

Tribological Properties of Synthetic and Biosourced Lubricants Enhanced by Graphene and Its Derivatives: A Review

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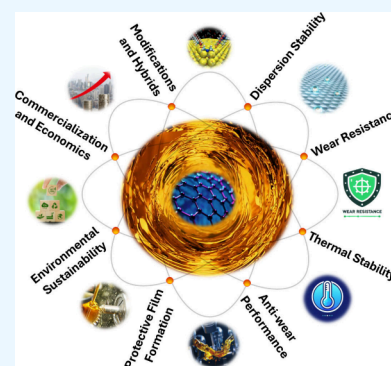
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ABSTRACT: This review explores the tribological properties of biosourced lubricants (biolubricants) enhanced by graphene (Gr) and its derivatives and hybrids. Friction and wear at mechanical interfaces are the primary causes of energy loss and machinery degradation, necessitating effective lubrication strategies. Traditional lubricants derived from mineral oils present environmental challenges, leading to an increased interest in biolubricants derived from plant oils and animal fats. Biolubricants offer high biodegradability, renewability, and low toxicity, positioning them as ecofriendly alternatives. This work extensively reviews the role of Gr-based nanoadditives in enhancing the lubrication properties of biolubricants. Gr with its exceptional physicochemical properties has shown promise in reducing friction and wear. The review covers various Gr derivatives, including Gr oxide (GO) and reduced Gr oxide (r-GO), and their performance as lubrication additives. The discussion extends to Gr hybrids with metals, polymers, and other 2D materials, highlighting their synergistic effects on the tribological performance. The mechanisms through which these additives enhance lubrication, such as the formation of protective films and improved interactions between lubricants and tribo-pairs, are examined. Emphasis is placed on the environmental benefits and potential performance improvements of Gr-based biolubricants. Finally, by analyzing current research and technological trends, the paper outlines future prospects for optimizing lubricant formulations with Gr-based nanoadditives, aiming for more sustainable and efficient tribological applications.



INTRODUCTION

Friction and wear are prevalent in mechanically engaged interfaces and are key contributors to energy loss and mechanical degradation of many industrial machinery parts.^{1,2} This major problem underscores the urgency of managing and reducing friction to improve productivity and sustainability.^{3,4} Overall, unchecked friction can lead to rapid deterioration of mechanical components, resulting in reduced service life and reliability.^{5,6} Thus, lubrication is a pivotal method for mitigating friction and wear in tribo-pairs (interacting machine components). Historically, lubricants derived from vegetable oils, animal fats, and aqueous sources have been utilized.⁷ At the same time, contemporary advancements in lubrication technologies using advanced materials have significantly enhanced our ability to address tribological challenges.⁸ In general, lubricants are categorized into solid and liquid forms. Solid lubricants, though effective, may suffer from wear-out due to environmental factors, such as detrimental interactions with oxygen and water, thereby limiting their durability.⁹ On the other hand, liquid lubricants (notably mineral and engine oils) are preferred in mechanical industries for their robust antifriction capabilities. These lubricants are understood to form protective films on tribo-

pairs, reducing metal-to-metal contact.¹⁰ However, challenges arise under extreme conditions, such as engine start-up or shutdown, which has prompted research into improved boundary lubrication by utilizing additives in base oil formulations.¹¹ These additives have been shown to enhance lubrication, high-temperature viscosity, oxidation resistance, and wear-reduction properties of the base oil.¹² While traditional additives like zinc dialkyldithiophosphate (ZDDP) and molybdenum dithiocarbamate (MoDTC) offer stability and performance to oil, their environmental and corrosion concerns have led to the exploration of alternatives, including nanomaterials, ionic liquids, and organic compounds.^{10,13} Notably, carbonaceous nanoadditives have emerged as promising alternatives, demonstrating significant improvements in the lubrication and antiwear properties of oil.¹⁴ Research on carbonaceous materials as nanoadditives for

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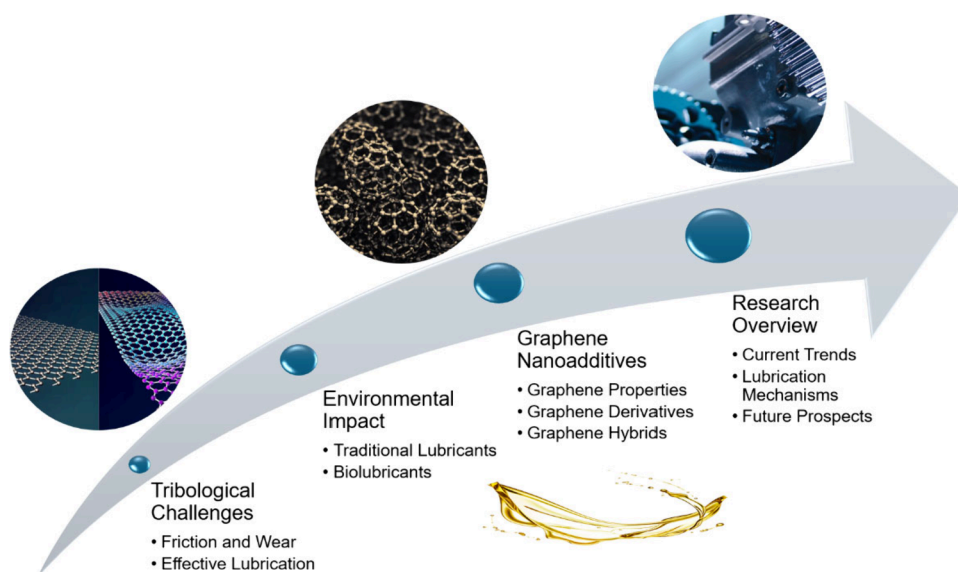


Figure 1. Structure of the current review paper.

lubricants is still ongoing, with graphene (Gr), graphite, fullerenes, and diamonds as typical examples. Gr is notable for its superior physicochemical and electrical properties.^{15,16} This two-dimensional carbonaceous nanomaterial is extensively used in both micro- and macro-tribological applications.¹⁷ Key derivatives of Gr include Gr nanoplatelets (GnPs), Gr oxide (GO), and reduced Gr oxide (r-GO), which differ in chemical composition and exhibit distinct chemical and structural properties. These Gr derivatives, especially GO and r-GO, have gained prominence in tribological research due to their exceptional performance as water-based and oil-based lubrication additives.¹⁸

Biosourced lubricants (biolubricants), which are primarily derived from plant oils and animal fats, have gained much attention as ecofriendly alternatives to conventional mineral-based lubricants. They are especially preferred over the latter lubricants due to their high biodegradability, renewability, and low toxicity.¹⁹ Today, biolubricants are utilized not only in industrial applications, but also in pharmaceuticals, cosmetics, and other consumer products.^{20–22} They comprise triglycerides, phytosterols, natural pigments, phospholipids, and fatty acids and provide an effective protective film on contact surfaces to mitigate friction and wear.^{23,24} Essential properties of an effective lubricant include good thermal stability, high viscosity index, low freezing point, high boiling point, and resistance to oxidation.^{25,26} Recently, the incorporation of Gr-based nanoadditives into biolubricants has been explored to further improve their lubrication properties. The development and discussion of this important aspect of the Gr bibliography would be a huge addition and a necessity for the development of this field of research. As this domain continues to expand at a rapid rate, reviews and surveys on the most recent advancements of Gr as a nanoadditive for biolubricants are much needed. In this respect, there is an immediate demand for a timely, thorough, and in-depth review on the latest advances in Gr-based biolubricants, which hold great potential as interesting 2D carbonaceous nanoadditives for tribology applications. This review addresses the application of Gr, its derivatives, and hybrids as nanoadditives specifically in biolubricants, an area that has not received as much attention as studies involving synthetic or mineral-based lubricants.^{27,28}

Unlike most previous research that has largely focused on synthetic lubricants, this work shifts the focus toward Gr-based nanoadditives in biolubricants, which are derived from renewable sources and support the goal of sustainable industrial practices. The study systematically categorizes these nanoadditives, including those modified with metallic elements, polymer coatings, and 2D heterostructures, revealing how different modifications impact friction reduction, wear resistance, and thermal stability.^{29,30} It further delves into the mechanisms by which GO and r-GO enhance tribological performance, emphasizing their role in forming protective films and enhancing lubricant interactions at the molecular level.^{31,32} Additionally, the review examines the synergistic effects observed with Gr-based nanoadditives and biolubricants, demonstrating that these combinations can outperform conventional mineral-based lubricants and provide insights into advancing sustainable tribological technologies from laboratory research to practical industrial applications.

To provide further focus and context to these efforts, the impact of Gr and its derivatives and hybrids on the tribological properties of biolubricants is reviewed herein. Specifically, the context and importance of using biolubricants in various industries are introduced first, followed by a detailed account of the use of Gr and its derivatives and hybrids as potential nanoadditives in biolubricants. This includes discussions on the classifications of Gr-based nanoadditives based on their modifications with metallic elements, polymers, and integration in 2D heterostructures. Emphasis is given to explaining how the exceptional properties of Gr can be harnessed to improve lubrication. The subsequent sections examine GO and r-GO as lubricant nanoadditives, showcasing their tribological performance in liquid lubricants. The mechanisms through which these nanoadditives influence lubrication are further explained, including their roles in forming protective films and enhancing the interactions between the lubricant and tribo-pairs. Next, the discussion shifts toward biolubricants, emphasizing their environmental benefits and observed synergistic effects with Gr-based nanoadditives. Finally, the future prospects of utilizing Gr-based nanoadditives in biolubricants are discussed, with a focus on the environmental benefits and potential performance improvements. The

structure of the review paper is summarized in Figure 1. By analyzing current research and technological trends, this review outlines the path forward for optimizing lubricant formulations with Gr-based nanoadditives to enable more sustainable and efficient tribological applications.

■ GRAPHENE AND ITS DERIVATIVES

Based on their dimensions, carbon nanostructures are categorized into 0D, 1D, 2D, and 3D. 0D structures include fullerenes, carbon dots (CDs), and Gr quantum dots (GQDs), 1D nanostructures encompass carbon nanotubes (CNTs) and carbon nanofibers (CNFs), and 2D nanostructures include Gr and graphitic carbon nitride (g-C₃N₄). Among these, Gr has attracted much attention across various scientific and technological fields due to its unique properties.³³ 3D carbon nanostructures often combine 1D and 2D nanostructures, such as Gr superstructures and nanotube-Gr hybrids. Gr, a 2D nanostructure, was first isolated using a simple sticky tape technique by Geim and Novoselov, sparking a revolution in research across multiple disciplines.³⁴ It is celebrated for its exceptional physicochemical characteristics, such as high thermal conductivity, mechanical strength, and biocompatibility, making it one of the strongest, thinnest, and lightest materials at the atomic level.³⁵ Gr is a single layer of sp²-bonded carbon atoms arranged in a honeycomb lattice, giving it a vast surface area, and making it impermeable even to helium atoms.³⁶ Its applications span biomedical, energy, environmental, tribology, biosensors, and water desalination fields, among others.^{15,37,38} Despite its hydrophobic nature and stability in air up to 200 °C, Gr tends to aggregate in aqueous media and is insoluble in organic solvents.³⁶ The commercial production of Gr faces challenges due to the diversity of synthesis techniques, broadly classified into top-down and bottom-up approaches.³⁹ The top-down method, suitable for industrial-scale production, includes exfoliation, electrochemical methods, and laser ablation. At the same time, the bottom-up approach, yielding high-quality Gr with superior electrical properties, comprises chemical vapor deposition, arc discharge, pyrolysis, and plasma synthesis.

Gr stands out for its exceptional antifriction and antiwear properties, setting it apart from conventional materials. It can serve as both a solid and colloidal liquid lubricant, leveraging its notable thermal, electrical, optical, and mechanical properties. The keys to its superior tribological performance include high chemical inertness, significant mechanical strength, and the ability to easily shear on its atomically flat surface, which reduces wear on contacting surfaces. Research by Lee et al.⁴⁰ highlighted Gr as one of the strongest materials known, with its atomically smooth surface and low surface energy, enhancing its role in minimizing adhesion and friction between tribo-pairs.⁹ Gr and its derivatives GO and r-GO differ slightly in their physicochemical properties but share a similar 2D structure.⁴¹ The electrical conductivity of GO is lower due to its oxygen-rich functional groups, but it can be converted to r-GO to achieve properties akin to Gr, which are not attainable from pure graphite.

