

<https://doi.org/10.26599/FRICT.2026.9441302>

Review

Green greases for the future: Biobased materials and emerging gel technologies

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Received: January 9, 2026; Revised: May 10, 2026; Accepted: August 10, 2026

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Abstract:

Lubricating greases are widely used in bearings, gears, and other heavily loaded machine elements, but conventional lithium- and calcium-soap greases based on mineral oils and metal-containing additives raise increasing concerns regarding toxicity, persistence, and regulatory compliance. This review summarises recent progress in green greases, defined as greases that employ renewable or environmentally acceptable base oils, thickeners, and additives to reduce environmental impact while maintaining functional performance. After briefly outlining the

composition and operating principles of traditional greases, we discuss the physicochemical properties, advantages, and limitations of bio-based base oils, with emphasis on vegetable and synthetic esters and on ionic liquids used either as base fluids or co-base oils. We review renewable and biodegradable thickener systems, including modified soaps, polysaccharides, lignin-related structures and mineral or hybrid networks, and relate their molecular structure and gel microstructure to rheology, oil retention and mechanical stability. Particular attention is paid to oleogels, hydrogels and ionogels as emerging gel technologies that enable semi-solid green lubricants. Natural antioxidants, bio-based friction modifiers and environmentally acceptable anti-wear additives are surveyed, and representative formulations are compared with commercial reference greases in terms of friction, wear and other key performance indicators. In addition, green synthesis routes for bio-based gelators and mild, energy-saving processing conditions for grease manufacture are discussed in the context of green-chemistry principles and industrial scalability. On this basis, the environmental performance, application domains and market prospects of green greases are analysed, and the main trade-offs between performance, cost and sustainability are identified. Finally, the remaining challenges, especially high-temperature reliability, long-term stability, standardisation and large-scale implementation, are discussed, and directions are outlined for the rational design and broader adoption of next-generation green greases.

Keywords: Green greases; Bio-based lubricants; Renewable and biodegradable thickeners; Sustainable tribology

1 Introduction

Tribology deals with friction, wear, and lubrication in interacting surfaces and is essential for the reliability and energy efficiency of mechanical systems [1, 2]. Lubricants are introduced between moving surfaces to separate asperities, carry away heat and contaminants, and protect components from corrosion [3, 4]. Depending on their physical form, lubricants are generally classified into three categories: liquid lubricants (oils), semi-solid lubricants (greases), and solid lubricants (such as graphite or molybdenum disulfide) [5]. Among these, lubricating greases are particularly suitable for applications where liquid lubricants fail to remain in place or where a continuous oil supply is impractical [6]. The primary functions of lubricants are to separate contacting surfaces, minimize direct asperity contact, dissipate heat generated during sliding or rolling, and extend component lifetime [3]. In industrial practice, the selection of lubricants depends on operating conditions such as temperature, load, and speed [7]. With increasing demands for energy efficiency and environmental protection, modern lubrication science is also focusing on developing sustainable, biodegradable, and high-performance lubricants to replace traditional petroleum-based systems [8, 9].

Conventional greases are semi-solid lubricants based on petroleum-derived oils and metallic soap thickeners (e.g., lithium, calcium, or aluminum soaps), with functional additives to enhance performance [10]. They have been widely used in industrial and automotive sectors due to their good mechanical stability, wide service temperature range, and effective lubrication properties. Structurally, conventional greases consist of three primary components: base oil, thickener, and additives[11]. As shown in **Figure 1**, the base oil, typically making up 70%-90% of the formulation, provides lubrication, while the thickener, comprising 5%-30%, converts the base oil into a semi-solid structure, giving grease its consistency and adhesion properties [12]. Additives, which generally account for up to 10%, enhance performance by improving oxidation resistance [13, 14], reducing wear [15, 16], and preventing corrosion [17-19].

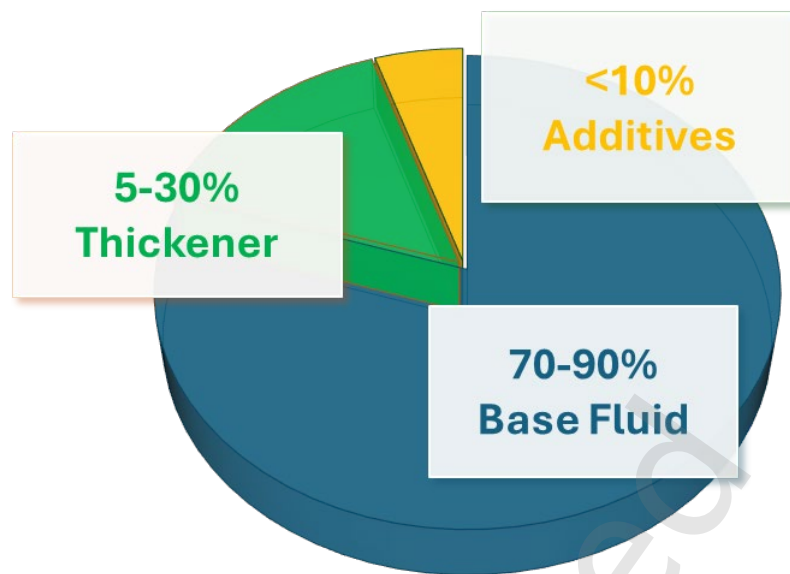


Figure 1 Typical composition of lubricating grease

Unlike lubricating oils, grease has lower fluidity, allowing it to adhere effectively to mechanical components and maintain a stable lubricating film, even in conditions where liquid lubricants may fail [6, 20]. As illustrated in **Figure 2**, its stability is crucial in ensuring consistent performance, as oil separation can reduce lubrication efficiency and accelerate wear [21]. These characteristics make grease indispensable across various industries, including automotive, aerospace, metallurgy, and petrochemicals, where it is used to lubricate high-stress components such as bearings, gear systems, and engines [22, 23]. Although traditional grease formulations have long relied on petroleum-based base oils, metal-containing thickeners, and fossil-derived additives, such formulations raise significant environmental concerns because of pollution risks and the generation of non-biodegradable waste.

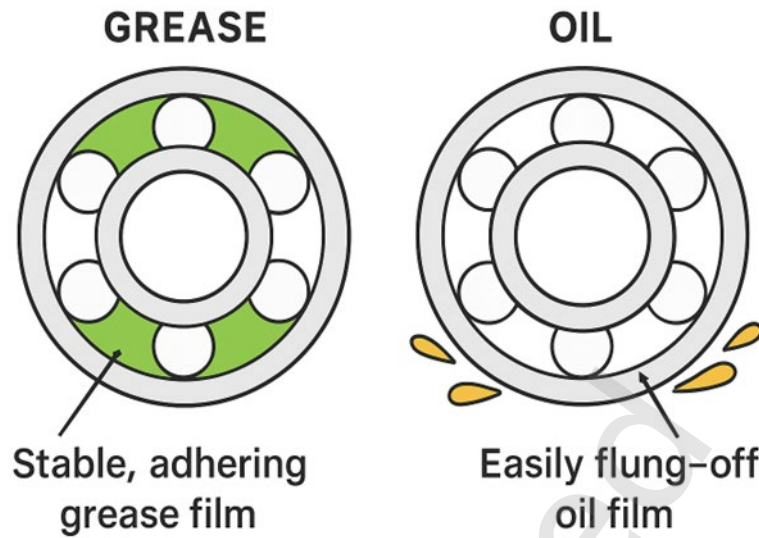


Figure 2 Grease vs. Oil comparison in bearing. Mechanism of grease lubrication in contact

Green greases are increasingly being considered environmentally preferable alternatives to traditional lubricants because they can offer both reduced environmental impact and effective lubrication performance [2, 10]. Formulated with biodegradable base oils, non-toxic thickeners, and eco-friendly additives, it significantly reduces environmental impact without compromising functionality [24, 25]. Unlike traditional mineral oil-based greases, which often contain heavy metals and toxic chemicals, green grease is designed to be safer for both human health and ecological systems. Its biodegradable nature allows it to break down naturally, minimizing long-term pollution, while its non-toxic composition lowers risks associated with handling and disposal [26]. These attributes make it particularly appealing for industries committed to sustainability initiatives. Additionally, green grease retains the key lubricating properties of conventional formulations, enabling its application in demanding environments characterized by extreme temperatures, high speeds, and heavy loads [27, 28]. Beyond providing effective lubrication, it enhances wear protection, corrosion resistance, and overall equipment longevity, reducing maintenance costs and supporting the transition toward greener industrial practices.

Green greases play a vital role in addressing environmental and industrial challenges, providing a sustainable alternative to conventional lubricants. As global awareness of environmental sustainability continues to grow, stringent regulations such as the EU REACH Directive and international ISO standards are driving industries to adopt more environmentally friendly practices [29]. Green grease plays a significant role in reducing pollution and waste, thereby helping to comply with these regulatory frameworks. In addition to environmental benefits, it also enhances industrial sustainability by reducing dependence on fossil fuel-derived resources, lowering greenhouse gas emissions, and promoting the use of renewable and biodegradable materials [30]. Growing demand from emerging industries such as electric vehicles and smart manufacturing has further driven interest in high-performance, low-emission lubricants, making green greases a key player in the evolving market [27, 31-33]. In addition, consumer preferences are shifting towards environmentally friendly products, creating a favorable market for green greases, encouraging continued innovation and investment in their development [34]. In a broader context, the adoption of green greases is consistent with global efforts to mitigate climate change, protect ecosystems, and transition to low-carbon industrial processes.

In the past decade, substantial progress has been made in bio-based lubricants and environmentally acceptable lubricants (EALs). Several comprehensive reviews have summarized advances in bio-based base oils, chemical modification strategies, and eco-friendly additives for oils and hydraulic fluids, as well as sustainability assessments of lubricants at large. These works mainly focus on vegetable and synthetic esters, polyalkylene glycols, and other renewable base fluids, or on additive technologies for EALs, and they only touch on greases as one product form among many [35-37]. In parallel, the production and application of biodegradable lubricating greases based on vegetable oils, polyester or cellulose derivatives, and mineral soaps have been discussed in more specialized articles, yet often with emphasis on chemistries or single performance aspects, such as biodegradability.

However, a dedicated and up-to-date analysis of green greases—here understood as greases formulated with renewable or environmentally benign base fluids, thickeners, and additives that collectively reduce toxicity, persistence, and carbon footprint while maintaining functional performance—is still lacking. Existing reviews rarely address the rapidly growing family of gel-based lubricant technologies, where oleogels, hydrogels, and ionogels are deliberately designed as semi-solid lubricants. Nor do they systematically link molecular structure, gel network architecture and rheological behaviour to tribological and environmental performance across different material classes. As a result, it remains difficult for both researchers and practitioners to compare different green grease concepts on a common basis and to identify robust design principles for future formulations.

This review aims to close these gaps by providing an integrated perspective on green greases for the future. The available raw materials for green grease formulation, such as bio-derived base oils, renewable and biodegradable thickeners, and novel bio-based or low-toxicity additives, are summarized, and their physicochemical properties are examined concerning their influence on grease structure and operational parameters. Second, recent developments are classified according to gel technology—oleogels, hydrogels, and ionogels—and the influence of different network-forming strategies on tribological behaviour and environmental performance is analysed, highlighting common structure–property relationships. Third, green synthesis and processing routes, standardised test methods, and environmental assessment frameworks that are critical for industrial implementation are examined. Finally, key challenges and research needs are identified, and future directions are outlined for the design of green greases that concurrently optimise friction and wear performance, durability, environmental impact, and cost.

It should also be emphasized that a bio-based origin does not automatically guarantee overall sustainability. A grease formulated from renewable feedstocks may still involve energy-intensive synthesis, hazardous intermediates, poor atom economy, limited service life, or unfavorable end-

of-life pathways. Therefore, the sustainability of green greases should be assessed not only in terms of renewable content or biodegradability, but also by considering synthesis route, solvent and energy demand, durability in service, recyclability, and end-of-life management. In this context, life cycle assessment (LCA) provides a more balanced framework for evaluating the true environmental performance of bio-based grease systems.

