up to the waste stage is necessary, as compounds of special concern might be detected, and proper disposal measurements are taken (complex issue as application conditions vary affecting the degradation products). The potential for recyclability even for other application(s) than the original should be assessed to limit the need for original resources and to limit the waste production.

Furthermore, in many cases, an adaption of specifications will be necessary, including:

- Common condition monitoring (see Characterization and condition monitoring) tests are optimized for the evaluation of mineral-based lubrication oils and are often not suitable for plant-derived oils (147).
- Biodegradability tests e.g. according to OECD301, a guideline adopted in 1992, define a lubricant as readily biodegradable (and within the EEL criteria), if at least 60% of the formulation is fully biodegraded within a period of 28 days. However, how long it does take for the remaining 40% of the lubricant to fully biodegrade is not regulated (214), which means up to 50% of the formulation can be of petroleum origin with high persistency and bioaccumulation potential. Even with such composition, they would still fulfill the criteria of the EU Ecolabel (215).
- Toxicity values of a substance are strongly varying with the type of organism used and therefore, are not consistently meaningful if not tested comprehensively.

Concluding remarks

The emission of lubricant into the environment via micro drops or oil mist (e.g. in the case of metal-working fluids) is hardly preventable during use and especially accidental spills of petroleum-based lubricants cause a massive negative impact on the environment and human health. For example, contamination of the water surface through oil spills impairs the gas exchange at the water-atmosphere interface and leads to a decrease in oxygen levels in the water, threatening aquatic ecosystems (215).

So, it is already past due, that the focus must shift toward bio-based or bio-derived lubricant components

from renewable sources accompanied by proper eco-hazard tests (biodegradability, bioaccumulation, and toxicity), that imitate common synthetic lubricant structures and serve as intermediates in established industrial scale production (on the way to a sustainable production). For scarce and limited resources (e.g. metals) within the production processes recycling could act as a driver. In general, recycling used lubricants and enforcing a circular economy are necessary not only for harmful substances but also for renewable materials as a matter of sustainable use.


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