■ CLASSIFICATIONS OF GRAPHENE-BASED NANOMATERIALS

Unmodified or Structurally Imperfect Gr. The intrinsic lamellar features of Gr and its structural features and defects, such as the number of layers, interlayer spacing, surface

characteristics, atomic vacancies, and edge bonds, critically influence its lubrication properties. These factors impact the structural strength, shear barriers, and stress distribution, which are essential under extreme conditions.^{10,42} The tribological performance of Gr is determined by layer configurations and spacing, which influence surface energy and the potential for frictional contact.^{43,44} Gr nanosheets synthesized through chemical red-ox or mechanical exfoliation show improved tribological due to structural evolution toward order under frictional forces.^{45,46} However, atomic-scale variations like vacancies and heterogeneity can lead to inconsistent experimental results.^{47,48} Even minimal atomic vacancies can significantly increase the friction coefficient due to the enhanced reactivity of adjacent bonds and strain distribution.⁴⁸ Additionally, atomic doping with elements such as nitrogen enhances, whereas boron deteriorates tribological performance by altering the potential energy corrugation during sliding, with nitrogen reducing and boron increasing the frictional forces.^{47,49,50}

Metal-Modified Gr. Metal-based nanomaterials such as Cu, Ag, Zn, and Sn enhance lubrication under harsh conditions by forming robust boundary tribofilms via adsorption, infiltration, and chemical reaction at interfaces.^{51–53} Modifying the Gr surface with metal nanoparticles leverages the film-forming abilities and shearing characteristics of Gr to optimize lubrication in demanding environments. For example, research by Gan et al.⁵⁴ introduced a novel GO/Cu nanocomposite developed through a liquid-phase method, which, when added to polyethylene glycol, markedly improved its friction and wear resistance by up to 47%. Additionally, Ag nanoparticles are recognized for their effectiveness of lubrication, though their application is hindered by agglomeration and complex synthesis.⁵⁵ A single-step laser irradiation method has been developed to modify Gr nanosheets with Ag nanoparticles, improving its lubrication and wear resistance through self-lubrication, rolling effects, and a self-healing mechanism.^{55,56} The lubrication efficacy of Gr is dependent on the optimal size and density of metal nanoparticles, which prevent agglomeration and enhance synergistic lubrication effects.^{54,57} Metals such as Al, Mn, Fe, and Ni, which readily oxidize, form oxides that improve the lubrication and mechanical properties of Gr nanosheets.^{58,59} Integrating metal oxide/Gr nanocomposites into self-lubricating substrates elevates tribological properties and mechanical strength.^{60,61} For example, Al₂O₃/Gr dispersed in a tungsten carbide ceramic matrix notably increases lubrication, refines grain size, and enhances hardness (from 16.18 to 18.34 GPa) and fracture strength (from 8.08 to 11.02 MPa m^{1/2}).⁶¹ Similarly, Gr nanosheets coated with Fe₃O₄ nanorods improve thermal stability, mechanical strength, and tribological performance in bismaleimide substrates.⁶⁰ Moreover, metal oxide nanoparticles anchored to Gr reduce van der Waals interactions, facilitating better dispersion in lubricants and increasing the lamellar spacing for enhanced lubrication.^{51,62} A prominent example includes Mn₃O₄/Gr nanocomposites, prepared via hydrothermal and green synthesis, which achieve uniform dispersion and deeper penetration of nanoparticles into Gr layers for improved stability and lubrication.⁵⁹

Polymer-Modified Gr. Polymers such as polytetrafluoroethylene (PTFE), polyether ether ketone (PEEK), and polyimide (PI) are recognized for their self-lubricating qualities, which are crucial in demanding lubrication environments.^{63,64} By manipulating their molecular weight through

synthesis conditions, diverse morphologies and structures are achieved to meet specific lubrication needs.^{64,65} Gr nanosheets, known for their large specific surface area, serve as a robust base for polymer growth, thus enhancing lubrication performance by increasing its adsorption energy and film-forming ability in severe conditions.^{64,66} This synergy has spurred interest in polymer-modified Gr for improved lubrication. Polymer nanomaterials like polydopamine (PDA), inspired by mussel proteins, are used for surface modification of Gr and as intermediates for grafting nanoparticles, to improve its lubrication and mechanical properties.^{63,66} PDA not only enhances van der Waals forces and the adsorption capability but also acts as a cross-linker for grafting nanoparticles, such as Cu, onto Gr, combining the benefits of Gr and nanoparticles while improving the anchoring strength.^{66–68}

PI nanocomposites are utilized for their mechanical strength, thermal stability, and lubrication properties.^{69,70} PI modifies the Gr surfaces through reactions that adjust van der Waals forces and π – π interactions to optimize lubrication characteristics.^{70,71} Additionally, Gr serves as an effective nanoadditive in self-lubricating polymers such as PEEK and PTFE, reinforcing the polymer matrix to enhance wear resistance and mechanical properties, particularly under high contact stress.^{72,73} It also boosts thermal diffusion in high-temperature applications.^{72,74,75}

■ GRAPHENE-BASED 2D HETEROSTRUCTURES

Monolayers of 2D nanomaterials have adjustable stacking structures thanks to van der Waals forces between the layers, which affect their properties.^{76,77} Combining these nanomaterials into heterostructures can enhance their properties, such as superlubricity, making them suitable for harsh conditions. For example, a Gr-MoS₂ heterostructure improves lubrication under high stress by forming a durable tribofilm. In contrast, Gr-hexagonal boron nitride (hBN) heterostructures achieve superlubricity through incommensurable contact due to differing lattice constants.^{78,79} These heterostructures, including Gr-transition metal dichalcogenides (TMDs) and Gr-MXenes, demonstrate great potential for lubrication performance enhancements.^{79–81}

Gr-hBN Heterostructures. Gr nanosheets with their excellent mechanical properties, high thermal stability, and low shear strength serve as effective solid lubricants or lubricant nanoadditives. Similarly, hBN offers high thermal stability and self-lubrication due to its strong covalent bonds.⁴² Gr-hBN heterostructures can be created through covalent or noncovalent methods, exhibiting enhanced tribological performance. For example, covalently linking oxidized hBN with GO through chemical functionalization, as demonstrated by Samanta et al.,⁸² improves lubricant adhesion, tribofilm formation, and friction stability. Noncovalent stacking exploits the incommensurability between Gr and hBN for superior lubrication, leveraging the difference in interlaminar shear strength to reduce friction. Qi et al.'s⁸³ simple method of synthesizing noncovalent Gr-hBN materials by drying their solution on silicon wafers shows promise despite potential nanosheet agglomeration issues. These heterostructures, whether covalently or noncovalently bound, showcase advanced friction properties through the manipulation of chemical and physical interactions at the nanoscale.

Gr-TMD Heterostructures. Despite its superb lubricating qualities, Gr faces limitations in engineering applications due to its poor ability to form boundary tribofilms at frictional

interfaces, which is attributed to its high thermal stability and chemical inertia.^{42,84} TMDs, which include sulfur, readily form covalent bonds at frictional interfaces under mechanical stress and frictional heat, thereby creating a durable boundary tribofilm.^{79,85} The synthesis of Gr-TMD heterostructures through in situ growth or mechanical stacking enhances lubrication performance significantly, with MoS₂ and WS₂ being the primary TMDs used for lubrication.^{81,86} The unique structure of MoS₂, which is characterized by a sandwich-like assembly and weak van der Waals forces, facilitates shear processes. The natural lattice mismatch between Gr and MoS₂ and the resulting Moiré patterns contribute to low friction states.⁸⁷ Various methods, such as hydrothermal treatment and grafting with cross-linking agents, have been employed to yield Gr-MoS₂ heterostructures with superior dispersion stability in lubricants thanks to van der Waals interactions that minimize nanomaterial agglomeration.^{79,88} Similarly, WS₂ shares structural similarities with MoS₂, offering low friction, high film-forming capability, and excellent lubrication under extreme conditions.⁴² Techniques involving layer-by-layer transfer of WS₂ onto Gr have produced heterostructures with superlubricity under ambient conditions and enhanced the lubrication and mechanical strength of polymers.^{89,90} The lattice mismatch within these heterostructures promotes effective lubrication, while the mechanical strength of Gr improves wear resistance and extreme-pressure lubrication performance.⁹¹

Gr-MXene Heterostructures. MXenes are synthesized by selectively etching an A element (Group IIIA or IVA) from the MAX phase, resulting in a structure consisting of a transition metal and carbon or nitrogen.⁹² Through etching and surface functionalization, MXenes exhibit diverse structural changes such as variations in lamellar spacing, layer count, and terminal functional groups. These attributes significantly enhance their surface potential and van der Waals interactions, making them suitable for use in extreme lubrication conditions, particularly in high-temperature and high-stress environments.^{42,81,93} The integration of MXenes with Gr to form 2D heterostructures has been increasingly explored for lubrication purposes.^{81,86} For example, combining Gr and MXene on silicon substrates has enhanced lubrication and resulted in superlubricity and improved wear resistance against diamond-like carbon (DLC)-coated steel balls in dry nitrogen environments.⁹⁴ Also, uniform Ti₃C₂-GO nanocomposite coatings produced via electrophoretic deposition have been shown to significantly reduce friction, illustrating the potential of these heterostructures in advanced lubrication applications.⁸⁶ Gr-MXene heterostructure nanoadditives have demonstrated the potential to improve heat conduction in lubricants and reduce oil viscosity, thereby enhancing thermal and tribological properties.⁹⁵ For example, adding nitrogen-doped Gr-Ti₃C₂ to engine oil was observed to increase thermal conductivity by 6.62% and reduce viscosity by 4.71%.⁹⁵ Additionally, when used in polymer self-lubricating materials, such as an epoxy coating filler, they have been shown to significantly reduce wear rates (by 81–88%) compared to pure epoxy and boost anticorrosion properties by preventing corrosive media diffusion.⁹⁶

■ GO AND r-GO AS LUBRICANT NANOADDITIVES

Gr derivatives, particularly GO and r-GO, are explored extensively due to their aromatic rings, free π – π electrons, and reactive functional groups.⁹ GO is a single-atom-thick carbon sheet adorned with oxygen-containing functional

Table 1. Dispersion of Gr and Its Derivatives and Hybrids in Liquid Lubricants

Nanoadditive	Lubricant	Dispersion Procedure	Stability	Method	ref
Gr	PAO ₄	surfactant (Span80) + ultrasonication	~4 weeks	physical	104
in situ exfol. Gr	water	surfactant (Triton X-100) + ultrasonication	>a month	physical	105
exfol. graphite	aqueous medium	surfactant (SDS) + mechanical stirring	-37.09 mV ^a	physical	106
exfol. graphite	aqueous medium	surfactant (SDBS) + mechanical stirring	-40.20 mV ^a	physical	106
exfol. graphite	aqueous medium	surfactant (CTAB) + mechanical stirring	43.90 mV ^a	physical	106
exfol. graphite	aqueous medium	surfactant (NPE) + mechanical stirring	-23.70 mV ^a	physical	106
Gr	simaroua biodiesel	surfactant (SDS) + ultrasonication	~8 weeks	physical	107
Gr	simaroua biodiesel	surfactant (SDBS) + ultrasonication	~8 weeks	physical	107
r-GO	engine oil	inclusion of long alkyl chains through amide linkages + mechanical stirring	~30 days	Chemical	57
r-GO	petroleum	covalent functionalization + mechanical stirring	~30 days	Chemical	108
r-GO	polyol lube	alkylation + mechanical stirring	~20 days	Chemical	109
r-GO	hydroisomerization dewax oil	alkylation + mechanical stirring	>24 days	Chemical	110
r-GO	PAO6	alkylation + mechanical stirring	>120 days	Chemical	111
r-GO	10W40 oil	alkylation + mechanical stirring	>a month	Chemical	112
Gr	PAO10 and TMP	mechanical stirring	~ 1 day	Physical	113
crumbled Gr ball	PAO4	mechanical stirring	~ 1 day	Physical	114
highly exfol. Gr	PAO6	mechanical stirring	~ 4 days	Physical	61
superhigh exfol. Gr	hydraulic oil	mechanical stirring	~ 10 days	Physical	62
ZnO/Gr	ester oil	mechanical stirring	>5 days	Physical	115
WS ₂ /Gr	PAO4	mechanical stirring	~ 7 days	Physical	116
r-GO	paraffin	mechanical stirring	~ 1 day	Physical	117
Sc-Au/GO	paraffin	mechanical stirring	~ 10 days	Physical	118
MoS ₂ /GO	sunshine oil	mechanical stirring	~ 5 days	Physical	119
ZrO ₂ /r-GO	paraffin	mechanical stirring	~ 2 days	Physical	120
Ag/r-GO	liquid paraffin	mechanical stirring	~ 60 days	Physical	121
PEG/Gr	water	mechanical stirring	~ 7 days	Physical	122

^aZeta potential.

groups, making it significantly different from Gr.⁹⁷ In general, GO is less electrically conductive, does not absorb visible light, and is more chemically active. Unlike Gr, its solubility in water and other solvents presents an advantage for industrial applications. The production of GO involves methods such as oxidation and exfoliation of graphite that lead to different structures, such as those represented by the Lerf-Klinowski model, widely accepted based on solid-state nuclear magnetic resonance (NMR) and X-ray diffraction (XRD) analyses.⁹⁸ The oxidation process introduces structural imperfections into GO, affecting its electrical, optical, and adsorption properties. To counter this, GO can be converted into r-GO using chemical, thermal, microwave, or photochemical reduction methods, which results in films with significantly reduced oxygen content.^{99,100} Despite its promising tribological applications, the poor dispersibility of Gr in liquid lubricants due to its large surface area poses challenges. Surface modification of Gr has been explored to improve its dispersibility, but these may have negative environmental impacts.¹⁰¹ Gr derivatives enhance liquid lubricants by providing low shear resistance, an appropriate size for reaching contact regions, and effective heat distribution. The interaction types, including electrostatic, van der Waals, covalent, and noncovalent bonding, influence the dispersibility and chemical stability of GO in solutions, potentially offering significant improvements in antiwear and antifriction properties in applications such as engine oils. The conversion of GO to r-GO enhances its compatibility with nonpolar liquids such as oils by reducing its hydrophilic characteristics, thus improving the compatibility. The high thermal conductivity of GO makes it a valuable addition to cutting fluids for machining tough

materials.¹⁰² However, its interaction with organic lubricants can introduce defects. r-GO nanoadditives, being more affordable and having fewer morphological defects than GO, present a promising option for sustainable lubrication solutions with their large surface area, high mechanical strength, and excellent oxidation resistance.¹⁰³ A summary of the dispersion procedures reported in the literature for different Gr-based nanoadditives in liquid lubricants is provided in Table 1.

Enhancing Liquid Lubricants with GO and r-GO: Tribological Properties and Applications. Gr derivatives, recognized for their superior characteristics and stability, are promising as lubricant nanoadditives and solid lubricants (applied through deposition techniques). However, their effectiveness as solid lubricants is limited due to wear and susceptibility to environmental conditions. As a solution, researchers have explored the potential of Gr in enhancing liquid lubricants, including water-based, synthetic, and bio-oils. Gr nanoadditives, especially GO and r-GO, have been shown to significantly reduce wear and friction in different oils. The tribological performances of these lubricants depend on factors such as Gr concentration, temperature, speed, load, tribo-pairs, and base oils.