2 Raw materials for green grease

Base oils constitute 70-90% of grease formulations and significantly influence lubricating properties. Plant oils, synthetic esters, and ionic liquids (ILs) have emerged as viable green alternatives to petroleum-based base oils. In a grease-lubricated contact, the base oil is stored in the thickener network and gradually released as bleed oil to form the lubricating film, while thickener particles accumulate around the contact and form a deposited layer that helps retain the oil, as schematically illustrated in **Figure 3**.

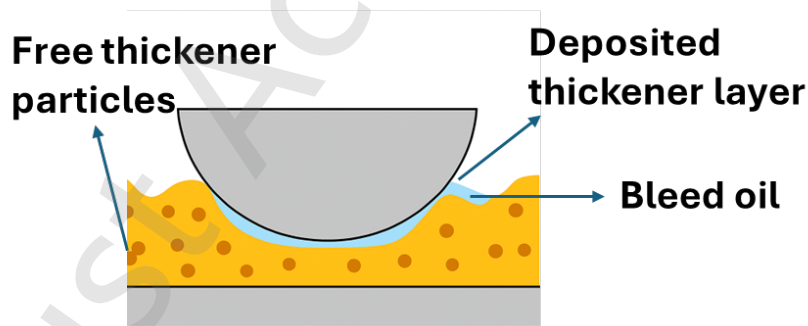


Figure 3 Schematic illustration of grease lubrication in concentrated contact. The thickener network acts as a reservoir for the base oil, which bleeds out to form a lubricating film between the surfaces, while free thickener particles and a deposited thickener layer accumulate around the contact and assist in film replenishment and retention.

2.1 Sustainable and bio-based base fluids

2.1.1 Plant-derived oils

Plant oils have been extensively investigated as sustainable base oils in green grease formulations due to their biodegradability, renewability, and favourable tribological properties. Unlike

petroleum-based oils, which are non-renewable and environmentally persistent, plant oils offer a bio-based alternative with lower ecotoxicity and high biodegradability, often exceeding 70% degradation within 28 days under OECD 301C test conditions [38, 39]. These characteristics make them ideal candidates for applications in ecologically sensitive areas such as agriculture, forestry, and marine systems [39].

Chemically, plant oils are composed of triglycerides containing varying ratios of saturated and unsaturated fatty acids [40]. This structural composition imparts high viscosity indices (typically 180-220) and excellent lubricity, which are advantageous for maintaining oil film thickness under a wide range of temperatures and loads [41]. The polar ester groups in vegetable oils enhance metal surface affinity, contributing to superior boundary lubrication [42].

Regarding tribological performance, plant oil-based greases—especially those formulated with lithium soap thickeners—have demonstrated comparable results to their mineral oil-based counterparts. A recent study by Singh et al. showed that jojoba oil-based grease enhanced with carbon-based nano additives achieved a 38% reduction in friction and up to 60% improvement in load-carrying capacity in rolling bearing tests, highlighting the synergistic effect of polar esters and nanostructured additives [43].

However, a significant limitation of unmodified vegetable oils lies in their poor oxidative stability, primarily due to the presence of polyunsaturated fatty acids [44]. Exposure to oxygen, elevated temperatures, and catalytic metal surfaces can lead to the formation of peroxides, sludge, and varnish, compromising long-term lubrication performance [45]. According to Erhan et al., the oxidation induction time (OIT) of unmodified soybean oil is significantly shorter than that of synthetic esters; however, its oxidative resistance can be improved through chemical hydrogenation or the addition of antioxidant packages [46]. For example, OIT measurements at 160 °C show that while unmodified soybean oil has an OIT of approximately 12 minutes, its

epoxidized and hydrogenated counterparts exhibit OITs exceeding 60 minutes, indicating a substantial enhancement in thermal and oxidative stability [47].

To facilitate a comparison between different plant-derived base oils used in green greases, their key physicochemical properties are summarised in **Table 1**. All the oils listed exhibit high inherent biodegradability and generally good lubricity, yet they differ markedly in viscosity index and oxidation stability, which strongly influence the operating window of the corresponding greases. Castor oil-based greases benefit from high polarity and good lubricity but have a relatively low viscosity index (~150) and poor oxidation stability, restricting their use to moderate temperatures [48-50]. Rapeseed and sunflower oils offer higher viscosity indices (~200–210) and moderate oxidation stability, representing a compromise between flow behaviour and durability [51, 52]. Jojoba oil combines excellent lubricity with the highest viscosity index (~240) and high oxidation stability, making it attractive for applications that require a wide service-temperature range, albeit at a higher cost [43]. For palm and mustard oils, viscosity indices are less well documented, but their oxidation stability is generally rated as moderate [53-55]. Overall, **Table 1** illustrates that the selection of plant oils for green grease formulations must balance biodegradability and lubricity with viscosity–temperature behaviour and resistance to oxidation, rather than being guided by renewability alone.

Table 1 Summary of Physicochemical Properties of Greases Based on Common Plant Oils

Base oil	Base oil's Biodegradability	Lubricity	Viscosity Index	Oxidation Stability	Reference
Castor oil	High	Good	~150	Low	[48-50]
Palm oil	High	Good	NR	Moderate	[53, 54]
Rapeseed oil	High	Good	~210	Moderate	[52]
Jojoba oil	High	Excellent	~240	High	[43]
Sunflower oil	High	Good	~200	Moderate	[51]
Mustard oil (WCO)	High	Good	NR	Moderate	[55]

Note: NR = Not reported in the original reference.

Overall, plant-derived oils offer an attractive combination of high viscosity index, strong polarity and excellent lubricity, but their high degree of unsaturation limits thermal and oxidative stability. For grease formulation, saturated or moderately unsaturated esters (e.g., high-oleic grades) and chemically modified triglycerides therefore provide a better balance between low-temperature flow and resistance to oxidation than highly polyunsaturated oils. The presence of polar functional groups is beneficial for boundary lubrication and adsorption on metal surfaces, but needs to be complemented by suitable antioxidants and, where necessary, partial hydrogenation or epoxidation to suppress oxidative degradation. In practice, selecting plant oils for green greases should focus on tailored fatty-acid profiles, good inherent oxidative stability, and compatibility with bio-based thickeners and additives, rather than relying solely on renewability.

2.1.2 Ionic Liquids: Sustainable Functional Fluids

While plant-derived oils represent the "green-by-origin" approach, Ionic Liquids (ILs) contribute to green lubrication through "green-by-design" principles, specifically via pollution prevention and energy efficiency. Characterized by negligible vapor pressure and non-flammability, ILs offer a distinct environmental advantage over mineral oils by virtually eliminating volatile organic compound (VOC) emissions during operation [8, 56].

From a green chemistry perspective, ILs contribute to sustainability in several key ways. First, their extremely low volatility virtually eliminates evaporative emissions, reducing air pollution and workplace exposure risks [57]. Second, many ILs-especially those based on non-halogenated and phosphonium or borate chemistries-can be synthesized from less hazardous, potentially bio-derived precursors, making them compatible with environmentally conscious design strategies.

In recent studies, ILs have been successfully incorporated into lithium- and polymer-thickened greases. These formulations demonstrated not only excellent tribological performance, but also a reduced need for traditional anti-wear additives such as ZDDP or MoS₂. For example, de la Presilla et al. reported that greases containing 10 wt% of trihexyl(tetradecyl)phosphonium

bis(mandelato)borate ([P₆₆₆₁₄][BMB]) provided superior protection in fretting tests, maintaining low friction and minimal wear across 1000 cycles [58]. Importantly, this formulation achieved these results without relying on environmentally concerning metallic additives.

Tribologically, IL-based greases have shown superior anti-wear and friction-reducing properties compared with conventional mineral-oil-based greases. Mozes et al. evaluated greases formulated with [BMIM][TFSI] and [P6,6,6,14][TFSI] and observed better elastic modulus, shear-thinning behavior, and thermal stability under oscillatory shear, together with excellent metal surface wetting, which is crucial for maintaining film integrity under dynamic conditions [59]. Similarly, Fan et al. reported that lithium-based ILs such as [Li(G2)] NTf₂ and [Li(G4)] NTf₂ outperformed commercial greases in wear resistance, particularly under high load conditions (e.g., 150 N), while also offering electrical conductivity advantages for greases used in electromechanical systems [60]. In addition to enhancing lubrication performance, ILs can reduce the total chemical additive content in grease formulations due to their multifunctionality. Acting both as base oil and as surface-active agents, ILs can form robust boundary lubrication films, thus decreasing wear and friction while extending mechanical component lifetimes. This dual functionality not only simplifies formulation but also reduces the additive load, which further contributes to the environmental friendliness of the grease system. Recent advances further demonstrate that supramolecular ionogels derived from imidazolium-based ILs and urea-functionalized gelators exhibit enhanced tribological behavior due to synergistic hydrogen bonding and hydrophobic effects. Yu et al. reported that these ionogels maintain stable low friction coefficients across a wide temperature range (−50 to 50 °C), outperforming neat ILs under the same conditions (**Figure 4**) [61].

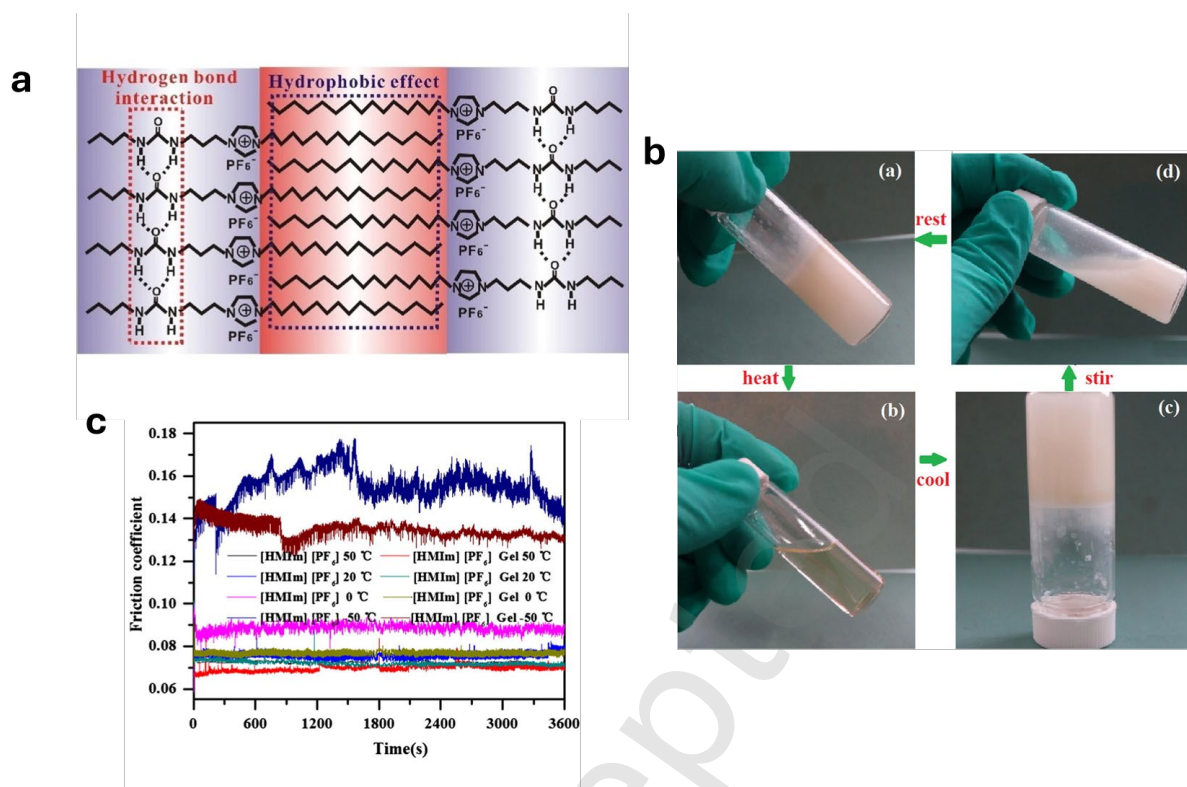


Figure 4 a) Schematic illustration of hydrogen bonding and hydrophobic interactions in supramolecular ionogels; (b) friction coefficient evolution of [HMIm][PF₆] and ionogels with gelators across temperatures; (c) thermoreversible gel–sol transitions based on self-assembly under heating, cooling, and stirring. Adapted from Yu et al [61].

