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Review

The application of nanocellulose in eco-friendly lubricants: A review

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Abstract: Lubricants are widely employed in mechanical systems to reduce energy loss and the wear induced by friction. With increasing concern for environmental protection, the development of eco-friendly lubricants has become increasingly significant. Nanocellulose, a natural material derived from cellulose, attracts increasing research attention in the field of lubrication due to its renewable, non-toxic, and biodegradable properties. Focusing on this emerging research topic, this state-of-the-art review first analyzes the theoretical basis of applying nanocellulose as a thickening agent in eco-friendly lubricating formulations. The following presents an overview of research advances in cellulose nanocrystals (CNCs) and cellulose nanofibrils (CNFs), categorized by their applications as additives or thickeners in aqueous, oil, and grease lubricants. This study also highlights the challenges associated with eco-friendly lubricants based on nanocellulose and offers suggestions for future investigations in this field. It is hoped that this review will inform the research direction of eco-friendly lubricants and promote the development of nanocellulose materials for industrial lubrication applications.

Keywords: eco-friendly lubricants; cellulose nanocrystals; cellulose nanofibrils; lubricating additives; thickening agents

1 Introduction

Friction leads to wear and energy loss, which directly affects the efficiency and lifespan of mechanical systems [1]. To mitigate these effects, proper lubrication is essential for reducing friction and wear. Lubricants play a critical role in this process, and their performance is often enhanced by functional additives, such as anti-wear and friction-reducing agents [2]. Traditional additives usually rely on reactive elements, such as phosphorus, sulfur, chlorine, and halogen compounds, which function through friction-induced chemical reactions to reduce wear and friction at the interface [3]. However, the presence of these reactive elements contributes to environmental pollution, leading to increased interest in alternative, eco-friendly materials [4-6].

Nanomaterials have emerged as promising candidates to address this challenge, owing to their unique size-dependent properties and versatile lubrication mechanisms [7-9]. Numerous studies [10-13] have investigated the tribological effects of nanoparticles, demonstrating their ability to improve lubrication efficiency across various applications. Unlike traditional additives, nanomaterials can reduce friction and wear through multiple physical mechanisms, such as the micro-rolling effect, mending effect, polishing effect, and the formation of protective films [14].

As a nanomaterial derived from natural cellulose sources, such as wood, animals, or bacteria, nanocellulose is attracting increasing research attention (Fig. 1) [15]. Nanocellulose possesses several desirable physicochemical properties at the nanoscale, including a high specific surface area and high mechanical strength [16,17]. In particular, the plentiful hydroxyl groups on CNC surfaces facilitate effective surface modifications, making them adaptable across different fluids [18]. Based on morphology and preparation techniques, nanocellulose is broadly classified into three types: cellulose nanocrystals (CNCs), cellulose nanofibers (CNFs), and bacterial nanocellulose (BNC) [19,20]. Among these, CNCs and CNFs have shown the most potential in lubrication-related applications.

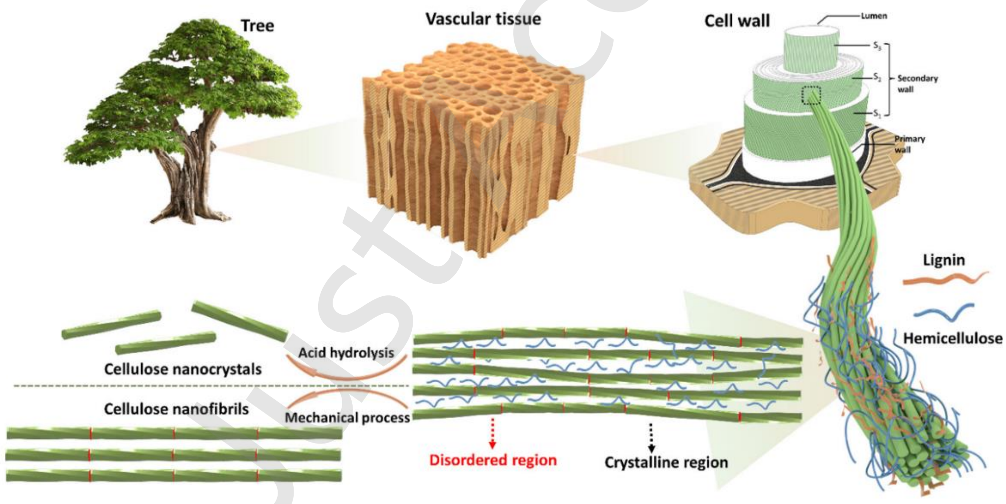

Fig. 1 The obtainment and classification of nanocellulose. Reproduced with permission from Ref. [15]. © Chemical Reviews, 2023.