Water-based lubricants are essential in various engineering applications, such as oil extraction, cutting, polishing, and hydraulic operations. They are favored for their cost-effectiveness, availability, excellent heat conduction, and environmental compatibility. Despite their poor compatibility with materials such as steel, the integration of GO into these lubricants has shown promising results. The solubility of GO is derived from its carbon-oxygen groups, which facilitates its dispersion in liquid media through ultrasonication and

Table 2. Tribological Performance Enhancements of Lubricants with Graphene-Based Nanoadditives

nano additives	synthesis method	base oil	friction materials (conditions)	COF	improvement in COF (%)	wear performance	improvement in wear (%)	ref
W ₂ C/GnP	API Group III chemical reduction	Group III Base Oil	AISI 8620 steel (ball-on-flat, 1 mm/s, 1.4 GPa), SiC (ball-on-flat, 10 m/s, 1.8 GPa)	~0.095 (0.05 wt %)	30%	~0.42 mm (wear depth)	25%	141
MG/Gr	IRIS-200BB grease	Mineral Oil	GCr15 steel (four-ball, 1200 r/min, 392 N)	~0.103 (1 wt %)	20%	~0.52 mm (wear scar)	18%	142
Fe ₃ O ₄ /GnP	BMI resin (coprecipitation)	Synthetic Oil	MM-200, 196 N	~0.07 (0.6 wt %)	40%	3×10^{-6} mm ³ /Nm (wear rate)	32%	143
Al/GnP	Cutting fluid (physical mix)	Water-Based Cutting Fluid	AISI 304 steel (pin-on-disc, 2000 rpm, 1000 N)	~0.1 (1.25 wt %)	15%	~0.332 μm (pin wear)	12%	135
CeO ₂ /GnP	Paraffin (hydrothermal)	Paraffin Oil	GCr15 steel (SRV-1, 6 cm/s, 50 N)	~0.1 (0.06 wt %)	25%	~0.45 μm (wear rate)	20%	117
α-Fe ₂ O ₃ /GO	Paraffin (hydrothermal)	Paraffin Oil	GCr15 steel (four-ball, 3.412 m/s, 19.6 N)	~0.2 (0.5 wt %)	10%	~0.1 mm (wear scar)	15%	40
ZrO ₂ /r-GO	Paraffin (hydrothermal)	Paraffin Oil	GCr15 steel (ASTM D6425-05, 6 cm/s, 100 N)	~0.12 (0.06 wt %)	28%	2.2×10^{-3} mm ³ /Nm (wear rate)	22%	120
SiO ₂ /GnP	Water (physical mix)	Water	Magnesium alloy/steel (ball-on-plate, 0.08 m/s, 3 N)	~0.062 (0.1:0.4 wt %)	35%	5×10^{-3} mm ³ /Nm (wear volume)	30%	136
SiO ₂ /GO	Ethylene glycol (chemical reduction)	Ethylene Glycol	Stainless steel (pin-on-disc, 0.008 m/s, 68.9 N)	~0.11 (0.125 wt %)	22%	3.5×10^{-6} mm ³ /Nm (wear rate)	25%	141
MoS ₂ /GnP	Dibutyl phthalate (hydrothermal and CVD)	Dibutyl Phthalate	GCr15 steel (four-ball, 1200 rpm, 392 N)	~0.075 (0.02 wt %)	38%	~0.825 mm (wear scar)	28%	42
MoS ₂ /GnP	Polyalkylene glycol (chemical reduction)	Polyalkylene Glycol	Stainless steel (SRV-IV, 700HV)	~0.08 (0.5 wt %)	36%	1×10^{-4} mm ³ /Nm (wear volume)	30%	144
MoS ₂ /GnP	Esterified bio-oil (physical mix)	Bio-Oil	ASTM E52100 bearing steel (four-ball, 1000 rpm, 300 N)	~0.018 (0.3:0.2 wt %)	60%	~0.45 mm (wear scar)	50%	145
WS ₂ /GnP	PAO4 (mechanochemical)	PAO ₄	GCr15 steel (UMT-2, 5 mm/s, 10 N)	~0.16 (0.01 wt %)	18%	250×10^{-3} mm ³ /Nm (wear rate)	20%	116
Cu/r-GO	Paraffin (chemical reduction)	Paraffin Oil	45# steel (UMT-2, 0.025 m/s, 19.6 N)	~0.105 (1.5 wt %)	27%	~0.1 mm (wear scar)	30%	146
Cu/r-GO	PAO (chemical reduction)	PAO	AISI-52100 steel (four-ball, 1450 r/m, 392 N/490 N)	~0.055 (0.5 wt %)	40%	~0.35 mm (wear scar)	35%	147
Se-Cu/GO	Paraffin (chemical deposition with supercritical CO ₂)	Paraffin Oil	GCr15 steel (four-ball, 1200 rpm, 200 N)	~0.065 (0.05 wt %)	42%	~0.26 mm (wear scar)	38%	39
L-Ag/r-GO	Paraffin (caser irradiation)	Paraffin Oil	GCr15 steel (four-ball, 1200 rpm, 392 N)	~0.065 (0.1 wt %)	20%	~0.5 mm (wear scar)	25%	121
Se-Ag/GnP	Engine oil (chemical reduction)	Engine Oil	GCr15 steel (four-ball, 1200 rpm, 343 N)	~0.08 (0.75 wt %)	32%	~0.38 mm (wear scar)	28%	148
Se-Au/GO	PAO6 (chemical reduction)	PAO6	GCr15 steel (MS-T3000, 0.1 m/s, 10 N)	~0.08 (0.1 wt %)	35%	~0.03 mm ³ /Nm (wear rate)	40%	118

significantly enhances its antifriction and wear properties compared to Gr. Studies have demonstrated that a 1 wt % GO concentration can notably decrease friction and wear in steel/steel tribo-pairs through tribofilm formation.¹⁷ Further research has consistently shown the superior performance of GO in reducing the coefficient of friction (COF) and wear under various conditions, with its ability to form tribo-chemical films playing a crucial role.^{123–125} Research has also explored the synergistic effects of GO with other nanoparticles, such as nanodiamonds (NDs) and inorganic particles, such as Al₂O₃, yielding further improvements in lubrication quality. For example, a combination of 0.1 wt % GO and 0.5 wt % ND was shown to significantly reduce COF to about 0.03 and wear track depth to 5 nm, which were attributed to the low shearing resistance of Gr, effective tribofilm formation, and the ball-bearing effect of ND.¹²⁶ Similarly, the GO-Al₂O₃ blend was shown to enhance the antifriction and wear properties, as well as surface quality, indicating that a balanced mass ratio of these nanoparticles can optimize the tribological performance.¹²⁷ These findings highlight the potential of GO and its nanocomposites to enhance the performance of water-based lubricants across varied operational conditions and material pairings, making them highly effective for industrial applications.

The use of GO and r-GO as lubricants in nonaqueous environments has been a focal point of global research due to their proven efficacy in reducing friction and wear. Studies have consistently shown that these materials can significantly enhance lubrication performance by forming stable tribofilms between contact surfaces. For example, Eswaraiah et al.³⁰ demonstrated that a small addition of GO to engine oil reduced the COF by 5-fold at a concentration of 0.025 mg/mL and increased its load-carrying capacity. Research by Senatore et al.¹²⁸ found that 0.1 wt % GO in SN 150 oil effectively improved lubrication in boundary and elastohydrodynamic lubrication regimes, markedly reducing both COF and wear. Further investigations report consistent reductions in friction and wear with GO inclusions in various oil formulations, with effects such as a 16% average reduction in friction and up to 30% in wear, noted by Sarno et al.¹²⁹ under mixed lubrication conditions. Wu et al.³² found that an aviation lubricant with 0.5 wt % GO under Si₃N₄/steel contact exhibited reduced friction and wear by 15% and 34%, respectively. Moreover, Javeed et al.¹³⁰ achieved a 37% improvement in wear resistance with GO nanoadditives in a diesel engine oil, suggesting that even lower GO concentrations might benefit certain applications. These findings collectively underscore the potential of GO and r-GO-based lubricants, offering significant improvements in wear resistance, friction reduction, and load-bearing capacities in different lubrication applications.

To mitigate the aggregation of GO nanosheets in lubricants, Kinoshita et al.¹³¹ developed a polyalphaolefin (PAO)-based lubricant with varying dispersibility of GO, utilizing alkylamine for dispersion or heating between 130 and 250 °C. They discovered that lower dispersibility facilitated better coverage of the contact surfaces by GO aggregates, achieving a COF as low as 0.05, which suggests that sliding GO layers reduce COF by targeting weak shear layers. Despite the effectiveness of conventional oils with additives, their environmental impact promotes the use of biolubricants, with Anand et al.¹³² finding optimal tribological performance in a 0.75% GO-dates bio-oil blend. However, higher GO concentrations may decrease lubrication by increasing surface abrasion. r-GO, recognized for

its cost-effectiveness and fewer morphological imperfections, has shown promise as a lubricant nanoadditive. Patel et al.¹³³ reported that 0.01 wt % r-GO in ester oil significantly reduced friction and wear, while improving dispersibility. Comparisons between GO and r-GO in a PAO oil by Zhao et al.¹³⁴ indicated that 0.5 wt % r-GO considerably lowered COF and wear thanks to consistent particle size and effective tribofilm formation. The impact of r-GO microstructure on its performance was notable, with regular-edged r-GO outperforming other forms in reducing friction and wear.¹¹⁷ Furthermore, modifications to r-GO to eliminate structural defects such as folds and wrinkles have been successful in enhancing tribological behavior, demonstrating the potential even in small amounts. For example, Kaleli et al.¹³⁵ showed a notable enhancement in tribological performance with just 0.02 wt % r-GO in synthetic engine oil, highlighting a reduced COF by 5% under boundary lubrication conditions. GO and its derivatives, typically challenged by agglomeration in oils due to their inorganic nature, require surface modification to enhance the oil solubility and stabilize dispersions. Advancements in chemically modifying GO have significantly improved the tribological properties of the base oils. For example, Lin et al.¹³⁶ demonstrated that lubricants containing 0.075 wt % modified GO, using stearic and oleic acids, enhanced antiwear and load-bearing capacities. Similarly, alkylated GO achieved better dispersion and significantly reduced the COF and wear scar diameter.¹³⁷ Cheng et al.¹³⁸ incorporated oleic diethanolamide borate-grafted GO into 500 SN base oil and reported substantial reductions in COF and wear scar diameter at just 0.02 wt % GO. Ionic liquid (IL) surface functionalization has also shown promise. For example, Fan et al.¹⁰⁵ reported that alkyl imidazolium ILs used for GO modification improved dispersion stability and decreased friction and wear by notable margins. Hybrid nanostructure materials, particularly Gr nanocomposites with nanomaterials such as MoS₂, have shown potential in developing advanced lubricants with enhanced tribological properties. Studies have demonstrated that Gr/MoS₂ hybrids in esterified bio-oil significantly improve antifriction, wear resistance, and load-bearing capacities, with a notable reduction in COF from 0.05 to 0.016, which is attributed to the synergistic effects between the mechanical support provided by Gr and low friction of MoS₂.¹⁰⁴ However, not all combinations yielded significant improvements. Indeed, some Gr and MoS₂ hybrids showed negligible effects on tribological performance.¹⁰⁶ Conversely, r-GO/MoS₂ hybrids in paraffin oil reduced COF by 50% and wear track depth significantly, highlighting the benefits of tribofilm development.¹⁰⁷ Studies on r-GO/metal oxide hybrids, such as r-GO/TiO₂, revealed variable outcomes with specific ratios, such as achieving reduced friction but only minor wear reduction. This behavior underscores the complex interplay between the materials and their proportions in optimizing lubricant formulations.^{139,140} A literature summary of the tribological performances of lubricants enhanced with Gr-based nanoadditives is provided in Table 2.

■ LUBRICATION MECHANISM

Incorporating 2D nanomaterials into macro-lubrication systems often results in the formation of protective films on tribo-surfaces, thereby enhancing surface roughness, wear repair, and load bearing capacity, while providing low shear strength.¹⁴ These benefits mainly arise from the lubricating mechanisms of the GO-based nanomaterials, including interlayer lubrication,

Table 3. Lubrication Mechanisms of Gr-Based and Other Nanoadditives

lubrication mechanism	description
Tribofilm formation	Nanoadditives aid in the film formation on tribo-surfaces, enhancing surface roughness, wear repair, load bearing, yielding low shear strength.
Interlayer lubrication	Nanoadditives act as nanoball bearings, filling surface imperfections and reducing friction through interlayer sliding.
Synergism in heterostructures	Combining 2D materials into heterostructures, such as Gr-MoS ₂ and Gr-hBN, improves lubrication under high stress.
Self-repair	Gr nanosheets reduce friction by creating lubricious films and filling surface imperfections, enabling self-repairing mechanisms.
Tribo-chemical reactions and adsorption	Gr-based nanoadditives enhance lubrication via adsorption and chemical reactions, forming tribo-chemical layers.
Fluid effects and molecular manipulation	Superlubricity is achieved through molecular interactions and maintaining a nanometer-thick water layer between the tribo-surfaces.
Formation of robust boundary tribofilms	Metal-based nanoadditives, such as Cu and Ag, enhance lubrication by forming robust boundary tribofilms.
Self-lubrication and rolling effects	Gr combined with other nanoparticles create self-lubrication and rolling effects, as well as a self-healing mechanism.
Improved wear resistance and mechanical properties	Gr-reinforced polymers, such as PTFE and PEEK, enhance wear resistance and mechanical properties under high contact stress.

protective film formation, and self-repairing actions. Liu et al.¹⁴⁹ detailed friction reduction mechanisms with Gr nanosheets, focusing on nanoscale phenomena such as electron–phonon coupling, puckering, and energy dissipation. These mechanisms highlight the importance of Gr-based materials in creating lubricious films, filling surface imperfections, and acting as nanoball bearings to improve lubrication system performance and reduce friction. Understanding the tribology of GO- and r-GO-based lubricants involves examining their lubrication mechanisms, which is still a topic of ongoing research. Researchers have identified several lubrication mechanisms for Gr-based nanomaterials in lubricating fluids, including tribofilm formation, microstructural modification, interlayer sliding, and synergetic actions, etc., emphasizing the complex interplay of factors, leading to lubrication through penetrating tribo-pair contact zones, adhering to tribo-surfaces, and altering fluid viscosity.¹¹² A summary of the different lubrication mechanisms of Gr-based and other nanoadditives is provided in Table 3.