While the molecular tunability of ILs is advantageous, a rigorous distinction must be drawn between "performance benefits" (e.g., low volatility) and "environmental safety" [62]. Early-generation ILs containing halogenated anions (e.g., [BF₄]⁻, [PF₆]⁻) or fluorinated groups (e.g., [NTf₂]⁻), despite their excellent tribological stability, pose significant ecological risks due to their resistance to biodegradation and potential hydrolysis into toxic hydrofluoric acid. Consequently, for an IL-based grease to be genuinely classified as "green," it should preferably employ halogen-free chemistries, such as phosphate-, borate-, carboxylate-, choline-, or amino-acid-based systems. Furthermore, the industrial viability of ILs as full base fluids (70-90 wt% of grease) faces steep economic hurdles. The synthesis of high-purity ILs often involves complex, multi-step reactions with poor atom economy and high energy consumption compared to the refining of vegetable oils

or synthetic esters. This creates a substantial cost disparity that currently limits the scalability of 100% IL-based green greases [63, 64]. Therefore, a more pragmatic pathway for the near future lies in using functionalized, halogen-free ILs as high-performance additives (at 1-5 wt% loading) within bio-based ester/hydrocarbon matrices, or as strategic phases in hybrid greases, rather than as bulk replacements for base oils. Future research must prioritize not only the tribology of these fluids but also Life Cycle Assessments (LCA) that quantify the trade-off between their synthesis footprint and their operational energy savings.

2.2 Renewable and biodegradable thickeners

Renewable and biodegradable thickeners have gained attention as sustainable alternatives to traditional metallic soap thickeners in lubricating greases [65]. Among the most studied are cellulose derivatives and chitosan-based materials.

Cellulose and its derivatives, such as ethyl cellulose, regenerated cellulose, and kraft pulp cellulose, are widely studied renewable and biodegradable thickeners. Their strong hydrogen bonding ability and network-forming capacity enable them to gel various vegetable oils. Cellulose pulp and ethyl cellulose have been successfully used to structure castor oil, producing greases with stable rheology and thermal resistance, especially when blended [66]. Regenerated nanocellulose has also shown promise, forming medium-consistency greases in polar base oils with excellent anti-wear performance at just 7 wt% concentration [67]. In this area, the group led by Prof. Franco at the University of Huelva has made particularly important contributions to the development of castor-oil-based green greases structured with cellulose derivatives and related biogenic thickeners. Their work helped establish key relationships between thickener chemistry, grease rheology, oil retention, and tribological behavior, and provided some of the early systematic demonstrations that renewable polymer-based thickener systems could serve as viable alternatives to conventional soap-based greases. [49, 66, 68, 69]

Chitosan, derived from deacetylated chitin, has also been explored as a thickener in vegetable oil greases. Despite higher friction coefficients observed in some formulations, especially under rotational tribological testing, its biodegradability and compatibility with acidic oils such as estolides and castor oil make it an attractive candidate[69]. The acylation of chitosan further enhances its dispersibility in oils, and greases thickened with acylated chitosan have shown lower friction in oscillatory mode tribological tests than traditional lithium-based greases[68].

Renewable and biodegradable thickeners such as cellulose derivatives, chitosan-based materials and small-molecule gelators have demonstrated that renewable networks can match or even surpass metal soaps in terms of oil binding and load-carrying capacity. Fibrous biopolymers provide tunable, shear-thinning structures but can suffer from limited high-temperature stability, whereas low-molecular-weight gelators offer very low friction at modest concentrations at the expense of more complex synthesis and sometimes higher cost. Effective design of green thickener systems therefore, hinges on balancing network density, fibre or fibril aspect ratio, and chemical affinity to the base oil to achieve sufficient yield stress, rapid structure recovery, and acceptable thermal stability. Hybrid networks that combine bio-thickeners with a small fraction of inorganic or polymeric reinforcements appear particularly promising for extending the operating window of green greases.

3 Green Additives in Lubricating Greases

In the development of green grease, the selection of functional additives is crucial. Compared with traditional additives (such as ZDDP, aromatic amines, and sulfur/phosphorus compounds), the additives discussed here are typically renewable, biodegradable, and/or low in toxicity [70]. They can improve lubrication performance and reduce the environmental burden. These environmentally preferable additives can be divided into four categories: natural antioxidants, environmentally acceptable extreme-pressure/anti-wear agents, bio-based friction modifiers, and green corrosion inhibitors.

3.1 Natural antioxidants

Oxidation is one of the main causes of grease failure. Traditional antioxidants such as butylated hydroxytoluene (BHT) [71], alkylated phenol, dithiophosphoric acid ester, and diphenylamine (APDD), butylated hydroxyanisole (BHA), and aromatic amine compounds are effective but have bioaccumulation and ecotoxicity issues. Natural antioxidants provide an environmentally friendly alternative.

3.1.1 Tocopherols, Polyphenols, and Gallic Acid

Plant-derived polyphenols such as tea polyphenols and gallic acid exhibit strong radical scavenging abilities. Tian et al. demonstrated that the incorporation of 2 wt% gallic acid into a soybean oil-based grease increased OIT by 353%, surpassing the performance of synthetic antioxidants like TBHQ. Tea polyphenols and α -tocopherol also significantly enhanced OIT by 231% and 93%, respectively. When used together, gallic acid and tocopherol exhibited a synergistic antioxidative effect, improving OIT by 454% compared to the base formulation [72], as illustrated in **Figure 5**.

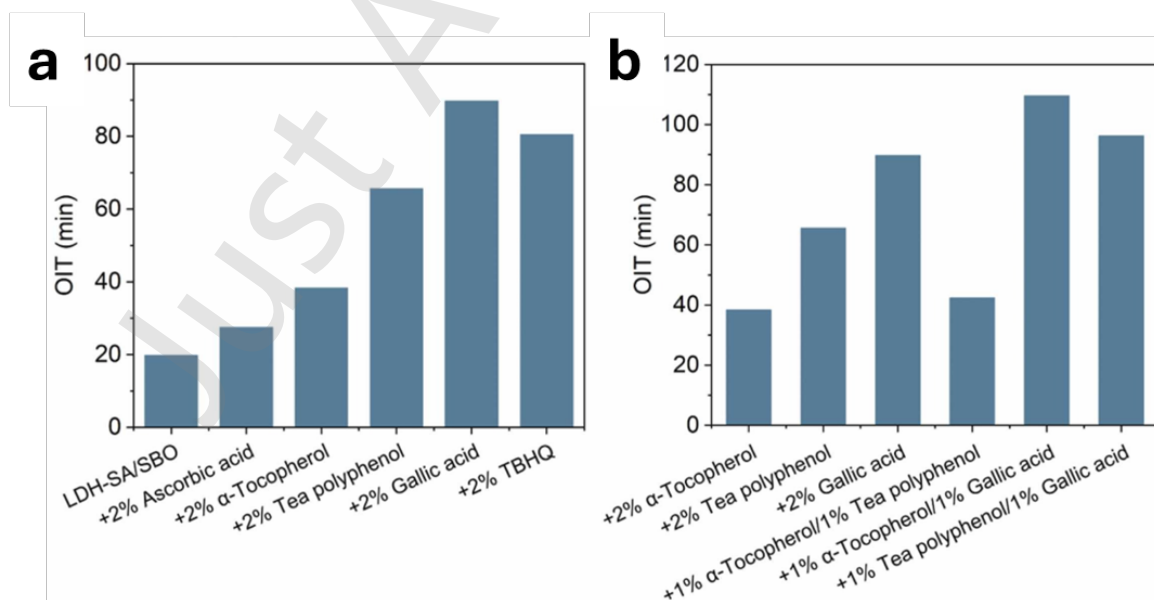


Figure 5 Effect of natural antioxidants and their combinations on the oxidation induction time (OIT) of soybean-oil-based greases. (a) OIT of greases containing 2 wt% α -tocopherol, tea

polyphenols, gallic acid or TBHQ. (b) OIT of greases containing combinations of α -tocopherol, tea polyphenols and gallic acid at 1-2 wt%. Adapted from ref. [72] with permission.

3.1.2 Lignin-based antioxidants

Lignin, a natural aromatic polymer rich in phenolic hydroxyl groups, has emerged as an effective green antioxidant. Wu et al. developed a lignin–castor oil-based oleogel grease that achieved an OIT of 1959 s without any synthetic antioxidant, compared to just 20 s for pure castor oil—a 98-fold improvement. This enhancement was attributed to the radical-trapping capability of lignin's hydroxyl groups, which terminate oxidation chain reactions. Notably, the lignin-based grease also exhibited strong tribological performance and was composed of over 80% renewable biomass, fulfilling both functional and sustainability goals [73].

3.1.3 Acylated chitosan

Chitosan, a natural polysaccharide derived from chitin, can be chemically modified to improve oil dispersibility and antioxidation. González et al. reported that castor oil-based greases structured with N-acylated chitosan and 1 wt% propyl gallate increased the oxidation onset temperature (OOT) by 66.5%, from 142.1 °C to 236.6 °C. This performance was superior to that of ascorbyl palmitate (OOT = 189.4 °C), indicating the efficacy of combining lipid-compatible antioxidants with biopolymer-based thickeners [74].

Taken together, plant-derived polyphenols, lignin-based antioxidants, and acylated chitosan show that effective oxidation control can be achieved with fully bio-based chemistries. Multi-phenolic structures provide strong radical-scavenging capability, while polymeric backbones such as lignin or chitosan improve thermal stability and compatibility with thickener networks. Compared to conventional amine or phenolic antioxidants, these natural systems can simultaneously enhance oxidative induction time and lower the overall toxicity and persistence of the grease. When designing antioxidant packages for green greases, it is therefore advantageous to combine low-

molecular-weight polyphenols for fast radical trapping with macromolecular phenolics that anchor in the grease matrix, considering solubility, colour, and long-term stability.

3.2 Bio-based friction modifiers

Friction modifiers play a critical role in lubricating grease formulations by reducing frictional losses, improving energy efficiency, and maintaining surface integrity. In the context of environmentally friendly lubrication, green friction modifiers—typically derived from bio-based, biodegradable, or non-toxic materials—are gaining momentum as sustainable alternatives to conventional additives such as graphite, molybdenum disulfide, and long-chain fatty amines.

3.2.1 Bio-based fatty acids: oleic acid

Oleic acid (OA), a naturally occurring unsaturated fatty acid, has been widely used as a green friction modifier due to its amphiphilic structure. The polar carboxylic head allows it to adsorb onto metal surfaces, while the long nonpolar alkyl tail promotes stable film formation. Saini et al. demonstrated that OA effectively reduces friction in CaCO_3 -based greases under boundary lubrication conditions. OA facilitated the dispersion of nanoparticles and contributed to the formation of a compact lubricating film, resulting in a friction reduction of approximately 30% under 20 N load conditions [75].

3.2.2 Ionic liquids as friction modifiers

Ionic liquids (ILs) are a versatile class of compounds increasingly explored as green multifunctional additives in grease formulations. Phosphonium-based ILs, such as trihexyl(tetradecyl)phosphonium bis(2-ethylhexyl) phosphate ($[\text{P}_{66614}][\text{BEHP}]$), have shown superior tribological performance in lithium complex greases. Zhao et al. reported that the addition of 5 wt% $[\text{P}_{66614}][\text{BEHP}]$ significantly reduced both the coefficient of friction and wear scar diameter in four-ball tests, particularly under high load (396 N) and elevated temperature (100 °C) conditions [76]. Furthermore, Chacón-Terán et al. studied five phosphonium-based ILs with varying anionic structures in model greases. The inclusion of ILs led to friction reductions of up to 20%, depending on the base oil polarity and IL concentration. Surface analyses indicated that

these ILs formed boundary layers on steel surfaces, effectively reducing direct metal-to-metal contact [77]. Imidazolium-based ILs functionalized with benzotriazole groups also serve as promising friction modifiers.