CNCs are typically rod-like nanocrystals with high crystallinity (54-88%), obtained by removing amorphous regions through acid hydrolysis. These particles typically have diameters of 5-20 nm and lengths of 100-500 nm [20,21]. The surface properties of CNCs depend on the type of acid used in hydrolysis. For instance, hydrochloric acid yields weakly negatively charged particles, while sulfuric acid introduces sulfate groups, imparting a stronger negative charge [22-24]. Specifically, CNC suspensions exhibit a phase transition from isotropic to partially anisotropic, and eventually to liquid-crystalline phases, at certain concentrations. This ordered structure may play a potential role during the lubrication process [25].

CNFs are primarily produced through intense mechanical treatments, such as shearing, grinding, or high-pressure homogenization. These methods partially disrupt the crystalline structure, yielding long, flexible nanofibers that retain both crystalline and amorphous regions. Compared with CNCs, CNFs also have diameters in the nanoscale (10-100 nm), but much larger lengths up to several micrometers (1-15 μm) and smaller crystallinity (51-69%) [26]. In solvents, CNF easily forms a three-dimensional network, displaying gel-like behavior and high viscoelasticity even at low concentrations [20]. These rheological features are particularly relevant for applications in greases and structured lubricants.

In recent years, CNCs and CNFs have been introduced into various lubricant systems, including aqueous lubricants, lubricating oil, and lubricating greases. This review provides a comprehensive overview of the application of nanocellulose in lubricants. It begins with a theoretical analysis of nanocellulose for lubrication. Recent advances in this field are then summarized, covering their tribological performance across different lubricant systems and the underlying lubrication mechanisms. Finally, the remaining technical challenges and future research prospects for developing eco-friendly lubricants are discussed.

2 Theoretical analysis of nanocellulose applied in lubrication

2.1 Environmental friendliness

Nanocellulose, characterized by its non-toxicity, high biodegradability, and excellent biocompatibility, offers clear environmental advantages as a lubricating additive, particularly in environmentally sensitive fields such as marine, food processing, and agriculture. In marine applications, the persistence of mineral oils in seawater poses risks to aquatic ecosystems. To mitigate these impacts, regulations such as the Vessel General Permit (VGP) and the Vessel Incidental Discharge Act (VIDA) require the use of Environmentally Acceptable Lubricants (EALs), which must meet strict criteria for biodegradability and toxicity. CNCs from CelluForce, for instance, achieve a 62.5% theoretical oxygen demand within 28 days (OECD 306), satisfying VGP's requirement of at least 60% mineralization within 10 days after biodegradation begins. This demonstrates that CNCs can be incorporated into marine lubricant formulations (e.g., water-based lubricants for enclosed stern bearings and greases using plant oils for wire ropes) without compromising overall environmental compliance. In the food processing industry, lubricant safety is paramount, as leakage from conveyor belts, mixers, and motor drive systems can directly contaminate food. Although traditional food-grade lubricants rely on synthetic esters or specialized mineral oils, they remain costly and are only partially biodegradable. Nanocellulose, by contrast, has been extensively utilized in drug encapsulation due to its exceptional biocompatibility and controlled release properties [27,28]. This well-established application in the pharmaceutical industry underscores its superior safety and reliability as a lubricating additive for food machinery. While classified as "Generally Recognized as Safe" (GRAS), it has not yet been approved by the U.S. Food and Drug Administration as a food ingredient, necessitating further toxicological studies before its large-scale adoption [29-31].

In other industrial fields, environmentally friendly lubricants are also required. Wind turbines, often deployed in ecologically sensitive areas, demand biodegradable lubricants to prevent contamination [32]. Similarly, lubricants used in the machining industry have been encouraged to be safe and healthy [33]. Agricultural machinery also faces environmental concerns, as lubricant leakage can contaminate soil and disrupt microbial ecosystems [34]. Therefore, nanocellulose is a promising material for green lubricating additives in various applications.

2.2 Physical properties

As mentioned above, cellulose nanocrystals (CNCs) and cellulose nanofibers (CNFs) typically have diameters ranging from approximately 5 to 20 nm and 10 to 100 nm, respectively, giving them the small-size effect commonly seen in nanoparticles. In theory, nanocellulose can function similarly to traditional nanoparticle additives, including metals, metal oxides, sulfides, carbon materials, and nanocomposites. It may help fill microscopic defects on friction surfaces, reduce surface roughness, and form protective films. These effects can improve lubrication performance [35-37].