Huang et al.¹²⁷ investigated the lubrication mechanisms of water-based lubricants with GO, Al₂O₃, and a GO-Al₂O₃ composite nanoadditive. Water alone failed to form protective tribo-oxide layers, showing minimal antiwear and friction protection. GO and Al₂O₃ nanoadditives improved performance over water but were less effective compared to the GO-Al₂O₃ composite, which produced a superior tribofilm and reduced shear resistance and polished asperities while removing debris. Mao et al.¹¹⁷ explored the impact of different micromorphologies of r-GO on lubrication. They found that regular-edged sheets formed a more effective, continuous tribofilm that significantly reduced the COF and wear rate compared to irregular-edged or wrinkled sheets, which caused tribofilm fragmentation. Li et al.¹¹¹ studied hybrid nano-hydraulic lubricants with a modified GO nanoadditive, noting that the protective coating of the base oil degraded quickly under harsh conditions. In contrast, the modified hybrid nanosheets formed a durable, Gr-like protective coating that significantly reduced wear and friction by preventing direct contact between the tribopairs.¹¹⁴

■ GRAPHENE-BASED ADDITIVES IN SOLID LUBRICANTS

Gr, a material only one atom thick with a low tendency to stick, significantly improves solid lubrication by minimizing both friction and the tendency to stick to various surfaces. Thanks to its incredibly slim profile, it serves as an excellent lubricant for solid surfaces. The phenomenon of superlubricity,

or the achievement of extremely low friction levels, in Gr results from two distinct types of interactions, i.e., those where the surfaces are perfectly aligned (commensurate) and those where they are misaligned (incommensurate).¹⁵⁰ In situations of commensurate alignment, the stability of Gr nanoflakes on top of Gr sheets is maintained as atoms leap across energy barriers, dissipating energy during the friction process.¹⁵¹ Conversely, with incommensurate alignment, where the surfaces are not perfectly matched, energy dissipation is significantly reduced, thus facilitating superlubricity. This effect has been noted for Gr interfaces with materials such as gold¹¹⁵ and WS₂.¹¹⁹ Studies conducted by Dienwiebel et al.¹²¹ in 2004, along with research by Feng et al. in 2013,¹²² underscored the crucial role of the alignment type in determining friction levels. In particular, they observed that the shift from commensurate to incommensurate alignment, aided by thermal fluctuations, drastically lowers the friction levels. Such findings indicate that manipulating the alignment between Gr layers could be the key to effectively controlling friction, offering promising avenues for boosting lubrication efficiency.

Investigating the frictional properties of perfect Gr interfaces has proven difficult, prompting the adoption of novel techniques such as chemical vapor deposition (CVD) to deposit Gr layers onto microspheres and envelopment of the atomic force microscopy (AFM) probes with Gr.¹⁵² Liu et al.¹⁵³ studied the frictional dynamics of Gr layers and the interface between Gr and hBN by employing a SiO₂ microsphere coated with Gr. They successfully achieved consistent superlubricity, unaffected by the rotational alignment of the interfaces and variations in the pressure (up to 1 GPa) and humidity. This phenomenon is attributed to the incommensurate contact between surfaces randomly coated with Gr.

The superlubricity of Gr and related materials has been extensively studied through both experimental and computational methods including simulations and first-principles calculations. For example, Cahangirov et al.¹⁵⁴ demonstrated superlubricity in multilayer Gr (MLG) coatings on Ni(111) surfaces, attributing the effect to the reduction of adhesion and sliding friction by shielding the Ni surface attraction. This was further enhanced by the weakened coupling between Gr layers due to chemical interactions between Ni and Gr orbitals. Wang et al.¹⁵⁵ achieved superlubricity in van der Waals heterostructures of Gr and MoS₂, with density functional theory (DFT) calculations showing that superlubricity arose from decreased lateral force constants due to reduced potential energy corrugation during sliding. Similar DFT studies on

fluorographene/MoS₂ heterostructures found almost no energy loss in sliding due to smooth potential energy surfaces.¹⁵⁶

The formation of a transfer film is also crucial for achieving superlubricity in Gr materials as it converts the contact from Gr-to-material to Gr-to-Gr. This process facilitates incommensurate contact at the nanoscale, enhancing superlubricity and significantly improving tribological performance at the macroscale.^{157,158} Li et al.¹⁵⁹ demonstrated nanoscale superlubricity at the SiO₂/highly oriented pyrolytic graphite (HOPG) interface under ambient conditions, showing it could be controlled by adjusting the contact pressure. Below 2.52 GPa, superlubricity was maintained, but above this pressure, the COF increased 10-fold due to top layer delamination on HOPG. The mechanism was linked to the presence of a transfer film of Gr nanoflakes on silica, creating an incommensurate contact with HOPG.

Recent research has delved into how structural modifications in Gr affect its tribological properties. Lee et al.⁴⁰ discovered that the friction between an AFM tip and 2D nanomaterials varies with the atomic layer count of the nanomaterial, attributing the thickness-dependent frictional behavior to both the contact area and the atomic-scale contact quality, including commensurability and local pinning states.¹⁶⁰ Zhang et al.¹⁶¹ revealed that the friction of Gr can be reversibly adjusted by applying mechanical strain, with increased tensile strain leading to reduced COF, enabling superlubricity on strained Gr due to improved contact quality via in-plane strain. Furthermore, Wang et al.¹⁶² explored how strain engineering of Gr affects friction between Gr layers, demonstrating that robust superlubricity can be achieved through both uniaxial and biaxial stretching beyond a critical strain. This superlubricity is mainly independent of the relative orientations of the layers, thanks to the complete lattice mismatch evident in Moiré patterns, which significantly influences lubrication behavior.¹⁶³

The evolution of Gr materials during friction is crucial for achieving superlubricity.¹⁶⁴ Gr sheets wrap around nanoparticles to form nanoscrolls, reducing contact regions and creating incommensurate contact, which, alongside nanoball bearing effect of the nanoscrolls, significantly reduces friction.¹⁶⁵ Li et al.¹⁶⁶ observed superlubricity using graphite and fullerene-like carbon in an N₂ environment, with Gr nanoscrolls at the interface acting as microball bearings to lessen interlayer shear and lower the COF. Furthermore, applying TMD nanoflake coatings on amorphous carbon substrates and observing load-driven graphitization during wear leads to van der Waals heterostructures and macroscale superlubricity. Berman et al.¹⁶⁷ introduced macroscale superlubricity by merging Gr with NDs or metal nanoparticles, forming a friction pair with DLC films, as well as nanoscrolls during friction. Jiang et al.¹⁶⁸ achieved superlubricity on steel pairs using a Gr/MoWS₄ composite in a dry argon environment, where wear debris formed nanoscroll structures, transforming the interface into a tribolayer–nanoscroll–tribolayer system, facilitating incommensurate contact. Zhang et al.¹⁶⁹ obtained superlubricity with a Gr-coated microsphere lubricant, achieving a COF of 0.006 in air, which was attributed to the low-shear exfoliated Gr flakes, as well as the buffering and stress-distributing role of the movement of the microspheres.

■ ROLE OF GRAPHENE IN IMPROVING SYNTHETIC LIQUID LUBRICANTS: MECHANISMS AND PERFORMANCE GAINS

Gr excels in both solid and liquid superlubricities and is used as a tribopair nanomaterial or lubricating nanoadditive across scales. Overall, Gr-based materials enhance lubrication via tribochemical reactions, adsorption, and fluid effects, demonstrating diverse mechanisms for achieving superlubricity. Gr achieves superlubricity at the nano- and microscales through special liquid molecule contact, demonstrating significant reductions in friction.¹⁷⁰ Li et al.¹⁷¹ reached superlubricity with a Gr/hydrophobic fluoroalkyl interface lubricated by water, recording a COF of 0.0003 under pressures below 14.5 MPa. This effect is attributed to a nanometer-thick water layer remaining between the Gr and the surface due to low pressure and weak interaction, facilitating smooth sliding. Similar superlubricity was observed with zwitterions in a lipid bilayer sliding against Gr under water, indicating water intercalation as a method to reduce friction.¹⁷² Zhang et al.¹⁷³ further demonstrated superlubricity on HOPG surfaces using ILs, where cation–anion interactions under varying voltages were observed to enhance the boundary layer properties, contributing to friction reduction. These findings offer new perspectives on achieving superlubricity and reducing friction between Gr and various surfaces through molecular and ionic manipulation. In general, Gr-based materials enhance wear resistance and reduce friction by forming protective, adsorption, and tribochemical layers, which prevent direct contact.³² Although GO nanoadditives lower the COF down to 0.02,¹⁷⁴ typical COF values range from 0.02 to 0.1, which are above superlubricity levels.^{175,176}

Ge et al.¹⁷⁷ demonstrated superlubricity at macroscale interfaces, such as Si₃N₄/SiO₂, Si₃N₄/Si₃N₄, and Si₃N₄/sapphire, through the combined use of GO and ethylene glycol (EG). After a 600 s wear-in period, the COF was observed to drop to below 0.01, further reducing to 0.0037 and stabilizing for two hours. The wear volume with GO-enhanced EG was just 5% of that with EG alone. They attributed this performance to the formation of a GO adsorption layer that prevented severe wear as well as a hydrated network at the GO/EG interface that reduced shear strength and contributed to superlubricity. They concluded that neither the GO adsorption layer nor the tribochemical layer could independently achieve superlubricity without the presence of a fluid film that also helps form a hydrated network, highlighting the necessity of their combined effect for liquid superlubricity. Liu et al.¹⁷⁸ proposed a novel strategy employing hydrophobic Gr coatings and glycerol aqueous solutions to address the dispersity issue of Gr in lubricants, achieving macroscale superlubricity with a COF of 0.004 and reduced wear thanks to a critical in situ-formed Gr nanoflake tribolayer.

Gr-based nanomaterials show different properties based on their functional groups; however, the specific roles of these functional groups in enhancing lubrication are not fully understood. Ge et al.¹⁷⁹ discovered that the -NH₂ group in GO-based lubricants excels in achieving superlubricity over -OH and -COOH due to its stronger adhesive force with contact surfaces. The result is a more durable GO-NH₂ adsorption layer, as confirmed by the high-resolution transmission electron microscopy (HRTEM) analysis of worn surfaces. This layer protects the surfaces, reduces wear, and facilitates a shift from solid contact to weak interlayer shearing

with the GO-NH₂ layer, thereby achieving superlubricity. This research advances our knowledge of the influence of the functional groups of GO on lubrication and guides the design of Gr-based lubricants.

■ BIOLUBRICANTS

Biolubricants represent an emerging frontier in pursuing ecofriendly technologies, offering a promising alternative to traditional petroleum-based lubricants.¹⁸⁰ Constituting nontoxic and easily degradable oils, biolubricants are distinguished by their minimal environmental impact, aligning with the growing global emphasis on sustainability. These lubricants boast many advantageous properties, including a high viscosity index, excellent COF, low evaporation rate, high ignition temperature, and notably lower emission into the atmosphere.¹⁸¹ With these attributes, biolubricants are poised to play a crucial role in reducing ecological damage and promoting the health of various ecosystems.¹⁸² Petroleum-based lubricants are complex mixtures of hydrocarbons, including paraffinic, olefinic, naphthenic, and aromatic compounds, typically consisting of 20 to 50 carbon atoms.¹⁸³ While effective in their function, these lubricants have significant drawbacks due to their toxic and non-biodegradable nature, which contributes to pollution, and nonrenewability. The combustion of these oils releases harmful gases and particles, posing risks to both the environment and public health. Furthermore, the reliance on petroleum, a nonrenewable resource, underscores the urgency for alternatives that can meet the lubrication needs of the modern global industrial complex without depleting natural reserves. Biolubricants offer a solution to these issues by providing effective lubrication while being nontoxic and biodegradable. They do not harm any species or ecosystems, presenting a stark contrast to their petroleum counterparts. Using biolubricants can significantly decrease friction and wear in machinery, extending their lifespan.¹⁸⁰ Despite their promising attributes, biolubricants face several technical challenges that must be addressed to fully realize their potential. These include a limited operating temperature range, less oxidative stability causing accelerated degradation, a restricted viscosity range, and a low pour point temperature.^{26,181} These limitations can hinder the performance and applicability of biolubricants under various operational conditions.

To overcome these obstacles, recent advancements have focused on enhancing the properties of biolubricants through the integration of nanoparticles. Research has shown that even small quantities of nanoparticles can significantly improve the tribological properties of biolubricants.¹⁸⁴ Nanoparticles, with their ultrafine sizes ranging from 1 to 100 nm and large specific surface areas, exhibit excellent surface activity.¹⁸⁵ The addition of nanoparticles to biolubricants not only improves their stability and performance but also expands their application potential. For example, Gr-based nanomaterials have shown exceptional promise in tribology due to their ability to form various nanostructures like nanoballs, nanorods, and nanowires, thereby enhancing lubricant performance.¹⁴⁶ The use of such nanoparticles in biolubricants can lead to reduced friction and wear, lower energy consumption in machinery, and extend maintenance intervals.¹⁸⁶

Gr-Based Nanoadditives in Biolubricants. The energetic footprint of the lubrication is significant. A study in 2017 revealed that nearly a quarter (approximately 23%) of the global energy consumption could be attributed to tribological

contacts, underscoring the substantial role of friction and wear in our energy landscape.¹¹⁸ Moreover, the environmental repercussions of conventional lubrication practices, primarily reliant on crude-oil-derived base oils, present a daunting challenge. These substances pose detrimental effects on soil, groundwater, vegetation, animals, and human health.^{187–190} This perspective aligns with a growing consensus among researchers on the importance of developing ecofriendly and biodegradable lubricant substitutes,^{116,191} albeit recognizing their limitations, such as diminished performance at low temperatures.¹⁸¹ As a general practice, lubricants are enhanced by an array of additives, including friction modifiers and antiwear agents, aimed at improving their tribological properties. Integrating nanomaterials, such as metals, carbon-based nanostructures, and oxides, into lubricants has been documented in numerous reviews to confer significant tribological benefits.¹⁰ Gr and its derivatives distinguish themselves as a promising class of carbonaceous nanomaterials. The efficacy of GnPs as lubricant nanoadditives has been validated by various researchers,^{192–194} who have demonstrated notable enhancements in friction and wear performance with the judicious incorporation of GnPs (0.02–0.5 wt %) into lubricant oils.