3.2.3 Environmentally compatible surface-modified nanoparticles

The incorporation of inorganic nanoparticles—such as calcium carbonate (CaCO_3) and hydroxyapatite (HA)—has been explored as a strategy to enhance frictional behavior in greases. When combined with bio-compatible dispersants like oleic acid or used in composite form with other green materials, these nanoparticles serve as rolling elements and contribute to smoother tribological interfaces. Ali et al. studied CaCO_3 and hBN (hexagonal boron nitride) as combined solid additives in palm oil ester-based calcium complex grease. At a formulation of 5 wt% CaCO_3 and 3 wt% hBN, the grease showed a ~24% reduction in friction and ~17% wear reduction, attributed to the synergistic formation of a protective tribofilm and improved thermal stability [78]. In a related study, the use of CaCO_3 and hydroxyapatite (HA) was investigated for their potential to replace conventional metallic solid additives. When tested in combination, these biomaterials enhanced both the load-carrying capacity and friction-reducing ability of the grease while maintaining environmental compatibility [79].

Bio-based friction modifiers—including fatty acids such as oleic acid, ionic liquids, and environmentally compatible surface-modified nanoparticles—illustrate complementary mechanisms for reducing friction. Fatty acids and their esters chemisorb or physically adsorb on metal surfaces to form boundary layers. ILs contribute strongly polar and sometimes reactive species that tailor interfacial interactions, and nanoparticles can act as nano-bearings or third bodies that smooth asperity contacts. Yet, these benefits must be weighed against potential issues such as corrosion, thermal stability, and environmental fate of the additives. Rational formulation should therefore match the friction modifier to the expected contact regime: fatty acids for mild boundary lubrication, ILs for high-load or mixed lubrication where strong adsorption is critical,

and biocompatible nanoparticles for heavily loaded or contaminated environments, always ensuring that the additives remain dispersible in the chosen based oil and thickener system.

Environmentally compatible surface-modified nanoparticles, such as functionalised hBN, CaCO_3 , or hydroxyapatite, clearly demonstrate substantial benefits in terms of friction reduction and wear protection, and their mineral or biomimetic origin is often cited as evidence of greenness. Nevertheless, the long-term environmental fate of nanoscale solids released from greases is still poorly understood. Issues such as potential inhalation or dermal exposure during manufacture and handling, accumulation in aquatic or terrestrial ecosystems, and the challenges of recovering or recycling nanoparticles from used greases remain largely unresolved. Consequently, nanoparticle-based additives for green greases should be designed and evaluated within a “safe-by-design” framework, including thorough ecotoxicological testing, control of particle size and surface chemistry, and consideration of end-of-life scenarios.

3.3 Environmentally acceptable anti-wear additives

Environmentally acceptable anti-wear (AW) additives are essential for reducing surface damage in high-load grease applications while minimizing ecological impact. A promising direction involves imidazolium-based ILs functionalized with benzotriazole groups, which combine antiwear and anticorrosion properties. Cai et al. synthesized several such ILs, including [BTAMIM][BF₄], [BTAMIM][PF₆], and [BTAMIM][NTf₂], and evaluated them in polyurea greases and PEG-based lubricants. With just 1-2 wt% IL addition, wear volume was reduced by up to 12-fold compared to the base grease [80]. As shown in **Figure 6**, the addition of ILs significantly decreased the friction coefficient (a) and wear rate (b), while SEM (c-h) and AFM (A–D) analyses confirmed narrower, shallower wear tracks compared to neat PEG, directly evidencing the formation of protective boundary films. Surface analyses confirmed that ILs formed protective boundary films containing FePO₄, FeF₂, and Fe₂O₃. These tribochemical layers effectively minimized metal-to-metal contact. Moreover, the ILs showed high thermal stability

(>300 °C), no heavy metals, and excellent solubility in both synthetic and polymeric grease matrices, highlighting their potential as green replacements for ZDDP and other conventional AW additives.

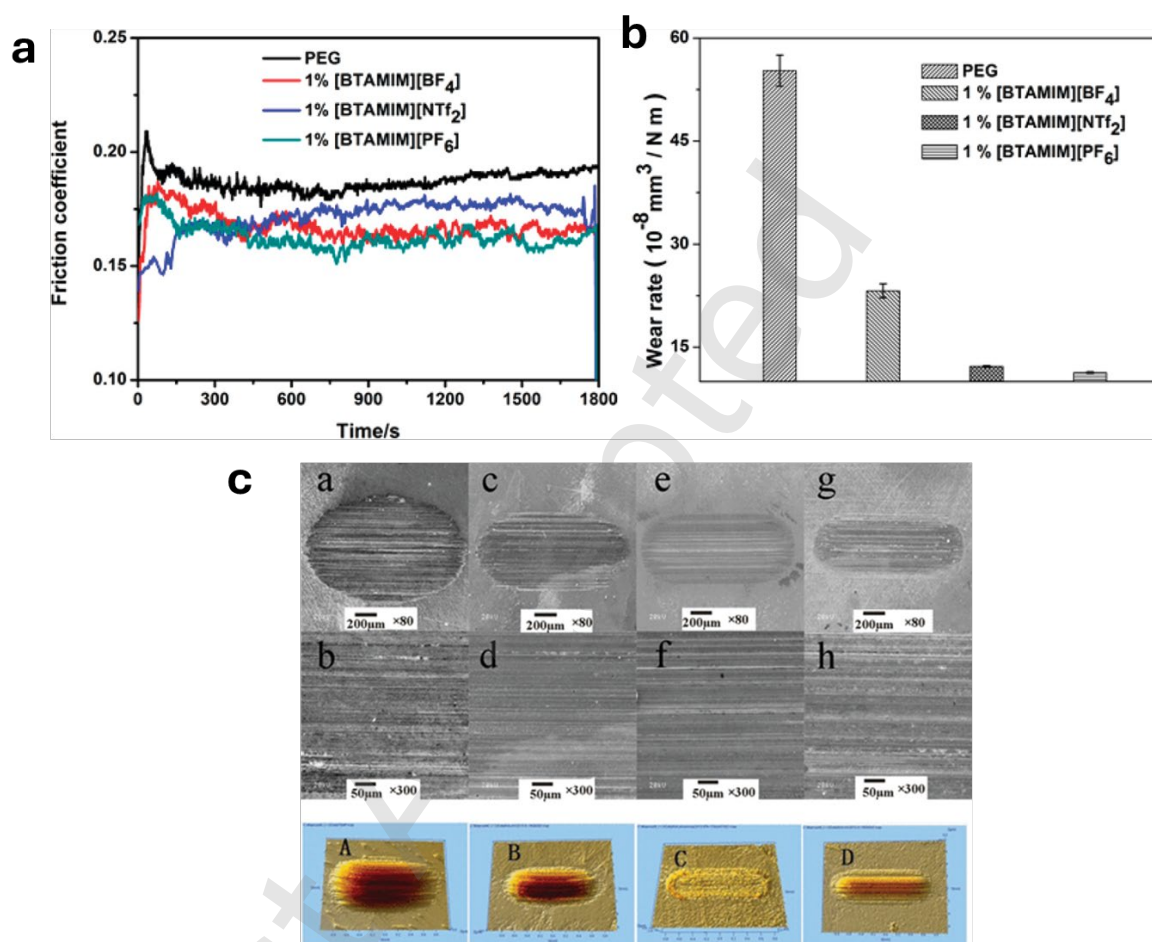


Figure 6 a) Friction coefficient evolution of PEG and PEG lubricants with 1 wt% ILs; (b) wear rate comparison; (c–h) SEM images of wear scars at low and high magnifications; (A–D) 3D AFM images of wear tracks. Adapted from Cai et al.[80]

Environmentally acceptable anti-wear additives aim to replace traditional zinc dialkyldithiophosphates and other metal-containing chemistries with phosphorus-, sulfur- or metal-free alternatives that still form protective tribofilms. Recent studies show that poly(ethylene glycol)-based systems, bio-derived phosphate esters, and functionalised nanoparticles can significantly lower wear by generating boundary layers enriched in oxygen, nitrogen, or benign

metal species. The key design challenge is to ensure robust film formation under a wide range of temperatures and loads without sacrificing biodegradability or introducing persistent elements. Future AW packages for green greases should emphasise multifunctionality—combining anti-wear, corrosion inhibition, and friction modification-while maintaining low toxicity and compatibility with both bio-based base oils and thickeners.

3.4 Summary of Green Additives in Lubricating Greases

To provide an integrated overview, **Table 2** summarizes recent advances in green additives for lubricating greases, **highlighting** a clear trend toward renewable, biodegradable, and multifunctional chemistries. Natural antioxidants such as α -tocopherol, tea polyphenols, and lignin-derived phenolics exhibit remarkable oxidative stability, extending the grease lifetime through radical scavenging and hydrogen-donation mechanisms. Bio-based friction modifiers—including oleic acid, phosphonium ionic liquids, and biogenic nanoparticles such as CaCO_3 and CaO —demonstrate efficient boundary film formation, leading to substantial reductions in friction and wear while maintaining compatibility with bio-oil matrices.

Environmentally friendly anti-wear additives, such as imidazolium-based ionic liquids bearing benzotriazole groups, show exceptional tribochemical activity and film-forming ability, enabling robust protection under high load and temperature conditions without the use of metals or halogenated compounds. Together, these additives provide a synergistic balance between tribological performance and environmental compliance.

In summary, the integration of green additives highlighted in **Table 2** underscores a paradigm shift from performance-driven to sustainability-oriented grease design. The future of bio-based lubricating greases will rely on the cooperative interactions between antioxidants, friction modifiers, and anti-wear agents to achieve long-term oxidation stability, friction control, and biodegradability within a unified, eco-friendly formulation framework.

Table 2 Summary of verified green additives in lubricating greases

Category	Representative Green Additive(s)	Main Function / Mechanism	Key Performance Improvement	Environmental & Sustainability Attributes	Reference
Natural Antioxidants	α -Tocopherol, Tea Polyphenols, Gallic Acid	Radical scavenging; inhibit oxidative degradation in base oils	Increased oxidation induction time (OIT) by 93–454% in soybean-oil-based greases	Renewable, biodegradable, non-toxic plant-derived phenolics	[72]
	Lignin-based antioxidant (from castor oil oleogel)	Radical-trapping via phenolic hydroxyl groups in lignin	OIT improved from 20 s $\rightarrow$ 1959 s ($\approx 98\times$ increase); improved tribological stability	> 80% biomass-derived; fully renewable, low ecotoxicity	[73]
	Acylated Chitosan + Propyl Gallate	Enhanced oil compatibility and antioxidation in castor-oil greases	Oxidation onset temperature (OOT) increased 142 °C $\rightarrow$ 236 °C	Renewable polysaccharide-based; biodegradable; low toxicity	[74]
Bio-based Friction Modifiers	Oleic Acid (OA)	Amphiphilic adsorption film formation on metal surfaces	Friction coefficient reduced by $\sim$ 30% under 20 N load	Biodegradable fatty acid; bio-derived; low cost	[75]
	Phosphonium-based ILs ([P ⁶⁶⁶¹⁴][BEHP])	Surface adsorption; film-forming friction reduction	Friction and wear reduced by 20–40% , particularly at high load/temperature	Non-volatile, tunable molecular design, partially biodegradable	[81]
	CaCO ₃ nanoparticles (palm ester bio-grease)	Nano-bearing and tribofilm formation	Friction $\downarrow$ 24%, wear $\downarrow$ 17%; improved load capacity	Mineral/biogenic origin, non-toxic, recyclable	[78]
	Waste eggshell-derived CaO nanoparticles (in mustard oil)	Tribofilm formation; enhanced oxidation resistance	Friction $\downarrow$ 32%, wear $\downarrow$ 28%; improved thermal stability	Upcycled biowaste, renewable CaCO ₃ source, eco-friendly	[82]
Environmentally acceptable Anti-wear Additives	Imidazolium-based ILs with Benzotriazole ([BTAMIM][BF ₄], [BTAMIM][PF ₆], [BTAMIM][NTf ₂])	Formation of tribochemical films (FePO ₄ , Fe ₂ O ₃) preventing metal contact	Wear volume reduced up to 12$\times$, friction reduced significantly	Metal-free, thermally stable (> 300 °C), non-volatile	[80]

4 Classification of green greases based on gel technology

Gels have emerged as a promising platform for green lubricating greases due to their three-dimensional (3D) network structures, which offer enhanced structural integrity, flow control, and shear adaptability [83]. These networks can effectively immobilize base fluids, enabling consistent lubrication under dynamic or extreme conditions. Unlike traditional metallic soap-based greases, which depend only on point attractions between thickeners and base oils and therefore suffer from poor structural stability, gel-based lubricants form continuous and stable 3D matrices that effectively immobilize base oils. This structural advantage not only enhances integrity and stability but also improves flow control and adaptability under shear stress, ensuring reliable lubrication in dynamic or extreme environments (**Figure 7**).