From a mechanical perspective, CNCs exhibit exceptionally high stiffness, with Young's modulus between 60 and 200 GPa [38], comparable to that of Kevlar fibers [39]. This rigidity contributes to their load-bearing capacity, potentially reducing friction under high-load lubrication conditions. By combining their rod-like morphology, CNCs have the potential to transform sliding friction into rolling friction (i.e., the micro-rolling effect), as observed in nanodiamond [40] and carbon nanotube [41] additives. In addition to their impact on the sliding surfaces, CNCs can also affect lubrication by altering the rheological properties of lubricating fluids, particularly at high CNC concentrations.

Compared with CNCs, CNFs possess longer and more flexible fibers. Through chain entanglement and intermolecular interactions, including hydrogen bonding, electrostatic forces, or van der Waals forces, CNFs form intense three-dimensional networks in the oil [42]. Therefore, CNFs can serve as thickening agents at specific concentrations, which are much more eco-friendly than metallic soaps in conventional greases. For instance, lithium-based greases are poorly soluble in water and exhibit extremely slow degradation rates in natural environments. Once released into the environment through road runoff or improper disposal, lithium greases can persist for long periods, contaminating soil and groundwater[43,44].

2.3 Chemical properties

CNC and CNF surfaces contain a large number of hydroxyl groups. Crystallographic models of native cellulose I_β show that the dominant hydrophilic faces expose about 5.4 hydroxyl groups per square nanometer (about 3.6 hydroxyl groups per square nanometer effectively free for bonding) [45]. Therefore, the compatibility of nanocellulose in both water- and oil-based lubricants can be further enhanced by surface chemical modification. Considering that the dispersion of nano-additives in lubricants is a universal problem [46], the tunable surface chemistry of nanocellulose is advantageous for lubrication applications.

To improve aqueous compatibility, nanocelluloses are commonly modified to introduce charged or hydrophilic surface groups. Typical strategies include sulfation and carboxylation. It should be noted that CNCs obtained by sulfuric acid hydrolysis are inherently sulfated due to the attachment of sulfonate groups during preparation. Here, the term "sulfation" refers to either this inherent modification in CNCs or to additional chemical sulfation treatments (e.g., chlorosulfonic acid) that can further increase surface charge density. Sulfonation attaches sulfonate groups to the surface, while TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl)-mediated oxidation selectively converts primary C6-OH to $-COO^{-}$ under mild conditions. These anionic surfaces greatly enhance colloidal stability in water. For instance, a one-step sulfonation of mechanically fibrillated CNFs was shown to yield sulfonated CNFs that disperse far more readily in both DMF and water than either unmodified CNFs or even TEMPO oxidation-modified CNFs [47]. Similarly, TEMPO oxidation-modified CNFs (TOCNs) have been successfully blended into waterborne coatings: by adding 0.1-5 wt.% TOCNs to a waterborne polyurethane (WPU) paint produced a uniform dispersion [48].

In oil-based or non-polar environments, nanocellulose must be hydrophobized by grafting hydrophobic chains or moieties onto its -OH groups. Standard chemistries include esterification with acyl chlorides or fatty acids, urethanization with isocyanates, and silylation with chloromethyl isopropylsilane or triethoxysilane. For example, hydrophobic modification of CNCs via esterification with medium-chain fatty acids can significantly improve their dispersibility in epoxy resins [49]. Grafting cetyltrimethoxysilane onto CNC surfaces has been shown to impart superhydrophobicity, with water contact angles exceeding 150° [50]. These findings underscore the versatility of nanocellulose surface chemistry and highlight its promising potential for lubrication applications.

2.4 Economic feasibility

Cellulose is the most abundant natural polymer in nature and can be extracted from a wide range of renewable sources. In addition to wood, cellulose can be derived from agricultural residues such as straw, corn stalks, and bagasse, as well as industrial by-products and waste streams, including paper sludge, textile waste, and marine biomass [51]. These sources are widely available and low-cost, which aligns well with the principles of the circular economy and sustainability. However, it should be noted that common methods for producing CNCs, such as strong acid hydrolysis, may raise environmental concerns due to the generation of acidic effluents, which highlights the need for more sustainable preparation strategies.