It has been shown that the thickness and lateral size of carbon-based nanoadditives influence the antiwear and antifriction properties of biolubricants. For example, Liñeira del Río et al.¹⁹⁵ conducted a comprehensive study on the tribological behavior of a biodegradable polymeric ester lubricant (BIOE), enhanced with two distinct types of pristine GnPs, designated as GnP7 and GnP40. These GnPs are characterized by their lateral dimensions of 7 and 40 μm and thicknesses of 3 and 10 nm, respectively. The study utilized four different concentrations of nanoadditives: 0.015, 0.035, 0.055, and 0.075 wt %. The stability of these lubricants was assessed through visual observation and by monitoring the changes in the refractive index over time, which indicated marginally improved stability for lubricants containing GnP40, the GnPs with a larger lateral dimension. Moreover, compared to the base BIOE without nanoadditives, all lubricant formulations demonstrated lower COF values (Figure 2a) and reduced wear areas (Figure 2b,c). Further analysis confirmed that the enhanced tribological performance of the lubricants is attributed to the formation of a protective film and a surface repair mechanism.

Biopolymers offer remarkable versatility in their applications across different industries.^{196–199} Among biopolymers, cellulose nanocrystals (CNCs)²⁰⁰ present a multitude of benefits for tribological applications. The efficacy of CNCs as nanoadditives in lubricants has been explored with both mineral-based lubricants, such as SAE40 oil,²⁰¹ and biodegradable lubricants, such as polyol ester (POE) oil.^{202,203} It is worth mentioning that PAO based oils are widely used for their thermal stability and oxidation resistance, and they are synthetic hydrocarbons derived from petroleum and, in general, do not qualify as biodegradable lubricants.²⁰⁴ However, their biodegradability depends on their synthesis route and source precursors, such as sustainable feedstock (vegetable oils).

A prominent strategy for imparting biodegradability to lubricants involves incorporating nanoparticles derived from biological sources into such oils. Key among these nanomaterials are nanofibers, cellulose, and GnPs, which can be integrated into POE oil to fabricate lubricants tailored for

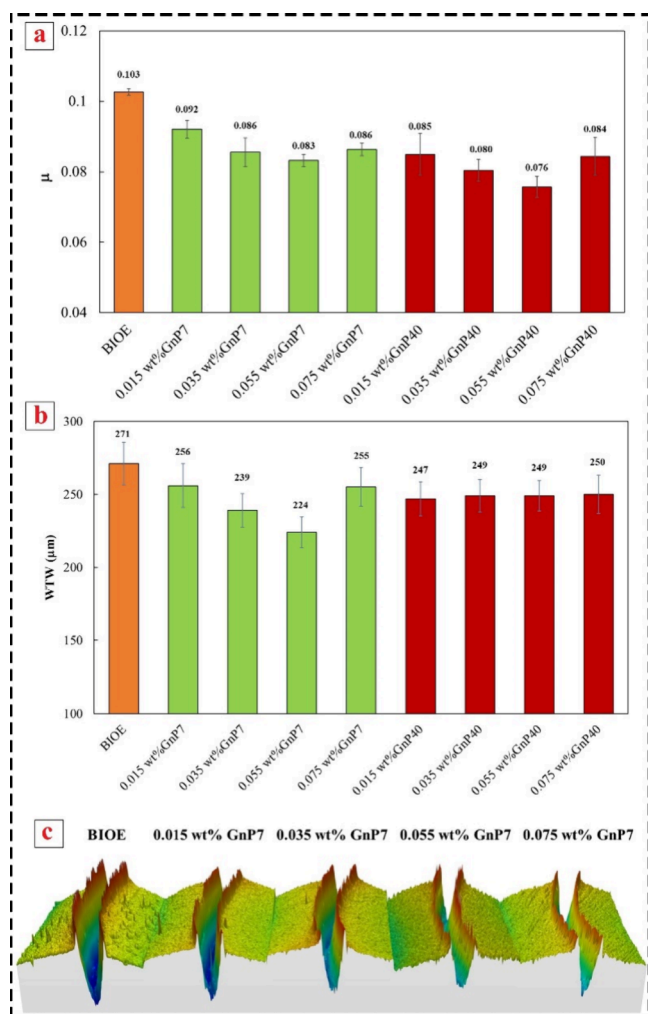


Figure 2. Overview of the BIOE-based lubricant test results. (a) Mean friction coefficients (μ) for the BIOE-based lubricants. (b) Width of the wear tracks on the disks lubricated with various BIOE-based lubricants. (c) 3D surface topography of the wear tracks on surfaces lubricated with BIOE/GnP7 lubricants with different GnP7 weight fractions (reprinted from ref 195. Copyright 2022 Elsevier).

tribological applications. Fuadi et al.²⁰⁵ indicated that the tribological efficacy of POE-based lubricants containing bacterial cellulose (BC) could be augmented by the inclusion of GnP. This augmentation was evidenced by a 20% reduction in the specific wear rate of the materials upon addition of GnP to the BC-enriched POE oil. The enhancements in friction and wear characteristics can be attributed to the formation of a tribofilm at the contact interfaces. Energy dispersive spectroscopy (EDS) analysis revealed a significant role of oxygen in the friction and wear of the lubricant based on the POE oil, enhanced with BC and GnP nanoadditives, with the tribofilm displaying considerable oxygen content. While the inclusion of GnP in the POE oil eliminated the oxygen-rich layer, it did not alone diminish the wear rate. Nonetheless, the concurrent integration of GnP and BC into the POE oil markedly improved its friction and wear performance, even though oxygen was still detectable at the contact interfaces.

Research has highlighted the potential of biobased oils as environmentally friendly alternatives to petroleum-based oils, owing to their biodegradability, reduced toxicity, and tribological superiority.²⁰⁶ Despite their advantages, bio-

lubricants face challenges such as oxidation at high temperatures and lower load-carrying capacities compared to conventional oils.^{207,208} Table 4 summarizes the key properties and challenges of different biolubricants along with the specific enhancements achieved through incorporating graphene-based nanoadditives. These improvements, particularly in tribological performance, illustrate the significant potential of graphene derivatives and hybrids to address the limitations of conventional biolubricants.

However, advancements in modifying their chemical structure, such as converting glycerol esters to sugar peresters, or incorporating polyol esters, have enhanced their thermal stability and oxidation resistance.²⁰⁷ Additionally, it has been suggested that these deficiencies can be overcome by dispersing various additives, such as nanoparticles, tribological enhancers, and friction modifiers.^{141,166} In this context, Banavathu et al.²⁰⁸ investigated the physicochemical and commercial characteristics of biolubricant and commercial lubricant blends, specifically focusing on the incorporation of GnP into the base oil. Pentaerythritol (PE) ester, derived from *Calophyllum inophyllum* (Tamanu tree) seed oil, was blended with commercial SN motor oil in volumetric concentrations of 10, 20, 30, and 40%. The initial phase of their study identified the optimal ratio of PE ester to commercial SN motor oil for an enhanced lubrication performance. Subsequently, the optimal blend of biolubricant and commercial oil was enriched with GnP in weight fractions ranging from 0.025% to 1%. A formulation consisting of 30 wt % biolubricant in commercial oil with 0.05% GnP significantly improved friction and wear characteristics of the base oil. In another study, Sadiq et al.¹⁴³ investigated the oxidation stability and tribological performance of biolubricants enhanced with nanoparticles, specifically Gr and maghemite ($\gamma\text{Fe}_2\text{O}_3$) dispersed in coconut oil at a volume concentration of 0.1%. Their findings indicated that the incorporation of nanoadditives into the biolubricants significantly enhanced their oxidation stability by delaying thermal degradation. Specifically, the oxidation onset temperature of biolubricants containing maghemite and GnP increased by 18.75 and 37.5 °C, respectively. These improvements positively affected both the viscosity and the tribological performance of the biolubricants. Table 5 summarizes the key properties and challenges associated with different biolubricant types and highlights the specific enhancements achieved through the incorporation of graphene-based additives, as discussed in recent studies.

For the Gr-enhanced lubricant, there was a reduction of 10.4% in the COF and 5.6% in the wear scar diameter (WSD). In comparison, the maghemite-enhanced lubricant saw reductions of 3.43% in COF and 4.3% in WSD compared with those of the pure coconut oil. Additionally, the viscosity index of the lubricants increased by 7.36 and 13.85% for maghemite and Gr lubricants, respectively. Continuing this exploration of biolubricants, Hettiarachchi et al.¹²⁰ investigated the tribological enhancements achieved through the formulation of nanobio lubricants employing 2D nanocomposites of Gr/ Al_2O_3 as innovative nanoadditives in coconut oil. This lubricant demonstrated a performance comparable to that of traditional mineral-based engine oil 15W40. The optimization of this 2D nanocomposite within the coconut oil base led to a 28% decrease in the COF and an 8% reduction in the specific fuel consumption (Figure 3a), as well as a noticeable decrease in the exhaust emissions (CO , SO_2 , and NO_x) (Figure 3b).

Table 4. Overview of Biolubricants

type of biolubricant	key properties	challenges	graphene-based enhancements	tribological performance improvements	references
Plant Oils	High biodegradability, renewability, low toxicity	Poor oxidative stability, low thermal stability	Graphene (Gr), graphene oxide (GO), reduced graphene oxide (r-GO)	Enhanced antiwear properties, improved thermal stability, formation of protective films	132, 184, 209
Animal Fats	High viscosity index, effective in lubrication	Limited temperature range, lower pour point	Gr and its derivatives	Enhanced friction reduction, improved load-carrying capacity	180, 181, 210
Biodegradable Esters	Low toxicity, ecofriendly	Limited viscosity range, low oxidative stability	Gr-based hybrids with metals and polymers	Improved wear resistance, formation of tribological films, increased thermal conductivity	181, 184, 211
Triglyceride-Based Oils	High flash point, good lubricating properties	Poor cold temperature performance	Gr and GO additives	Better dispersibility, improved film formation, reduced friction coefficient	116, 118, 191

Bhaumik et al.²¹³ formulated a biodegradable lubricant by incorporating different volume percentages of cashew nut shell liquid (CNSL) into the neat castor oil (NCO) and explored its potential as a substitute for non-biodegradable mineral oils. The formulation demonstrating the most promising tribological properties was further enhanced with different weight percentages of r-GO, after which additional tribological evaluations were conducted. A lubricant blend consisting of 40% CNSL and NCO outperformed a commercial mineral oil by 45.8%. The inclusion of 0.5% r-GO into this blend further enhanced its performance, achieving a 61.7% improvement over that of commercial mineral oil. When augmented with nanoparticles and tested in a gearbox, this innovative biodegradable mixture underscored the superior performance of the biodegradable lubricant, showcasing its viability for industrial use. In another study, Mushtaq et al.²¹⁴ aimed to enhance the lubrication capability of Karanja oil (KO) by incorporating nanosized MoS₂ and Gr as nanoadditives into this oil. Based on their findings, the addition of these nanoparticles significantly reduced the wear on the test balls, with the minimum WSD observed at 0.5 wt % nanoadditive content, corresponding to a 27.33% reduction in wear compared to the untreated KO. Moreover, it was found that both the COF and wear rate decreased as a function of load and speed, a phenomenon attributed to the formation of a protective tribo-layer on the wear surfaces.

Continuing this trend of incorporating Gr-based nanoparticles to improve the lubrication efficiency of biolubricants, Jie Jason et al.²¹⁵ conducted a comprehensive study on the tribological behavior of Pongamia oil (PO) and 15W40 mineral engine oil (MO), both with and without the addition of GnPs. The introduction of 0.05 wt % GnPs into both PO and MO resulted in the most significant reductions in friction and wear, corresponding to 17.5% and 12.24% reduction in friction (Figure 3c), and 11.96% and 5.14% reduction in wear (Figure 3d) for PO and MO, respectively. Moreover, the study indicated a surface improvement on worn surfaces, which was attributed to the polishing effect of GnPs. It was determined that the deposition of GnPs onto the friction surfaces, coupled with the formation of a protective film, effectively prevented direct surface contact, thereby significantly reducing friction and wear. In another work, Suresha et al.²¹⁶ studied the impact of GnPs on the physico-tribological properties of Madhuca indica (Mahua) oil. The incorporation of 0.25 wt % GnPs into the oil led to notable reductions of 17% in the COF and 15% in the WSD. A wear preventive characteristics analysis revealed the formation of micro tracks on the steel balls, indicating the presence of wear. An extreme pressure characteristics evaluation further highlighted that Mahua oil blended with 1 wt % GnPs exhibit superior antiwear performance, thereby identifying this concentration as the optimal GnP loading from a weld load perspective. Finally, a GnP content of 0.1 wt % was determined to be the most effective from an antiwear perspective.