To fundamentally understand this stability, the viscoelastic nature of the gel network must be considered. Unlike traditional lithium-soap greases, which rely on the mechanical entanglement of fibrous metallic soaps, green gel lubricants often form supramolecular networks driven by non-covalent interactions such as hydrogen bonding and van der Waals forces. This difference dictates their structural dynamics under shear.

Under high shear rates characteristic of elastohydrodynamic lubrication (EHL) contacts, the physical networks of bio-based thickeners exhibit superior thixotropic reversibility compared to the often brittle breakage of soap fibers. Specifically, the non-covalent bonds can temporarily dissociate under high stress (where the loss modulus G'' exceeds the storage modulus G'), allowing the grease to flow and lubricate, and then rapidly re-associate upon exiting the contact zone.

This reversible self-assembly mechanism is critical for maintaining a dynamic equilibrium between flow and retention, offering a distinct advantage over the static "bleed-only" model of conventional greases.

At the same time, long-term grease performance is governed not only by the initial network structure, but also by its structural evolution under tribological stress. Repeated shear, local flash

temperature, pressure-induced rearrangement, and tribo-oxidative effects may progressively alter the gel network, leading to fiber fragmentation, irreversible structural collapse, oil bleed, hardening, or loss of replenishment capability. Therefore, a practical green grease should be evaluated not only by its initial rheological and tribological properties, but also by its ability to maintain network resilience and functional recovery during prolonged operation.

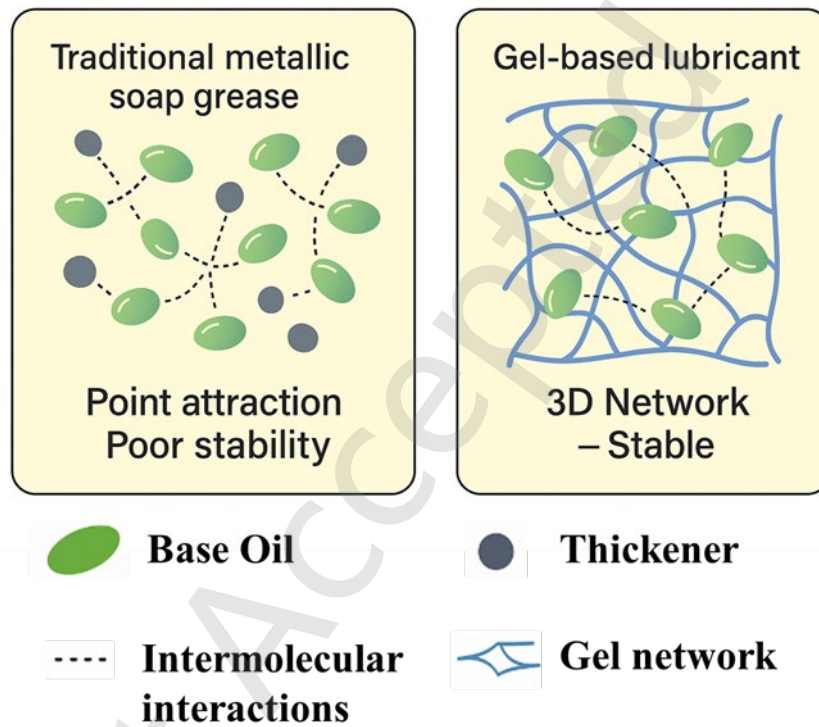


Figure 7 Comparison of the microstructures of traditional metallic soap grease and gel-based lubricants.

4.1 Green oleogel

Green oleogels have emerged as a promising class of sustainable lubricating greases, offering a bio-based, biodegradable alternative to conventional petroleum-derived products. These systems are typically formulated by structuring vegetable oils—most commonly castor oil—using various naturally derived or non-toxic gelators to create a semi-solid matrix with desirable rheological and tribological properties.

Several recent studies have demonstrated the effectiveness of oleogels prepared from cellulose derivatives, lignin-based gelators, clays, and low molecular weight organic compounds.

Cellulose derivatives such as methylcellulose, ethyl cellulose, and α -cellulose have also been successfully employed as structuring agents. In castor oil systems, cellulose-thickened greases exhibited viscoelastic moduli and flow curves comparable to traditional lithium soap greases. However, formulations with ethyl cellulose alone showed higher oil separation at elevated temperatures, suggesting the need for blending or composite approaches to improve stability [84, 85]. Mineral-based thickeners like sepiolite have also shown promise in forming oleogels. Martín-Alfonso et al. prepared gel-like dispersions of sepiolite in castor oil, reporting that a 30 wt% sepiolite formulation achieved stable shear-thinning behavior, rapid structural recovery, and wear protection on par with commercial greases [86]. Low molecular weight gelators derived from renewable resources have demonstrated strong self-assembly capabilities and tribological performance. Cai et al. introduced a glucose-based gelator (N-octadecyl-D-gluconamide) that formed thermoreversible, thixotropic oleogels. The formulation provided significantly lower friction coefficients and wear rates under boundary lubrication conditions compared to base oil alone [87].

Recent advances have explored renewable and biodegradable thickening systems that can directly utilize plant oils. For example, Tian et al. developed layered double hydroxide–stearic acid (LDH-SA) greases using soybean oil and other vegetable oils as the base oils (**Figure 8**) [72]. These LDH-SA/plant oil greases displayed good rheological behavior, while maintaining low friction coefficients and reduced wear volumes compared with lithium greases. Importantly, the oxidative stability of the greases was significantly enhanced by the incorporation of natural antioxidants such as tocopherols and gallic acid, demonstrating that plant oils, when paired with renewable and biodegradable thickeners, can achieve both high tribological performance and improved durability.

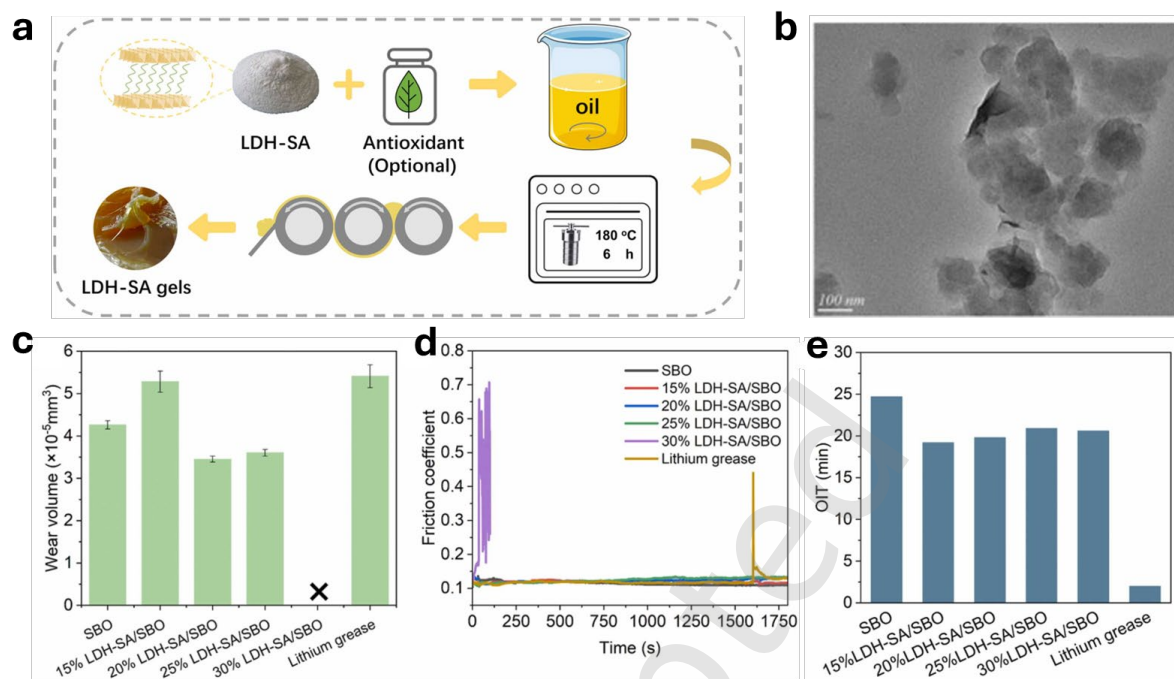


Figure 8 Preparation and performance of LDH-SA/soybean oil gels: (a) schematic of gel preparation, (b) TEM image of LDH-SA, (c) wear volume, (d) friction coefficient, and (e) oxidation induction time (OIT), compared with soybean oil and lithium grease. Reproduced from ref. [72] with permission.

Gelators are a class of thickeners that form three-dimensional networks to trap base oils, giving lubricating greases their semi-solid structure. As alternatives to traditional lithium or polyurea thickeners, organogelators such as glyceryl and sorbitan monostearates have shown potential as green thickeners due to their biodegradability and renewable origin [68]. Recently, Wu et al. developed a lignin-based organogelator synthesized from lignin, malic acid, and epoxidized soybean oil using a green, isocyanate-free route [48](**Figure 9a**). This gelator was able to structure vegetable oils, particularly castor oil, into stable greases with strong shear-thinning behavior, improved friction and wear performance, and outstanding surface adsorption. A 35 wt% gelator in castor oil reduced friction by ~20% and wear by ~40% compared to the base oil (**Figure 9 b,c**).