In addition to the cost of raw materials, the cost of inputs in the production process also significantly impacts the product's price. The production of nanocellulose has reached a relatively mature stage, supported by scalable mechanical, chemical, and enzymatic processes. Industrial-scale manufacturing has been realized in several regions. For instance, Chuetsu Pulp & Paper has developed nanocellulose products for applications in lubricants, food packaging, and coatings. Other key institutions, including CelluForce, the Forest Products Laboratory (USDA), Melodea, Holmen, InnoTech Alberta, and FPInnovations, have also advanced nanocellulose production technologies and expanded their use into lubrication and related fields [52].

However, surface modification is often required to improve the dispersibility and functionality of nanocellulose in lubricants. These modification steps may increase the overall production cost and should be considered in economic evaluations. However, these additional processing steps, such as chemical grafting, purification, and drying, can significantly increase the overall production cost and complexity. Therefore, in addition to improving nanocellulose production, it is also crucial to enhance the efficiency and reduce the cost of its surface modification for lubrication applications.

3 Research advances in nanocellulose-based lubricants

Based on the above analysis, nanocellulose is theoretically feasible for lubrication applications, particularly with great potential for the development of environmentally friendly lubricating materials. In this research direction, numerous noteworthy studies have emerged in recent years. This section provides an overview of the advances in the application of nanocellulose in various types of lubricants, with a focus on its lubrication performance and mechanisms in aqueous, oil, and grease lubricants.

3.1 Applied as additives in aqueous lubricants

3.1.1. Dispersion behavior

The dispersion stability of nanoparticles in lubricating fluids is a key factor affecting their tribological performance. Although nanocellulose is rich in surface hydroxyl groups, agglomeration and sedimentation often occur in practice. On the one hand, nanocellulose exhibits an inherently high surface energy, arising from its large specific surface area, which is similar to that of other nanoparticles. On the other hand, the hydroxyl groups on the surface of nanocellulose can not only form hydrogen bonds with water but also form intermolecular hydrogen bonds of themselves (Fig. 2), which cause the fibers to stick together and form small aggregates [19]. In particular, for the CNCs obtained by sulfuric acid hydrolysis, although the introduced sulfate groups could increase the negative charge density on the surface (100-400 mmol/kg) [53], their dispersibility in aqueous lubricants still needs to be further enhanced, especially at high concentrations, considering the long service time in practical applications.

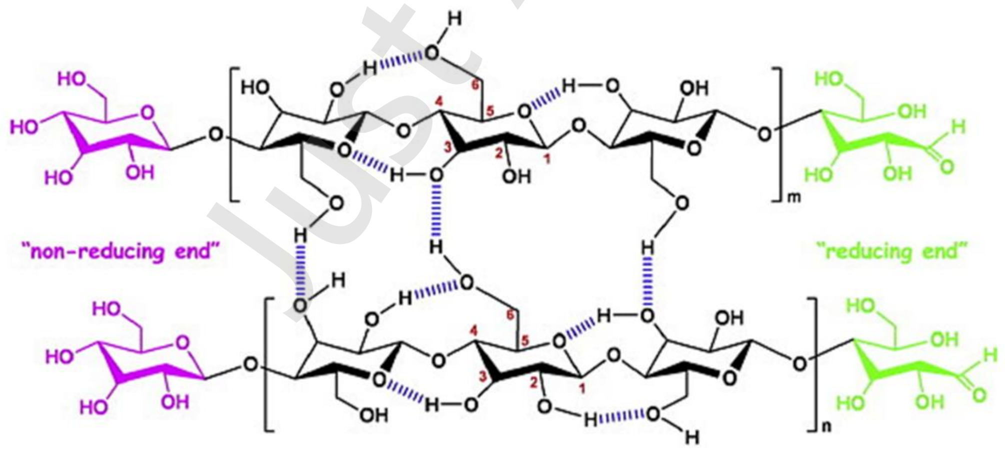

Fig. 2 The intra-molecular and inter-molecular hydrogen bonds in nanocellulose. Reproduced with permission from Ref. [19]. © European Polymer Journal, 2014.