Recent studies have explored how Gr-based nanoadditives affect the rheological (viscosity), wear resistance, and overall performance of biolubricants under various operational conditions. Anand et al.¹³² investigated the rheological and tribological properties of biolubricants derived from date (Balanites Aegyptiaca) oil, which were formulated with GO and stabilized with sodium dodecyl sulfate (SDS) surfactant to maximize nanoparticle dispersion within the oil. The study involved preparing samples with GO concentrations of 0.25%,

Table 5. Summary of Biolubricant Types, Challenges, and Enhancements Achieved through Graphene-Based Nanoadditives

year	graphene derivative/hybrid	base oil	friction reduction (%)	wear reduction (%)	other benefits	reference
2023	Graphene oxide (GO)	Trimethylolpropane ester	25%	30%	Ecofriendly, enhanced stability	132
2023	Al ₂ O ₃ /Graphene hybrid	Polyalphaolefin (PAO)	35%	40%	Improved load-bearing capacity	184
2024	Reduced graphene oxide (r-GO)	Castor oil	20%	28%	Enhanced thermal stability, reduced viscosity	120
2024	Graphene hybrid with MoS ₂	Ester-based oil	40%	35%	Improved load-bearing capacity	212

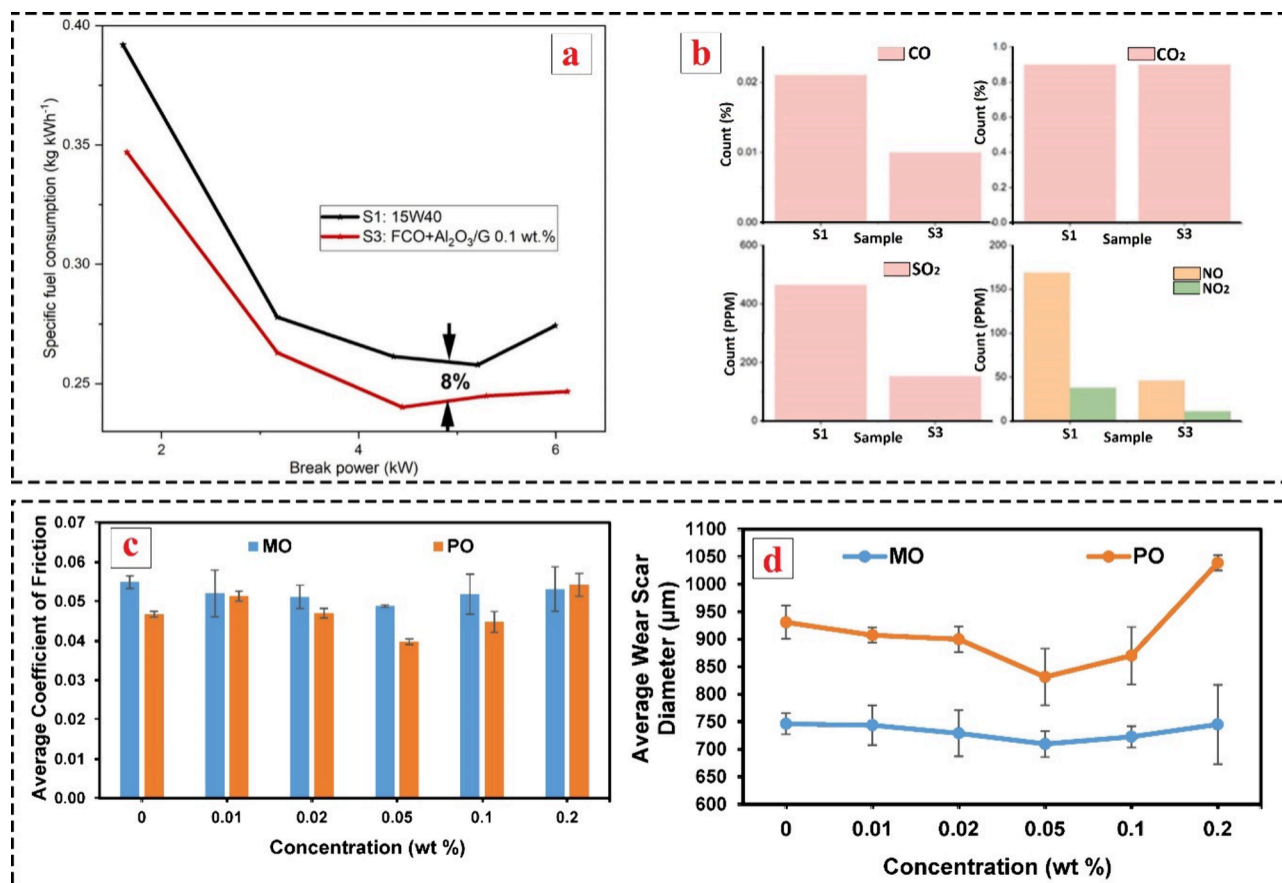


Figure 3. Comparative analyses of selected lubricant samples. (a) Relationship between the brake power and specific fuel consumption for samples S1 (100 wt % 15W40 oil) and S3 (formulated blend of 98 wt % coconut oil, 1.9 wt % oleic acid, and 0.1 wt % Al₂O₃-G). (b) Exhaust gas analysis results for blends S1 and S3 (reprinted with permission from ref 120. Copyright 2023 Elsevier). (c) Average coefficient of friction outcomes for the Pongamia oil (PO) and mineral engine oil (MO) with varying GnP concentrations (0–0.2 wt %). (d) Wear scar diameter (WSD) results for the PO and MO oils across the same range of GnP concentrations (reprinted from ref 215. Copyright 2021 MDPI).

0.5%, 0.75%, and 1% by weight in date oil and examining their rheological properties, such as viscosity and shear stress. Additionally, tribological assessments, including analyses of friction, wear, and WSD, were conducted using a pin-on-disc apparatus. These improvements positively affected both the viscosity and tribological performance of the biolubricants. Among the tested concentrations, the blend containing 0.75% GO demonstrated the most favorable properties for engine oil replacement. This optimal blend resulted in reduced viscosity, torque, friction, and wear. Furthermore, the WSD imagery indicated that the 0.75% blend is the most suitable for commercial applications, underscoring the potential of GO-enhanced date oil biolubricants as effective substitutes for traditional engine oils. In another work, Padgurskas et al.¹⁰⁸ explored the feasibility of producing biogrease from rapeseed oil, using beeswax as a thickener functional antiwear agent and antioxidant, along with carbon nanoparticles derived from GO

and thermally expanded graphite. Results indicated that the greases exhibited non-Newtonian behavior and that the rheological parameters varied with temperature. Although an increased concentration of beeswax negatively impacted the tribological properties of the grease (Figure 4a–g), the inclusion of functional additives and nanoparticles markedly enhanced the thermal properties, yield stress, viscosity, and tribological performance of the grease, notably decreasing its wear rate (Figure 4h,i). Similarly, Alqahtani et al.¹⁰⁹ explored the enhancement of SAE 5W30 oil with GnPs to investigate its tribological and rheological properties. The study assessed how varying concentrations of GnPs (0.03, 0.06, 0.09, 0.12, and 0.15 wt %) influenced the properties of the base oil. Based on their findings, the inclusion of nanoadditives resulted in 15% and 33% improvements in the WSD and COF, respectively. Compared to the base oil, enhancements were observed in flashpoint, thermal conductivity, kinematic viscosity, and pour

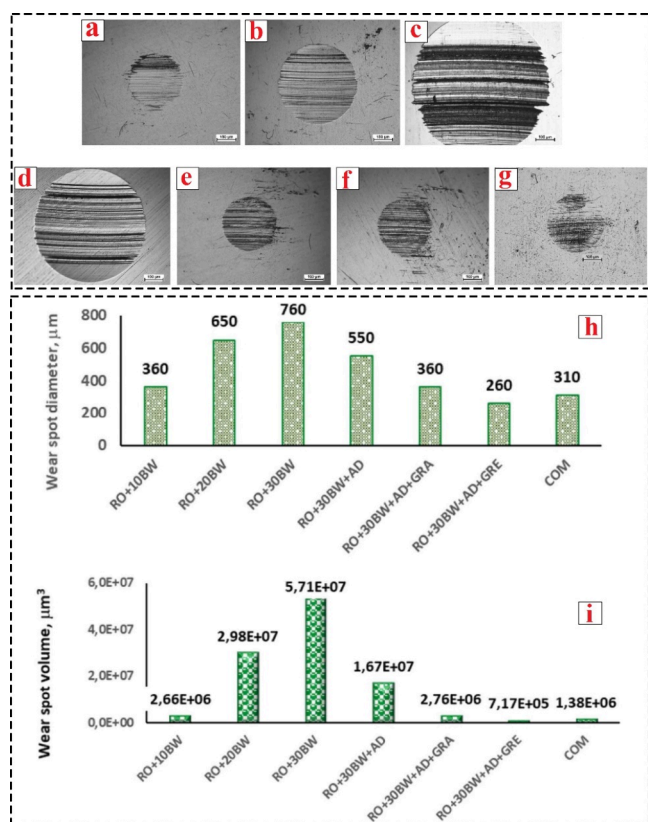


Figure 4. Surface imagery of wear scars on balls subjected to a 150 N load. (a) Rapeseed oil (RO) + 10% beeswax, (b) RO + 20% beeswax, (c) RO + 30% beeswax, (d) RO + 30% beeswax + antioxidants/antiwear nanoadditives, (e) RO + 30% beeswax + antioxidant/antiwear nanoadditives + GO, (f) RO + 30% beeswax + antioxidant/antiwear nanoadditives + graphite, (g) COM (reference commercial biological grease). Analysis of wear on the balls at 150 N load: (h) wear spot diameter, (i) wear volume (reprinted with permission from ref 108. Copyright 2023 Elsevier).

point, showing increases of 25.4%, 77.4%, 29.9%, and 35.4%, respectively. The study concluded that the incorporation of GnPs significantly enhances the performance characteristics of 5W30 engine oil.

The diminishing reserves of crude oil and global initiatives aimed at protecting the environment from pollution have spurred renewed interest in developing ecofriendly lubricants from vegetable oils.²¹⁷ Bhat et al.¹¹⁰ delved into the impact of varying concentrations of lithium stearate as a thickener and GnPs on the rheological behavior of coconut oil-based grease. Lithium stearate was incorporated at concentrations of 6, 12, and 18 wt %, while GnPs were introduced at 0.5, 1, and 2 wt %. The findings indicate an enhancement in the rheological properties of the coconut grease with higher concentrations of lithium stearate and nanoparticles. Specifically, at a GnP concentration of 2 wt %, the viscosity and shear stress of the grease were augmented by 25% and 33%, respectively. Additionally, the influence of GnP concentration on the dropping point of the grease was examined, revealing a 13.19% increase at a 2 wt % GnP concentration. The study concludes that coconut oil-based grease, with an inclusion of 2 wt % GnPs, represents a viable lubricant alternative for industrial applications with environmental concerns.

Nonedible oils are increasingly recognized as a sustainable source for biolubricant production, with castor oil being

particularly notable for its distinct hydroxyl group configuration, which enables its use in lubrication, dielectrics, and heat transfer applications.¹¹³ Vora et al.¹⁴⁷ conducted a study on the dynamic viscosity and thermal conductivity of castor oil and GnP/castor oil fluids synthesized via a two-step method, involving the mixing of GnPs with pure castor oil. Their findings indicated a notable reduction in the dynamic viscosity of the fluid with increasing temperature, whereas higher GnP contents showed a modest increase in the dynamic viscosity. Interestingly, GnPs maintained the Newtonian flow behavior of the castor oil. In addition, a significant enhancement in the thermal conductivity of the fluid was observed with the introduction of GnPs. These findings position the biodegradable GnP/castor oil fluid as a promising biolubricant with enhanced heat transfer capabilities and potential applications in thermal management.

The natural wax derived from the nonmedicinal part of *Codonopsis pilosula* offers a novel, sustainable resource for the production of biolubricants.¹⁴⁸ To enhance the lubricity and load-carrying capabilities of this wax (referred to as the P grease), multilayer Gr (MGr), which was synthesized through a magnesium metallothermic reaction, was incorporated into the P grease.¹⁴² Compared to the pure P grease, the P+MGr grease demonstrated a significantly enhanced load-carrying capacity of 300 N and maintained effective lubrication across a broad temperature range from room to 250 °C (Figure 5a-d). At room temperature, the interaction between the semisolid grease and MGr culminated in the formation of a superior tribofilm. This film significantly improved both the lubricity and the load-bearing capacity of the P+MGr grease relative to the pure P grease (Figure 5e). Moreover, beyond a temperature threshold of 150 °C, the MGr within the P+MGr grease was found to play a crucial role in diminishing the COF, wear, and oxidation experienced by steel pairs (Figure 5f).

Suresha et al.²¹⁸ conducted an investigation into the friction and wear characteristics of Mahua oil, enhanced with 0.25–1% by weight of GnPs. The incorporation of GnPs into the Mahua oil significantly improved its lubrication performance, evidenced by a reduction in both the COF and wear loss, as indicated by the diminished WSD on the steel balls. Notably, even a minimal addition of GnPs (0.25 wt %) to Mahua oil demonstrated the potential to serve as an effective biolubricant, offering enhanced mechanical and thermal efficiency suitable for automotive engine applications. In another study, Suresha et al.¹⁹⁴ investigated the tribological performance of GnPs as an antiweld nanoadditive in neem oil under extreme pressure. The study focused on the impact of blending neem oil with various concentrations of GnPs on the friction and wear characteristics of the base oil. Based on their findings, neem oil containing 1 wt % GnPs exhibited the lowest COF and smoothest WSD. The results of the extreme pressure test showed a significant enhancement in the seizure load capacity of neem oil, corresponding to an increase of 27.8% at a GnP concentration of 0.5 wt %, compared to pure neem oil. Zhang et al.¹⁷⁶ explored the tribological performances of vegetable oils enhanced by the addition of GO, designated as GO-D and GO-T, that were developed through a chemical reaction between the carboxyl groups of GO and 1-dodecanethiol and *tert*-dodecyl mercaptan, respectively (Figure 6a). Their findings indicated that GO-D exhibited superior dispersion stability and tribological performance when added to rapeseed oil (RO) compared to GO-T, with both additives outperforming the