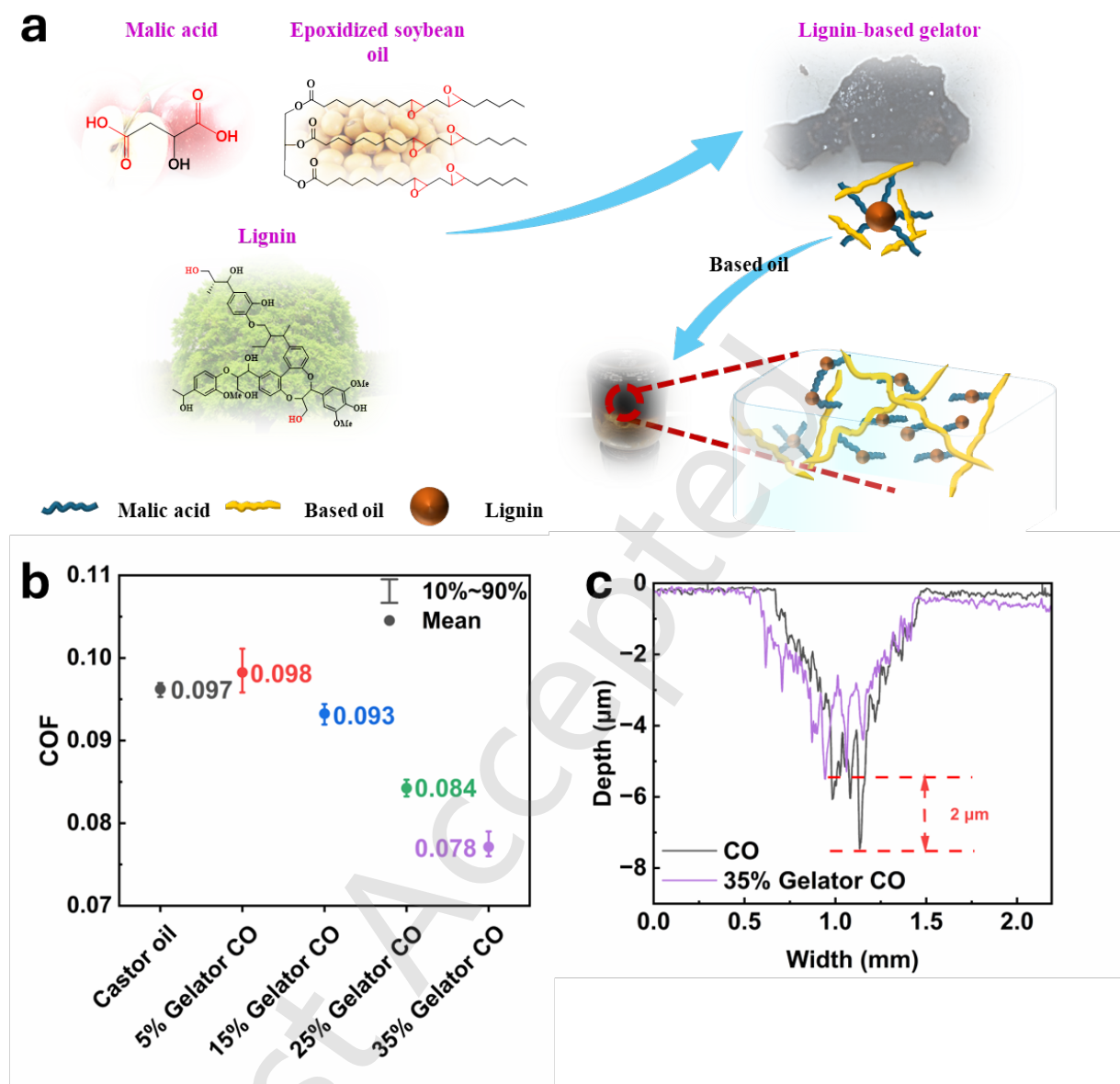


Figure 9 (a) Green, isocyanate-free synthesis route of a lignin-based organogelator from lignin, malic acid and epoxidized soybean oil, and schematic illustration of its self-assembly into a three-dimensional network that structures vegetable oils (exemplified by castor oil) into grease-like oleogels. (b) Coefficient of friction (COF) of neat castor oil and castor-oil oleogels with different gelator contents, showing a gradual decrease in COF with increasing gelator loading. (c) Wear-track profiles after sliding for castor oil and the 35 wt% gelator–castor-oil oleogel, highlighting the pronounced reduction in wear depth provided by the green gelator. Reproduced from **ref. [48]** with permission.

Green oleogels demonstrate that bio-derived gelators can transform low-viscosity oils into semi-solid greases with controllable yield stress and shear-thinning behaviour. Their performance strongly depends on the self-assembled microstructure of fibres, platelets or three-dimensional networks, which in turn is governed by gelator concentration, polarity and processing conditions. Tribological studies consistently show that networks which are strong enough to retain oil sufficiently labile to break and rebuild under shear deliver the best compromise between low friction, low wear and pumpability. Consequently, the design of oleogel-based green greases should focus on tuning gelator chemistry and network morphology to achieve fast structural recovery, moderate stiffness and good thermal stability, often supported by small amounts of co-thickeners or fillers.

4.2 Green hydrogel grease

Hydrogels have attracted increasing interest as water-based alternatives to conventional oil-based greases, aligning well with the goals of sustainable lubrication. These materials typically comprise hydrophilic polymer networks swollen with water, offering advantages such as biocompatibility, non-flammability, and environmental safety. Despite their potential, most green hydrogels have been explored for applications in biomedical coatings and drug delivery systems, with only a limited number directly applied as lubricating greases [88, 89].

Recent studies have demonstrated the viability of hydrogel systems for tribological use. For instance, Shetty et al. (2020) developed fully green hydrogel-based greases using sodium carboxymethyl cellulose (NaCMC) and hydroxyethyl cellulose (HEC) as gelators in a PEG/water base fluid. These greases showed stable friction coefficients (~ 0.07 – 0.10) and significant wear reduction under severe tribological conditions (2.5 GPa load, 50 Hz frequency), outperforming neat PEG/water solutions [89]. The superior performance of NaCMC-based greases was attributed to their anionic nature, promoting strong adsorption on metal surfaces and facilitating robust tribo-film formation.

Other strategies include hybrid gel systems. For example, hydrogels reinforced with functionalized microgels or fatty acid derivatives have shown improved mechanical integrity and reduced wear under boundary lubrication conditions. Blum and Ovaert (2013) functionalized polyvinyl alcohol (PVA) hydrogels with lauroyl and stearyl chlorides, demonstrating significant reductions in polymer loss and surface degradation during extended sliding tests against both metal and cartilage surfaces [90].

Hu et al. (2020) developed a superlubricious hydrogel system using proteinaceous lactoferrin microgels and κ -carrageenan. This reinforced network exhibited a friction coefficient as low as 0.01, even outperforming human saliva under oral tribological conditions [91]. Although this system was not originally designed for mechanical lubrication, it demonstrates the potential of combining soft colloids and hydrogels to achieve ultra-low friction in aqueous environments.

Hydrogel-based greases highlight the potential of water and other benign polar fluids as base media for lubrication, especially in biotribology and environmentally sensitive applications. Double-network hydrogels, fibre-reinforced systems, and hybrid hydrogels incorporating lubricious polymers or nanoparticles can reach ultra-low friction and acceptable wear under aqueous conditions. Nonetheless, limitations in mechanical strength, dehydration, and microbial stability still restrict their use in heavy-duty engineering contacts. Future hydrogel greases should therefore exploit toughened network architectures, humidity-resistant formulations, and antimicrobial strategies, while carefully balancing water content, elasticity, and interfacial chemistry to maintain both lubricity and long-term integrity.

4.3 Ionogels

Ionic gels (ionogels), composed ILs physically or chemically immobilized within a gel network, represent a promising class of green lubricating materials. These systems combine the unique physicochemical properties of ILs-such as high thermal stability, wide liquid range, and low

volatility-with the structural integrity and leak-proof characteristics of gels, making them ideal candidates for high-performance semisolid greases.

One representative approach involves the formation of supramolecular ionogels through the self-assembly of imidazolium-based ILs bearing urea functional groups as gelators. These ionogels exhibit thixotropic behavior, thermoreversibility, and significantly enhanced antiwear and anticorrosion properties compared to pure ILs. For instance, Yu et al. demonstrated that a 5 wt% urea-functionalized imidazolium gelator in [HMIm][PF₆] drastically reduced wear and friction... Although this specific study utilized a fluorinated anion ([PF₆]⁻) to establish the gelling mechanism, future green ionogels should apply this supramolecular strategy to the halogen-free ILs (e.g., phosphates) discussed in **Section 2.1.2** to ensure full environmental compliance[61]. Another significant advance involves the design of dual hydrogen bond networks within ionic gels. Zhou et al. developed ionic gels by combining polyacrylamide (PAM) with choline-based deep eutectic solvents (DESs) such as urea/choline chloride (UCC) and glycerol/choline chloride (GCC). These ionogels exhibited tunable viscosity, excellent self-healing ability, and strong boundary lubrication. Under a 1.35 GPa load, the UCC-PAM ionic gel reduced wear volume by up to 64% and maintained a low friction coefficient over extended operation. The improvement in tribological properties is attributed to the dual hydrogen bond interactions between PAM and DESs, which help stabilize the lubricating film during sliding [92]. Additionally, ionogels have been reinforced with carbon-based nanomaterials to further enhance their lubricity and mechanical strength. Chen et al. reported a self-healing supramolecular ionogel incorporating carbon nanotubes and ILs, which retained conductivity and displayed stable friction coefficients under various shear conditions, extending the gel's usability in flexible electronics and high-temperature lubrication [93]. Furthermore, Wang et al. introduced ionic supramolecular oleogels enhanced with liquid metal nanodroplets, offering superior adaptability to complex frictional environments and showing excellent performance in tribological tests even under thermal and mechanical stress [94].

Ionogels offer a powerful route to immobilise ionic liquids within polymeric or inorganic frameworks, combining the high thermal stability and polarity of ILs with the dimensional stability of gels. Properly designed ionogels can sustain high temperatures and loads, providing low friction and wear where conventional greases fail. However, the environmental profile of many ILs, the cost of ionogel fabrication, and uncertainties about recyclability currently limit large-scale adoption. Design of green ionogel greases should therefore prioritise ILs with proven low toxicity and good biodegradability, minimise IL content through efficient network confinement, and consider hybrid concepts where small ionogel domains are dispersed within more conventional bio-based grease matrices.

5 Low-hazard and energy-saving synthesis strategies for grease components

5.1 Low-hazard synthesis of bio-based gelators for oleogels

Recent developments in green lubrication research have placed increasing emphasis on the green synthesis of bio-based gelators, which function as both thickeners and performance enhancers. Unlike traditional thickeners synthesized via energy-intensive or toxic chemical routes, green gelators are often derived from renewable materials such as lignin, fatty acids, or sugars, and are produced via environmentally benign processes.

For example, Wu et al. reported an isocyanate-free and low-hazard route for synthesizing a lignin-based gelator using malic acid and epoxidized soybean oil[48]. This method eliminates hazardous reagents and operates under mild thermal conditions, resulting in a high-yield, biodegradable product with dual functionality as both a gelator and antioxidant. Similarly, sugar-based gelators (e.g., N-octadecyl-D-gluconamide) have been synthesized via solvent-free or ethanol-assisted self-assembly, aligning well with the principles of green chemistry [87, 95].

These approaches demonstrate that the synthesis pathway, not just the gelator's composition, is critical to evaluating a lubricant's environmental profile. Moving forward, integrating green

reaction conditions, such as ambient pressure, aqueous systems, and recyclable catalysts, will be essential for the scalable production of truly sustainable gelator-based lubricating greases.

5.2 Mild and eco-friendly processing conditions

Green grease production increasingly favors mild reaction conditions, such as low temperature, low pressure, and solvent-free or green solvent systems, to reduce environmental impact. Compared with conventional greases synthesized under high temperatures or with hazardous reagents, these approaches offer significant advantages in energy efficiency, safety, and sustainability.

For example, many bio-based gelators-such as those derived from lignin, amino acids, or sugars-can be blended with vegetable oils at temperatures below 80 °C to form stable oleogels without the need for catalysts or pressurized systems. Such conditions simplify processing and help preserve the functionality of sensitive natural ingredients. In some systems, self-assembly of low molecular weight gelators enables gel formation under ambient or slightly elevated temperatures, relying on hydrogen bonding or hydrophobic interactions rather than chemical reactions. These processes are inherently cleaner, generating minimal waste and avoiding toxic byproducts.

Overall, the use of mild, energy-saving synthesis routes enhances both the environmental performance and scalability of green greases. Future research may focus on further simplifying these methods through solvent-free blending, microwave-assisted processes, or bio-catalysis, making green grease production even more sustainable and practical for industrial applications.

5.3 Recent advances in green grease technology

Recent advances in green grease technology increasingly emphasise hybrid and multifunctional formulations. A recurring trend is the combination of bio-based base oils and thickeners with small fractions of nanoparticles, ionic liquids or reactive polymers to extend load-carrying capacity, high-temperature stability and oxidation resistance. These hybrids often outperform purely bio-based or purely conventional greases, demonstrating that carefully selected non-bio components can be used strategically without compromising overall environmental benefits.

Another important development is the growing focus on structure–property relationships across different gel technologies. Studies that correlate network morphology, rheological parameters (such as yield stress and storage modulus), and tribological indicators (friction coefficient, wear scar diameter, and film thickness) are beginning to reveal general design rules that apply to oleogels, hydrogels and ionogels alike. Greases that exhibit strong yet recoverable networks, pronounced shear-thinning, and stable microstructures under operating temperature and shear conditions tend to deliver robust lubrication performance.