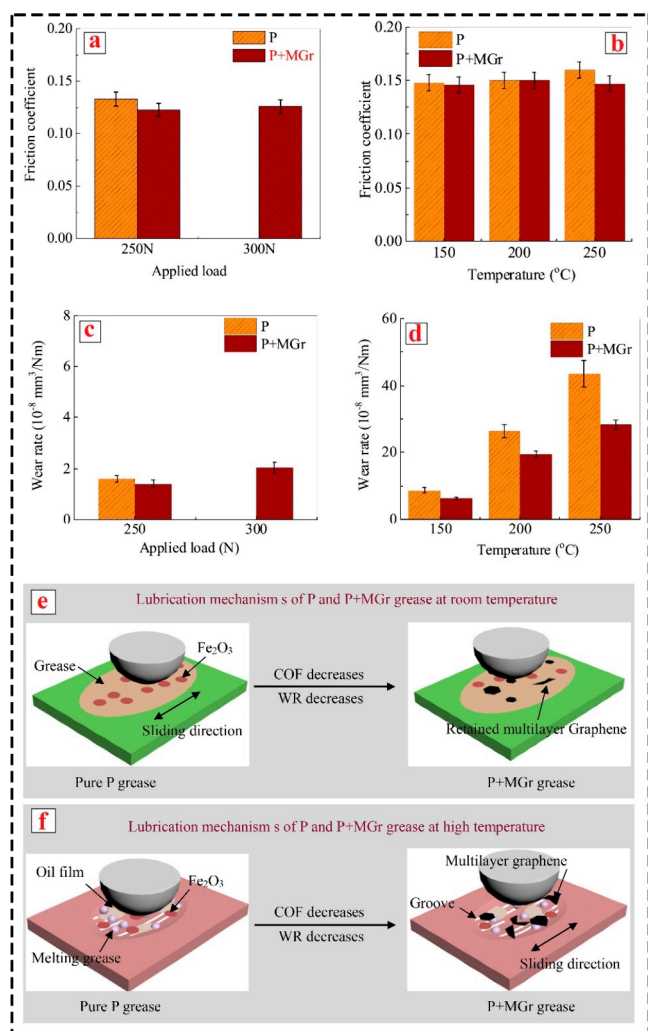


Figure 5. Friction and wear analysis of the AISI52100-AISI52100 steel pairs. (a) Friction coefficients for pairs lubricated by Codonopsis pilosula/multilayer Gr (P+MGr) and P greases under loads of 250 and 300 N. (b) Friction coefficients at varying temperatures for the same sliding pairs lubricated with P+MGr and P greases. (c) Wear rates for the AISI 52100 steel lubricated with P and P+MGr greases at a constant temperature of 25 °C under different loads. (d) Wear rates at a fixed load of 100 N across various temperatures. (e) Schematic representation of the lubrication mechanisms of the P grease at room temperature and (f) elevated temperature (reprinted with permission from ref 142. Copyright 2020 Elsevier).

base RO/GO (Figure 6b). Further analysis revealed that GO-D and GO-T nanoadditives could form absorptive and tribochemical films on the contact surfaces (Figure 6c-f), suggesting their effectiveness in improving lubrication.

The drilling industry relies heavily on drilling fluid for various tasks such as cooling the drill bits, removing debris, balancing formation pressure, stabilizing the wellbore, transmitting hydraulic pressure, and protecting oil and gas reservoirs. Integrating lubricants into drilling fluids offers numerous benefits, such as protecting equipment, improving drilling efficiency, maintaining wellbore integrity, and enhancing safety, all vital for successful drilling operations.¹⁴⁴ Geng et al.¹⁴⁵ introduced a novel biolubricant formulated from triolein. After synthesis of Gr and triolein, they integrated these into a drilling fluid system. In their study, they evaluated the efficacy of the biolubricant through tests measuring the COF, adhesion

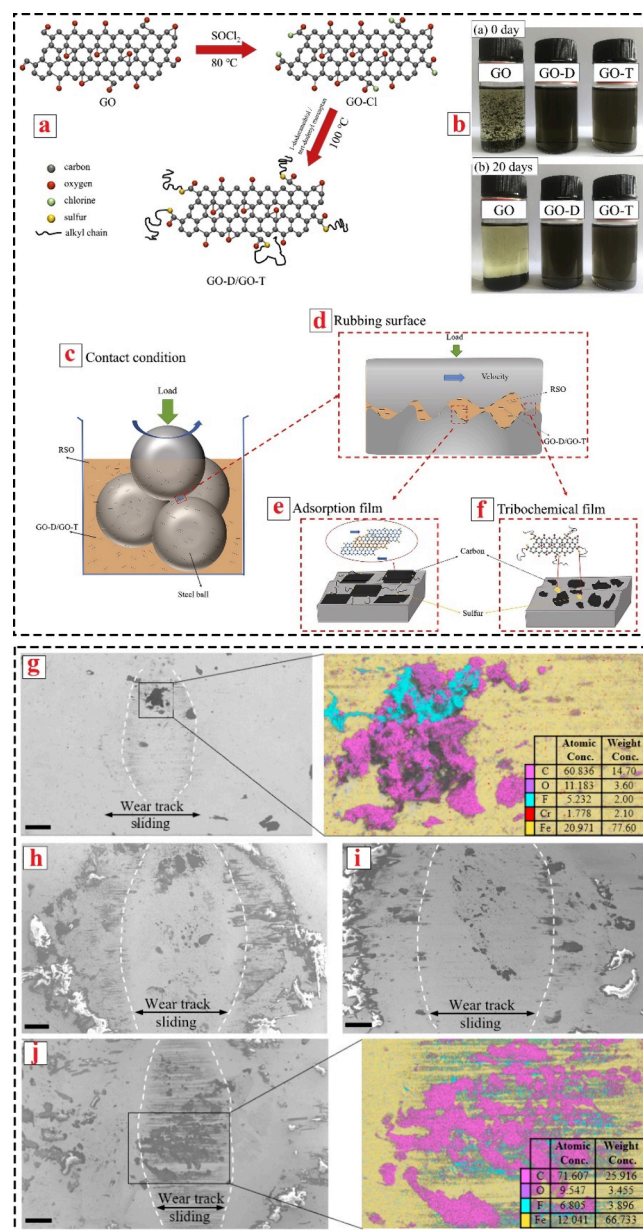


Figure 6. Overview of the GO-based dispersions and tribological testing. (a) Schematic illustration detailing the synthesis processes for GO-D and GO-T. (b) Digital images showing the dispersion of GO, GO-D, and GO-T (0.05 wt %) immediately and 20 days postsonication. Diagrammatic representation of friction processes involving steel balls in GO-D and GO-T dispersions: (c) friction process, (d) rubbing surface of steel balls, (e) adsorption film formation by GO-D and GO-T sheets, and (f) tribochemical film formation (reprinted with permission from ref 176; Copyright 2018 Elsevier). SEM images depicting surface conditions of steel balls after tribology tests for (g) K1 (PTFE:10g/PPTA:0.35g/GO:0.5g), (h) K2 (PTFE:10g/PPTA:0.7 g/GO:0.5 g), (i) K3 (PTFE:10g/PPTA:1.4 g/GO:0.5 g), and (j) K4 (PTFE:10g/PPTA:2.8 g/GO:0.5g) at a scale of 50 μm (reprinted with permission from ref 221. Copyright 2023 Elsevier).

coefficient, and extreme pressure lubricity. The addition of the biolubricants at 240 °C markedly reduced the adhesion coefficient of the drilling fluid to below 0.2 and down to 0.055, which signifies a reduction rate of over 70%.

In recent years, the development of self-lubricating polymers, e.g., PTFE, has emerged as a significant area of

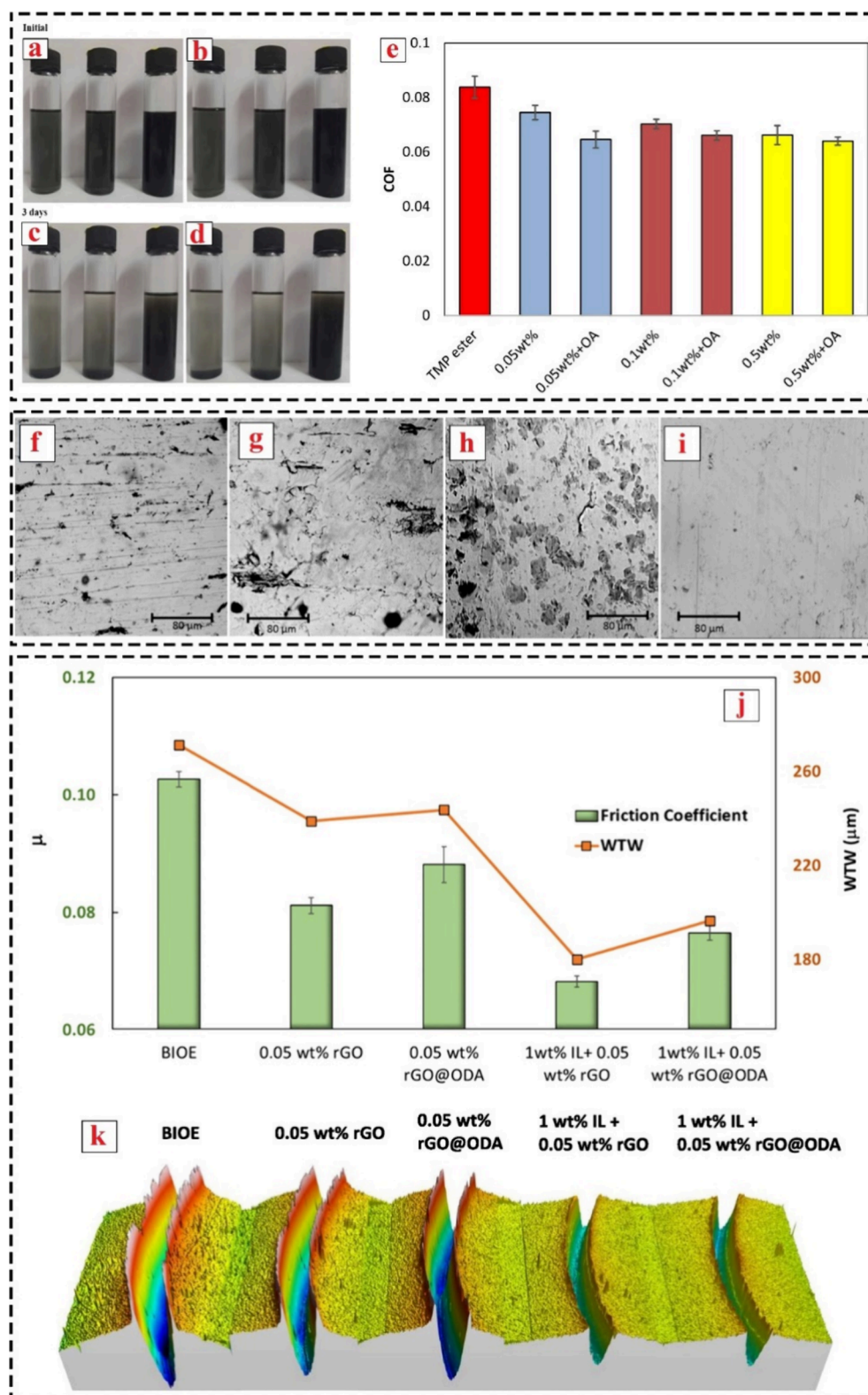


Figure 7. Analysis of the sedimentation, friction, and wear in lubricant testing. (a–b) Sedimentation test results for trimethylolpropane (TMP) ester/GO/oleic acid (OA) dispersions immediately following the mixing process. (c–d) Results after a period of 3 days, with the left side of each image showing samples without and the right side with OA. In each image, glass vials are arranged from left to right displaying TMP ester samples with increasing GO concentrations of 0.05, 0.1, and 0.5 wt %. (e) Average COF for all lubricants tested (reprinted with permission from ref 212. Copyright 2023 Elsevier). (f–i) SEM images of the wear on cast iron plates lubricated with TMP, TMP+graphene (G), TMP+ionic liquid (IL), and TMP+G+IL, respectively. Reprinted with permission from ref 224. (j) Comparative analysis of friction coefficients and wear track widths (WTW) for BIOE and BIOE lubricants. (k) 3D surface topography showing cross-sectional profiles of worn scars for all tested BIOE lubricants (reprinted from ref 225. Copyright 2021 Elsevier).

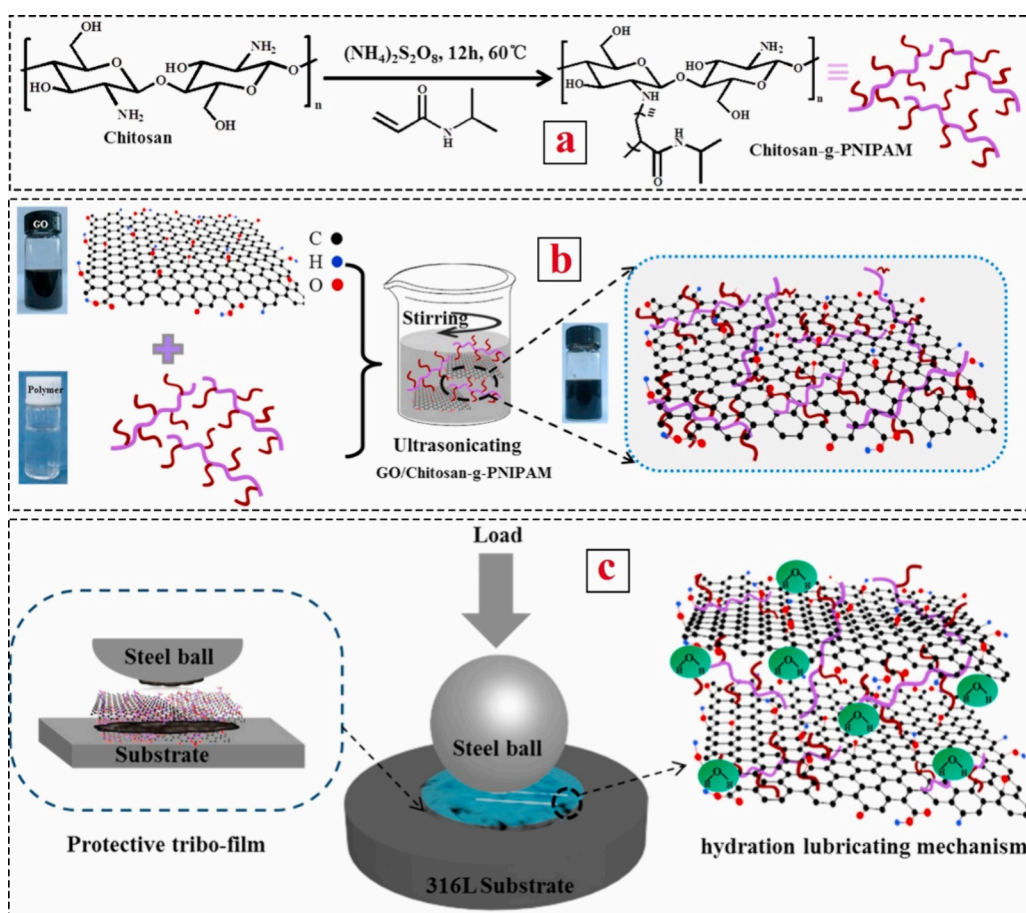


Figure 8. (a) Schematic representation of the synthesis process for the chitosan-g-PNIPAM copolymer. (b) Diagram illustrating the fabrication steps for the GO/chitosan-g-PNIPAM nanohybrids. (c) Conceptual schematic of the friction mechanism proposed for the GO/chitosan-g-PNIPAM nanohybrids when used as an additive in water-based lubrication systems (reprinted with permission from ref 71. Copyright 2021 Elsevier).

research within the field of advanced solid lubrication materials.²¹⁹ These materials are highly valued for their low COF, robust mechanical strength, corrosion resistance, and capacity for oil-free lubrication.²²⁰ Wang et al.²²¹ conducted a study aimed at enhancing the tribological performance of self-lubricating polymers by improving the properties of their transfer films. GO/PTFE/poly-*p*-phenylene terephthalamide (PPTA) composites (GPPs) were synthesized using a “dual-size” approach. The tribological behavior of the GPPs was evaluated using a ball-on-disk tribometer, demonstrating that GPPs exhibit a low COF (a minimum of 0.061) and significant hydrophobicity (a maximum of 137°). The hierarchical structure, composed of dual-sized PTFE and PPTA, was shown to mitigate the agglomeration of PTFE particles effectively. This configuration was found to hinder the mobility and extensive failure of the PTFE matrix, thereby diminishing the wear rate (Figure 6g-j). Additionally, it was determined that the presence of edges and defects in GO, which introduces chemically active sites, lowers the energy threshold required for polymer defluorination. Such alterations facilitate tribological chemical reactions, essential for the fortification of the transfer film and the enhancement of its adhesive properties.

While Gr demonstrates exceptional characteristics as a lubricant nanoadditive, its incorporation into lubricant oils is challenged by agglomeration. This issue arises due to cohesive interactions among nanoparticles, leading to the settling of Gr,

which in turn degrades the tribological performance of Gr-based lubricants.²²² The dispersion stability of Gr additives in nonpolar organic solvents is notably poor, resulting in diminished lubrication efficacy.¹¹² Therefore, overcoming agglomeration and sedimentation is a primary concern when Gr is employed in lubricant applications. One strategy to mitigate agglomeration involves using surfactants, achieved through physical modification to alter the interfacial properties of Gr.^{212,223} Strong physical interaction forces, such as hydrogen bonding and van der Waals forces, facilitate the binding of surfactant molecules to Gr, thereby reducing its surface tension and preventing agglomeration.¹¹² In a study conducted by Zulhanafi et al.,²¹² TMP ester served as the base lubricant, GO as the nanoadditive, and oleic acid (OA) as the surfactant. This combination, TMP/GO/OA, exhibited enhanced dispersion stability compared to that of TMP/GO alone (Figure 7a-d). Moreover, the incorporation of OA significantly reduced the coefficient of friction in samples with 0.05, 0.1, and 0.5 wt % GO by 22.9%, 21.1%, and 23.6%, respectively (Figure 7e).

The efficacy of biolubricants can be enhanced with the addition of Gr, though challenges in maintaining a stable suspension over extended periods have been noted.¹⁷⁶ Hasnul et al.²²⁴ explored a novel approach to combine Gr with an IL (triethyltetradecylphosphonium bis(2,4,4-trimethylpentyl)-phosphinate) to achieve stable dispersions of biolubricants.

The incorporation of IL, which was considered a surfactant in this study, not only stabilized the dispersion but also reduced the frictional performance by 33%. A comparative analysis revealed that using the IL or Gr independently resulted in 13% and 7% COF reductions, respectively. Examination of wear scars revealed that samples treated with the Gr/IL combination displayed smoother surfaces than those with corrosive spots or abrasive scars found in samples treated with only IL or Gr (Figure 7f-i). This outcome suggests a synergistic interaction between the two additives. Furthermore, the study determined that the IL contributed beyond dispersion stabilization, playing a critical role in the formation of the tribolayer, underscoring its multifaceted benefits in biolubricant performance enhancement. In a similar study, Liñeira del Río et al.²²⁵ explored the antifriction and antiwear synergies offered by two functionalized GO (r-GO and r-GO modified with octadecylamine, r-GO@ODA) when combined with a phosphonium IL as additives in a biodegradable ester base oil (BIOE). Four BIOE nanodispersions were prepared: two without the IL and two with the IL (forming hybrid lubricants). These formulations exhibited good temporal stability, particularly those containing the IL, with stability extending to at least 3 weeks. Moreover, compared to the neat BIOE oil, all hybrid and nonhybrid lubricants demonstrated reduced COF, with the most significant reduction in friction, 34%, observed for the lubricant containing 1 wt % IL+0.05 wt % rGO (Figure 7j). Similarly, discs lubricated with the prepared lubricants exhibited less wear than those lubricated with the BIOE oil alone, with the greatest wear reduction, 34%, also recorded for the 1 wt % IL+0.05 wt % r-GO lubricant (Figure 7j). Further analysis indicated that mechanisms of surface repair, synergistic effects, and tribofilm formation were present, contributing to the enhanced performance of the lubricants. 3D mapping of the wear scars on lubricated discs further showed that the wear resulting from the lubricants was less than that resulting from the BIOE base oil (Figure 7k).

Although adding small amounts of GO to water can decrease friction and enhance wear resistance,⁵⁴ as GO sheets form protective coatings on sliding surfaces,¹²⁴ its aggregation in water due to van der Waals forces and π - π interactions is a challenge. This challenge is often addressed by the chemical modification of GO with organic molecules or hydrophilic polymers to prevent clumping and enhance tribological characteristics.²²⁶ For example, attaching poly(ethylene glycol) (PEG) to carboxylated Gr nanosheets has been shown to result in improved friction-reducing and antiwear properties of the virgin Gr.²²⁷ However, the covalent modification of GO is complex and requires harsh conditions, potentially damaging its structure. In this respect, noncovalent modification offers a simpler alternative, which maintains the structure of GO, while enhancing its lubricating properties.²²⁸ Additionally, water-soluble polymers with brush-like structures from biological sources can improve water lubrication, but their high cost and limited availability make synthetic alternatives more viable for industrial use.²²⁹ Semisynthetic polymers, such as chitosan-derived copolymers, are promising due to their ecofriendliness, ease of modification, and low cost, despite the solubility challenges of chitosan, which recent strategies have begun to overcome.^{230,231} Wei et al.⁷¹ introduced a hybrid additive tailored for water-based lubrication, which synergistically combines GO with a brush-like, chitosan-based copolymer, specifically chitosan-graft-poly(*N*-isopropylacrylamide) (chitosan-g-PNIPAM) (Figure 8a,b). This GO/chitosan-g-PNIPAM

nanohybrid was synthesized through an efficient in situ, noncovalent assembly technique. The findings revealed that the nanohybrids exhibited exceptional friction-reduction and antiwear capabilities, especially under conditions of a high contact load. Notably, these nanohybrids were able to lower the mean COF by 40% relative to pure GO, and by 84% compared to the copolymer alone. Additionally, wear rates decreased by 15% and 47%, respectively. It was suggested that GO/chitosan-g-PNIPAM could adhere to friction-pair surfaces and form a hybrid protective film. This film prevents direct contact between the rubbing surfaces, thus providing effective lubrication and antiwear properties (Figure 8c). Furthermore, the presence of the chitosan-g-PNIPAM copolymer on the surface of GO is likely to enhance the adsorption of GO onto metal substrates and sustain the integrity of the boundary-lubricating film. This interaction facilitates a synergistic effect with GO, thereby enhancing the performance in reducing friction and wear.

■ FUTURE RESEARCH DIRECTIONS AND RECOMMENDATIONS

The potential integration of Gr-based nanoadditives into biolubricants heralds a transformative leap forward in lubrication technology poised to deliver both enhanced performance and significant environmental benefits. As we gaze into the future of this promising field, a critical analysis reveals several research avenues and strategies that must be prioritized to realize the full potential of these advanced materials. Although this study highlights the tribological improvements achieved by incorporating graphene-based additives into biolubricants, further research is needed to conduct a direct, systematic comparison with traditional mineral-based lubricants under identical conditions to fully establish their relative performance advantages. Such comparative studies will provide valuable insights into the practical benefits and potential limitations of graphene-enhanced biolubricants, guiding future developments in lubrication technology.

Enhanced Dispersion Techniques. One of the foundational challenges in the utilization of Gr-based nanoparticles in biolubricants lies in their dispersion stability. Future research must delve into the development of novel surfactants or dispersing agents. These agents should not only be effective at lower concentrations but also be environmentally benign, addressing both efficacy and ecofriendliness. Moreover, the exploration of mechanical methods, such as ultrasonic and mechanical mixing, promises to refine the uniformity of the dispersion without compromising the structural integrity of Gr. The critical question remains: can we achieve a balance between optimal dispersion and maintaining the intrinsic properties of Gr?

Advanced Gr Modifications. The customization of the properties of Gr through chemical and physical modifications represents fertile ground for innovation. Tailoring Gr for specific lubrication scenarios, such as high-temperature or high-pressure environments, could dramatically expand its applicability. Furthermore, the functionalization of Gr with both organic and inorganic molecules may offer new dimensions for modifying the tribological properties of biolubricants. The challenge here is to systematically ascertain the effects of these modifications and ensure that they are beneficial rather than detrimental to the desired lubrication characteristics.

Hybrid Nanocomposite Systems. The exploration of hybrid nanocomposite systems, wherein Gr-based nanoparticles are combined with other nanomaterials such as TiO₂, MoS₂, or nanodiamond, could unlock new synergies in lubrication technology. Research should focus on understanding the tribological behavior of these hybrid structures within diverse matrix materials like polymers and ceramics, targeting specialized applications. The critical consideration will be how these combinations affect the overall performance and stability of the biolubricants.

Scale-up and Commercialization. Transitioning from laboratory success to industrial scalability and commercial viability remains a formidable barrier. Research must address the cost-effectiveness of synthesizing Gr-based nanoadditive and their integration into biolubricants. This includes conducting lifecycle assessments and techno-economic analyses to evaluate the environmental impacts and commercial prospects rigorously. The overarching question is whether the benefits in laboratory settings can be replicated on a mass scale while remaining economically feasible.

Real-World Application Testing. Extensive real-world testing is imperative for Gr-enhanced biolubricants to undergo the transition from theory to practical applications. Long-term performance assessments in the machinery and automotive sectors will provide invaluable data on their efficacy and durability. Moreover, comparative analyses with traditional petroleum-based lubricants in field tests will illuminate relative performance metrics and environmental impacts, offering a clear benchmark for progress.

Environmental Impact Reduction. Finally, the environmental footprint of Gr-based additives must be meticulously evaluated. While graphene itself is not biodegradable, the biolubricant base oils used in the formulation maintain their biodegradable properties, ensuring that the lubricants still offer significant ecological benefits. Research into their biodegradability and ecological effects is crucial to ensure that these advanced materials do not inadvertently harm the environment. Ongoing research into the long-term environmental persistence of graphene-based additives is critical. Although their tribological benefits are well-established, more studies are needed to fully understand their long-term impact on ecosystems. Evaluations of the environmental footprint of graphene-enhanced biolubricants have begun, and more real-world testing is needed to assess their effects over extended periods. Moreover, strategies to enhance the recyclability and reusability of spent lubricants and Gr-based materials will be pivotal in promoting a circular economy.

By addressing these critical areas, the research community can enhance the technological and environmental benefits of Gr-enhanced biolubricants, setting the stage for their widespread adoption across industries. This will contribute not only to the sustainability of mechanical systems but also to the overarching goal of reducing ecological footprints.

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Notes

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■ ABBREVIATIONS

AFM, atomic force microscopy
BC, bacterial cellulose
BIOE, biodegradable polymeric ester lubricant
CD, carbon dot
BMI, bismaleimide
CNC, cellulose nanocrystals
CNF, carbon nanofiber
CNSL, cashew nut shell liquid
CNT, carbon nanotube
COF, coefficient of friction
CTAB, cetyltrimethylammonium bromide
CVD, chemical vapor deposition
DLC, diamond-like carbon
EDS, energy dispersive spectroscopy
EG, graphene Nanoplatelets
g-C₃N₄, graphitic carbon nitride
Gr, graphene
GnP, graphene nanoplatelets
GO, graphene oxide
GQD, graphene quantum dot
hBN, hexagonal boron nitride
HOPG, highly oriented pyrolytic graphite
HRTEM, high-resolution transmission electron microscopy
IL, ionic liquid
KO, Karanja oil

MG, multilayer graphene
 MGr, multilayer graphene
 MLG, multilayer graphene
 MO, mineral engine oil
 MoDTC, molybdenum dithiocarbamate
 NCO, neat castor oil
 ND, nanodiamond
 NMR, nuclear magnetic resonance
 NPE, nonylphenol ethoxylates
 AO, oleic acid
 PAO, polyalphaolefin
 PDA, polydopamine
 PE, pentaerythritol
 PEEK, polyether ether ketone
 PEG, poly(ethylene glycol)
 PI, polyimide
 PNIPAM, poly(*N*-isopropylacrylamide)
 PO, Pongamia oil
 POE, polyol ester
 PPTA, poly-*p*-phenylene terephthalamide
 PTFE, polytetrafluoroethylene
 r-GO, reduced graphene oxide
 rGO, reduced graphene oxide
 RO, rapeseed oil
 SDBS, sodium dodecylbenzene sulfonate
 SDS, sodium dodecyl sulfate
 SEM, scanning electron microscopy
 TMD, transition metal dichalcogenide
 TMP, trimethylolpropane
 WSD, wear scar diameter
 WTW, wear track width
 XRD, X-ray diffraction
 ZDDP, zinc dialkyldithiophosphate

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