Finally, the field is progressing from proof-of-concept demonstrations towards application-oriented optimisation. Long-duration tests, comparison with commercial reference greases and preliminary field trials in bearings, gears, and biotribological contacts indicate that several green grease concepts are approaching technological readiness. Future work should integrate green synthesis, lifecycle assessment, and techno-economic analysis into formulation studies, ensuring that emerging green greases are not only high-performing in the laboratory but also scalable, cost-competitive, and aligned with broader sustainability targets.

To consolidate the recent progress in sustainable lubricating grease development, the low-hazard and energy-saving synthesis strategies discussed in Sections 5.1-5.3 are summarized in **Table 3**. This table highlights representative approaches for the synthesis of grease components using renewable precursors, environmentally benign processing routes, and emerging environmentally preferable technologies such as halide-free ionic liquids and ionic supramolecular oleogels.

Table 3 Low-hazard and energy-saving synthesis strategies for grease components: renewable precursors, mild synthesis methods, and emerging environmentally preferable technologies

Category	Representative Strategy / Example	Key Reaction or Process Description	Environmental & Technical Advantages	Reference
Renewable and bio-based precursors	Vegetable oils (soybean, castor, palm, mustard) as	Transesterification and epoxidation of triglycerides via	Renewable, low toxicity, improved oxidative stability	[78], [51]

	base stocks for grease	enzymatic or heterogeneous catalysts		
	Lignin-, cellulose-, and chitosan-derived gelators	Esterification, acylation, or ionic crosslinking of biopolymers	Biodegradable, tunable rheology, no metal soap waste	[48], [49], [73], [74]
	Waste biomass-derived nanomaterials (e.g., CaO from eggshell, SiO ₂ from rice husk)	Thermal calcination or sol-gel conversion of agro-waste	Waste valorization, circular economy, low-cost synthesis	[82]
Low-hazard and mild synthesis methods	Isocyanate-free lignin-malic acid gelator formation	Solvent-free polycondensation between lignin and natural acids with epoxidized oils	Non-toxic, dual gelation and antioxidation	[48]
	Enzymatic catalysis for ester/amide bond formation	Lipase-catalyzed transesterification under solvent-free conditions	Mild temperature, recyclable biocatalyst, high selectivity	[51]
	Microwave-assisted polymerization and crosslinking	Rapid heating without solvents; uniform reaction environment	Energy-efficient, minimal by-products	[89, 90]
Emerging environmentally preferable grease technologies	Halide-free ionic liquids and bio-phosphate ILs	Halogen-free metathesis; metal-free ionic systems	Stable, biodegradable, tunable polarity	[81]
	Ionic supramolecular oleogels reinforced with liquid metal nanodroplets	Formation of supramolecular ionic networks incorporating Ga-based nanodroplets for improved load-bearing capacity	Excellent antiwear and heat dissipation performance; novel ionic supramolecular design	[94]

Overall, green synthesis strategies for lubricating greases demonstrate a clear transition from traditional metal-based thickener systems to biopolymer- and ionic-liquid-based sustainable alternatives. The integration of renewable feedstocks, mild catalytic processes, and advanced supramolecular or nanocomposite systems offers a viable route toward environmentally responsible and high-performance grease formulations. Accordingly, whether a grease component can be regarded as sustainable depends not only on its renewable origin, but also on the overall

footprint associated with its synthesis pathway, processing conditions, service life, and end-of-life fate.

6 Comparison of green greases with traditional Grease

6.1 Environmental performance

Green greases differ from traditional petroleum-based greases primarily in their environmental profile and synthesis strategy, while gradually approaching comparable tribological performance. As detailed earlier in this review, green greases typically use renewable base oils, bio-based thickeners, and biodegradable, low-toxicity, or otherwise environmentally acceptable additives, in contrast to the fossil-derived and chemically intensive components of conventional greases[36]. Performance-wise, green greases have made noticeable improvements. Though traditional greases still lead in extreme thermal and oxidative stability, modern green formulation, especially those incorporating hybrid strategies, can achieve similar levels of friction and wear resistance in many applications [27].

Environmentally, the advantages of green greases are more pronounced. Their synthesis under mild, solvent-free conditions, combined with high biodegradability and low toxicity, offers a clear sustainability edge [96]. This contrasts with the energy-intensive processes and lower environmental compatibility of conventional greases. At the same time, it should be recognized that bio-based composition alone does not necessarily imply superior overall sustainability. The net environmental benefit of a green grease also depends on factors such as synthesis complexity, energy consumption, solvent use, additive selection, service life, and end-of-life treatment. Therefore, a balanced comparison between green and conventional greases should ideally be supported by life cycle assessment rather than by renewable content alone.

While cost and industrial standardization remain challenges for green greases, their long-term potential is bolstered by lifecycle benefits and increasing demand for sustainable lubricants.

The contrast between the two types of greases is conceptually illustrated in **Figure 10**. The radar chart is intended to provide a qualitative visualization of general trends, highlighting the superior environmental attributes of green greases (such as biodegradability, low toxicity, and reduced carbon footprint), while indicating that traditional greases still retain advantages in thermal and oxidative performance.

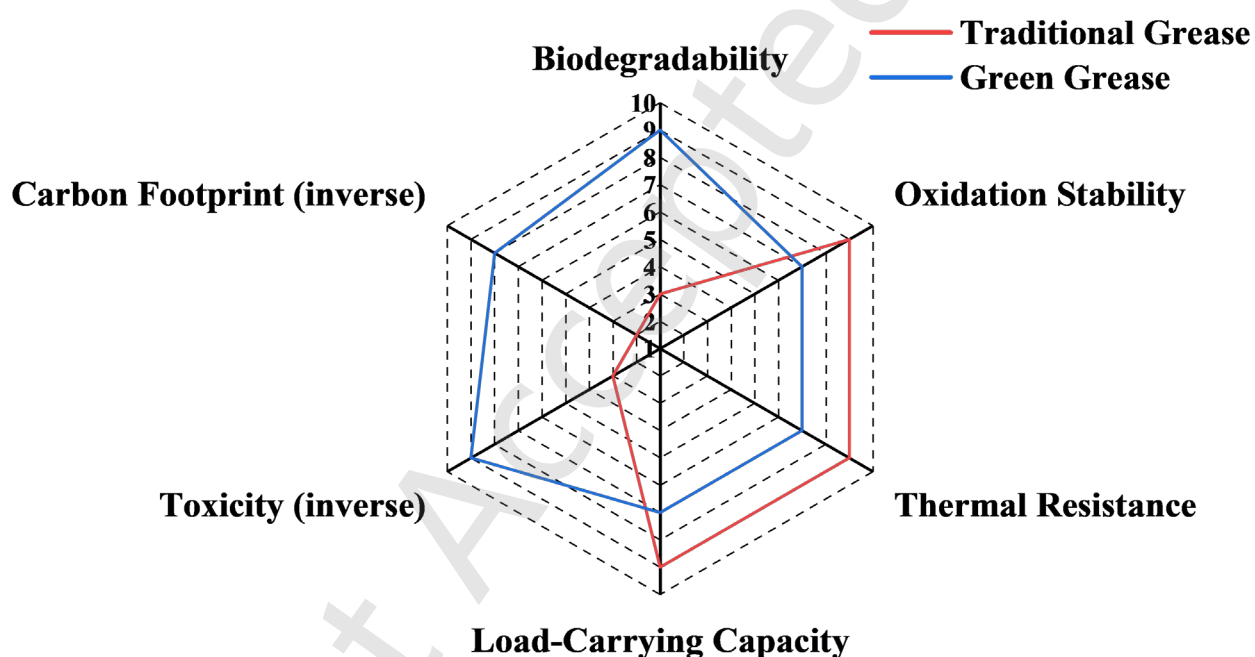


Figure 10 Conceptual radar chart qualitatively comparing general performance and sustainability trends of green and traditional greases. The plot is intended as a schematic illustration rather than a quantitative meta-analysis based on harmonized datasets.

In summary, current evidence shows that green greases can drastically reduce ecotoxicity, persistence and reliance on critical raw materials compared with traditional lithium- and calcium-soap greases, while providing comparable or better biodegradability and lower greenhouse-gas

emissions over the life cycle. The main trade-offs lie in higher material costs and, in some cases, shorter service life under extreme operating conditions. Design of environmentally optimised greases should therefore combine renewable base fluids and eco-friendly additives with long drain intervals, efficient sealing and recyclability, so that the overall system-level impact is minimised rather than focusing solely on individual components.

6.2 Tribological and physicochemical performance

Recent developments in green lubricating greases reveal that bio-based thickeners and new green gels-including lignin-derived gelators, cellulose derivatives, polysaccharides, and lignosulfonate hydrogels-can deliver friction and wear performances comparable to, or even surpassing, those of conventional lithium- and calcium-based greases. As summarized in **Table 4**, friction coefficients of green greases generally fall within 0.03-0.10, while wear-scar diameters typically lie between 0.17 and 0.80 mm, covering the full performance range of commercial mineral-oil-based greases [73, 85, 89, 97, 98]. Importantly, these systems achieve this level of performance while using biodegradable base oils (castor oil, ESO, palm ester, jojoba oil) or water/PEG fluids, and non-toxic biopolymeric thickeners, highlighting the strong potential of green alternatives to replace petroleum-based greases.

It should be noted, however, that the tribological data summarized from the literature were obtained using different test configurations, such as SRV, ball-on-disc, four-ball, HFRR, Brugger-type, and custom rigs, under different loads, temperatures, frequencies, counterpart materials, and lubrication regimes. Therefore, the reported friction and wear values should be interpreted primarily as indicative comparisons of general trends and formulation potential, rather than as directly equivalent quantitative metrics for strict one-to-one comparison across studies.

From a physicochemical standpoint, most green greases combine high biodegradability and low acute toxicity with acceptable drop points, mechanical stability, and oxidation resistance for low-to medium-severity applications [72]. Hybrid formulations further narrow the gap to high-

performance soap greases by incorporating nanoparticles or ionic liquids as multifunctional co-additives, thereby improving load-carrying capacity, oxidation-induction times, and resistance to oil separation, albeit sometimes at the expense of cost and formulation complexity.

In summary, existing data demonstrate that green greases can already meet tribological requirements in many bearing and sliding contacts, especially where moderate temperatures and speeds are involved, and environmental compliance is prioritized. Looking forward, systematic benchmarking under harmonized test protocols, long-duration field trials and deeper structure–property correlations will be crucial to identify the remaining performance gaps under high temperature, high speed, and extended relubrication intervals, and to guide the rational design of next-generation green greases that simultaneously optimize friction, wear, environmental impact, and total cost of ownership.

Table 4 Representative tribological and physicochemical performance of green greases under different test configurations

Grease Type	Base Oil	Thickener	Additives	Representative test configuration / conditions	COF, μ	Wear	Biodegradability	Toxicity	Reference
Green oleogel (lignin–castor oil)	Castor oil	Lignin-based gelator (malic-acid-modified organosolv lignin)	None	SRV ball-on-disc; 100 N; 50 Hz; RT; 2 h	0.103 ± 0.007	Wear volume ~64% lower than pure castor oil	High (vegetable oils + biopolymers)	Low	[73]
Lignosulfonate hydrogel grease	Water	Lignosulfonate-based hydrogel (APS initiated, Fe^{3+} crosslinking)	None	SRV ball-on-disc; 200 N; 25 °C; 25 Hz; 0.5 h	COF reduced 37.5% vs 0 wt% LS hydrogel	Wear volume reduced 31.25% vs 0 wt% LS	High	Low	[97]
PEG/water–cellulose gel grease	PEG 200 / Water (1:1)	HEC or NaCMC	None	SRV-III; ASTM D6425; 150 N; 25 °C; 50 Hz; 1 mm; 1 h	Lower than neat PEG/water, NaCMC < HEC	Wear lower than PEG/water; NaCMC < HEC	High	Low	[89]
P34HB–ethyl cellulose fully renewable grease	Castor oil	P34HB + ethyl cellulose composite	None	SRV reciprocating test; 10–25 N; 1–5 Hz; 60–80 °C	Lower COF than P34HB–Li and P34HB–Ca greases	Wear volume reduced by 39.20% vs reference P34HB grease	High	Low	[85]
Calcium-complex palm-biogrease with green additives	Palm-based ester	Calcium complex soap	CaCO_3 + HA nanoparticles	Four-ball test	Avg COF improved by about 6.5% vs	Wear scar slightly smaller than base grease; see	High	Low	[99]

Grease Type	Base Oil	Thickener	Additives	Representative test configuration / conditions	COF, μ	Wear	Biodegradability	Toxicity	Reference
Jojoba oil biogrease + plant-derived nanoadditives	Jojoba oil	Lithium soap	Activated carbon nanoparticles (0–1.5 wt%)	Brugger / rolling-bearing test rig	base palm bio-grease COF decreased by 38% vs plain jojoba grease	original data for exact values Wear scar reduced by 24% vs plain jojoba grease	High	Low–Medium	[43]
Corn-stover cellulose/ESO bio-grease	Epoxidized soybean oil (ESO)	MDI-modified cellulose from corn stover (St-C-MDI)	None	Ball-on-disc / four-ball	0.027 (lower than commercial Li grease, 0.084)	0.61 mm vs 0.64 mm for Li grease	High (agricultural waste cellulose + ESO)	Low	[98]
Fried-vegetable-oil Ca-soap grease with biopolymers	Waste sunflower / palm frying oils	Calcium (and lithium) stearate soaps (20 wt%)	Cellulose or lignin sulfate fillers (15 wt%)	HFRR; 60 °C	Avg COF = 0.030 – 0.062	0.169–0.250 mm	High (waste vegetable oils + biopolymers)	Low	[100]

Note: Because the data were obtained using different test configurations, loads, temperatures, frequencies, counterpart materials, and lubrication regimes, the reported tribological results are intended for qualitative or trend-level comparison only and should not be interpreted as directly equivalent quantitative metrics across studies. Where necessary, wear-scar values originally reported in μm were converted to mm for consistency.

7 Applications and market potential for green grease

Green greases are increasingly adopted in application domains that prioritize sustainability, safety, and environmental compliance. They have found promising use in sectors such as agriculture and forestry—where lubricant leakage poses risks to soil and water systems—as well as marine and offshore operations that require high biodegradability due to aquatic environmental constraints[101]. In the food and pharmaceutical industries, green greases offer non-toxic, safe alternatives for incidental contact scenarios[102]. Furthermore, their low volatility and clean-operating properties make them suitable for emerging technologies such as electric vehicles (EVs) and high-precision electronic components.

Beyond these sectors, green greases are gaining interest in construction, wind energy, and rail transport, particularly in regions with stringent environmental policies such as the EU and North America.

From a market perspective, global interest in environmentally acceptable lubricants (EALs), including green greases, is accelerating. It should be noted, however, that the market data cited here are drawn from different reports with different baseline years, category boundaries, and forecast horizons, and therefore should be interpreted as directional indicators rather than as directly harmonized market estimates. For example, one report values the global grease market at USD 6.05 billion in 2024 and projects it to reach USD 7.68 billion by 2030, corresponding to a CAGR of 4.1%[103]. In another report, the bio-based lubricants market is projected to grow from USD 2.5 billion in 2024 to USD 3.38 billion by 2032[104]. A separate market report projects the broader EAL market to expand from USD 97.7 billion in 2023 to USD 148.4 billion by 2031, with a CAGR of 12.5%[105]. Despite these differences in reporting windows and scope, all three sources indicate growing industrial and commercial interest in more sustainable lubricant technologies. The adoption of green lubricants is

accelerating, driven by stricter environmental regulations (e.g. REACH, VGP) and industry decarbonization goals, as well as a growing societal demand for eco-friendlier products. The cost pressures and the lack of greater standardization are enduring issues, but the ever-improving grease technologies and favorable policy frameworks are increasingly improving commercial viability.

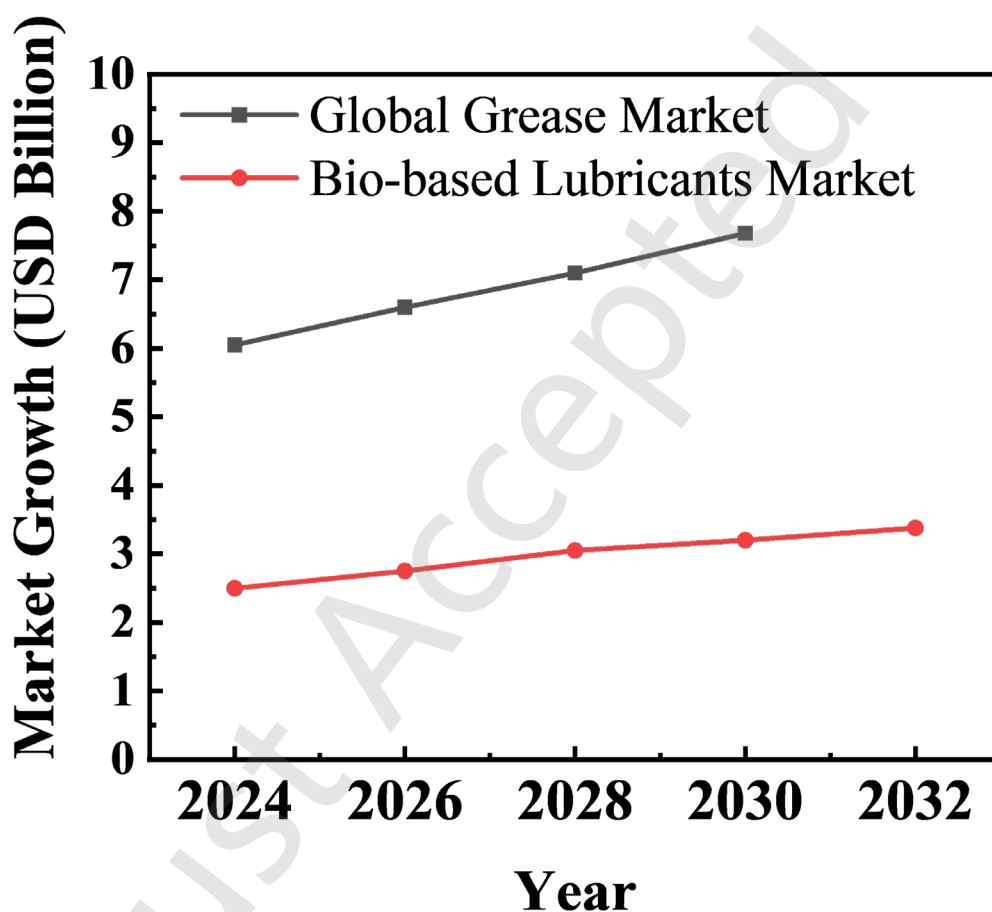


Figure 11 Projected market growth trends for selected lubricant-related market segments based on different industry reports. Because the cited reports use different baseline years, forecast horizons, and market scopes, the figure is intended to illustrate general growth tendencies rather than provide a directly harmonized quantitative comparison. Data sources: Grand View Research [103], Data Bridge Market Research [104].

8 Challenges of green grease

Despite the environmental imperative, the transition from mineral-based to green greases is hindered by distinct technical, economic, and operational challenges. A critical analysis of these barriers reveals that they are not merely "performance gaps" but fundamental physicochemical trade-offs that require targeted molecular engineering. A central issue behind several of these challenges is the structural degradation of the grease network under tribological stress. Even when a grease exhibits favorable initial rheological properties, its lubrication performance may deteriorate during service if the thickener network cannot withstand repeated shear, thermal exposure, oxidative attack, and mechanical disruption.

8.1 Oxidative stability and hardening-type failure

Oxidative stability remains a primary technical challenge, particularly for greases based on vegetable oils. The high degree of unsaturation in triglyceride structures makes them prone to oxidative polymerisation under thermal and catalytic conditions. Unlike mineral oils, which typically degrade via chain scission and viscosity loss, bio-based oils tend to harden over time due to cross-linking reactions. This atypical failure mode poses a serious reliability risk in fill-for-life or low-maintenance applications, as hardened grease may fail to reflow after idle periods. While antioxidants can extend oxidation induction times, long-term stability often requires structural modification of the base oil, such as partial hydrogenation or conversion to hindered esters.

8.2 Compatibility with legacy lubricants and non-drop-in behaviour

Compatibility with existing lubrication systems is another major obstacle. Many green greases employ polar base fluids that are thermodynamically incompatible with residual mineral oils or conventional soap-thickened greases. Even limited cross-contamination can lead to softening, excessive oil bleed, or structural collapse. As a result, green greases are rarely true drop-in replacements and typically

require thorough system cleaning, increasing downtime and transition costs. Improving tolerance to mixed-lubricant environments through hybrid base oils or co-thickener strategies remains an important research need [27].

8.3 Economic scalability and formulation complexity

Economic scalability further constrains adoption. Although renewable feedstocks are widely available, the synthesis of advanced green thickeners—such as ionic liquids or functionalised biopolymers—often involves multi-step, energy-intensive processes with low atom efficiency[106]. Without process simplification and economies of scale, high-performance green greases struggle to compete with commodity lithium-soap greases in cost-sensitive applications, restricting their use mainly to environmentally sensitive or total-loss scenarios [107].

8.4 Lack of standardised performance and sustainability metrics

Finally, the lack of standardised testing and certification frameworks hampers industrial confidence. Current biodegradability and ecotoxicity assessments are largely based on aqueous laboratory tests and may not reflect the environmental fate of grease in soil or mixed waste streams. Moreover, no widely accepted performance classifications exist that integrate tribological durability with environmental criteria for green greases. Establishing harmonised standards and application-relevant qualification protocols will be essential to support broader industrial uptake [52, 108-111].

9 Future directions and research prospects

Looking ahead, green grease is poised to become a cornerstone of sustainable lubrication technology. With increasing regulatory pressure, carbon neutrality goals, and consumer demand for safer, eco-friendly products, industries are actively seeking alternatives to traditional petroleum-based greases. Future development will likely focus on enhancing the performance of green greases to meet or exceed those of conventional products. This includes improving thermal and oxidative stability, load-bearing

capacity, and long-term durability through advanced formulation strategies and novel bio-based materials such as functionalized lignin, cellulose derivatives, or biodegradable nanoparticles. Lowering production costs and developing global standards will also support wider adoption. Collaboration between industry, academia, and regulatory bodies will be vital. In summary, green grease has strong potential to deliver efficient and safer lubrication while reducing environmental impact.

Notes

The authors declare no competing financial interest.

Acknowledgements

The authors thank the financial support from the Swedish Research Council for Environment, Agricultural Sciences and Spatial Planning (Formas, Project No. 2020-01258 and 2022-01047).

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Braham PRAKASH. He obtained his BSc Eng (mechanical) degree from Punjab Engineering College, Chandigarh (India), and M Tech (mechanical engineering) as well as Ph.D. (tribology) degrees from Indian Institute of Technology Delhi (India). He is a professor emeritus at the Division of Machine Elements of Luleå University of Technology in Sweden since 2019 and a distinguished visiting professor at the Department of Mechanical Engineering of Tsinghua University in China since 2018. He has earlier been professor and head of Tribolab at the Division of Machine Elements of Luleå University of Technology from 2002 to 2019. He has also been a faculty member at Indian Institute of Technology Delhi from 1981 to 2002, a visiting professor at Tokyo University of Science, Japan (2016) and Indian Institute of Technology, Ropar, India (2010–2013), Japan Society for the Promotion of Science (JSPS) postdoctoral fellow, Chiba Institute of Technology, Japan (1998–2000) and visiting researcher at Tokyo Institute of Technology, Japan (1985). Professor Prakash has over 45 years of experience, both in academia and industry. His research and teaching activities cover several aspects of tribology such as high temperature tribology, friction and wear of materials, solid and boundary lubrication, surface engineering, tribology of machine components, tribology of wheel-rail interface, tribotesting, and analysis and solution of tribological problems. He has mentored over 10 postdoc researchers, 18 Ph.D. candidates, and numerous students. He has made over 250 contributions in reputed journals, international conferences, and books.



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Just Accepted