To address this issue, various physical methods, such as ultrasonic treatment, high-pressure homogenization, and mechanical shearing, are used to weaken hydrogen bonds and facilitate nanocellulose dispersion. For example, Zakani et al. [54] investigated the effect of ultrasonic treatment on the lubrication performance of CNC aqueous suspensions. The sulfated CNC suspension provided by CelluForce was diluted to 0.25, 0.75, 1, 2, and 3 wt.% by adding deionized water. The results showed that at 1 wt.%, without any treatment, it exhibited significant aggregation, forming large agglomerates, indicating insufficient dispersion. As sonication energy increased, CNC aggregates progressively broke apart, leading to more homogeneous dispersions. Sonication at 0.5 kJ/g partially loosened the agglomerates, while higher energy (e.g., 1 kJ/g or above) effectively broke down larger aggregates into smaller clusters or individual nanocrystals, significantly improving dispersion uniformity (Fig. 3a-e). However, excessive ultrasound exposure might damage the structure and reduce the crystallinity or mechanical properties of CNCs [55-57]. Therefore, further studies should optimize ultrasound energy and treatment time to achieve a favorable dispersion effect while minimizing damage to CNCs' physical properties.

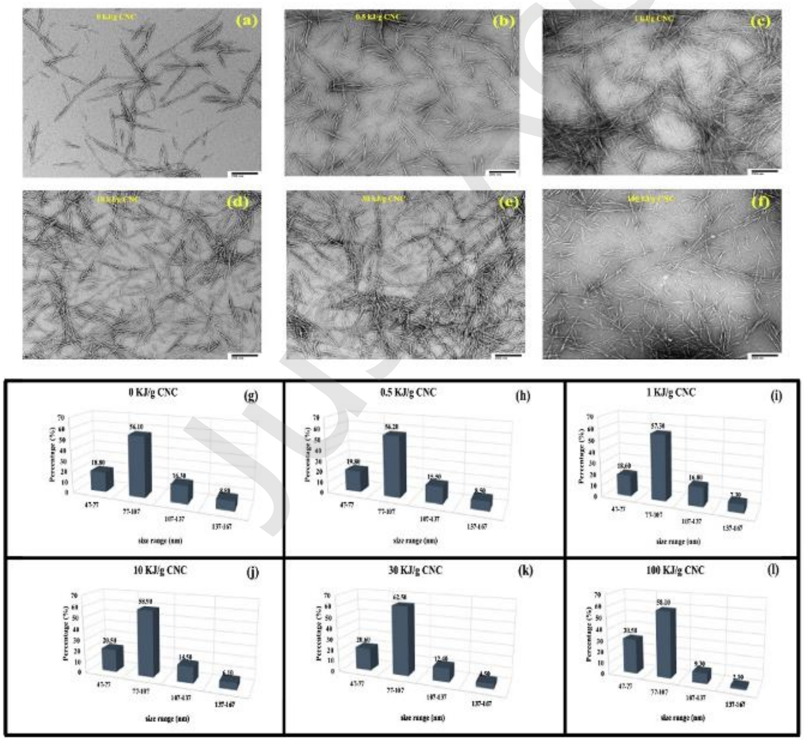

Fig. 3 TEM images representing the morphology of CNC nanoparticles at a magnification of 100,000×, under different sonication energy inputs of 0 (a), 0.5 (b), 1 (c), 10 (d), 30 (e), and 100 (f) kJ/g CNCs, respectively. Reproduced with permission from Ref. [54]. © Carbohydrate Polymers, 2022.

In addition to physical methods, surface modification can also enhance the dispersion stability of nanocellulose (Fig. 4) [53]. A common strategy to elevate electrostatic repulsion is to introduce anionic groups (e.g., sulfate, carboxyl, and phosphate) or cationic groups (e.g., amines and quaternary ammonium). Etale et al. [58] reported a method for hydrolyzing cellulose with phosphoric acid, followed by phosphite esterification, to introduce phosphate groups on the surface. The high density of phosphate groups (up to ~1.8 mmol/g) endowed the nanocellulose with a strong negative charge. Zaman et al. [59] prepared CNCs via sulfuric acid hydrolysis and grafted glycidyltrimethylammonium chloride (GTMAC) onto their surfaces. In such a way, the zeta potential of CNCs varied from -57 mV to +63 mV. Another strategy is to introduce long-chain polymers to enhance steric hindrance. Araki et al. [60] conjugated poly(ethylene glycol) (PEG) with an amino group to the surface of TEMPO-oxidized CNCs through amide bonds, which exhibited much better dispersibility than the unmodified CNCs in water. Furthermore, it is also feasible to increase steric hindrance by using surfactants through physical adsorption. For example, Shang et al. [61] adsorbed hexadecyl trimethyl ammonium bromide (CTAB) onto hydrochloric acid-hydrolyzed CNCs. Based on the steric resistance and electrostatic shielding, the average particle size of the CNC aggregates decreased from 2.35 μm to 470 nm.

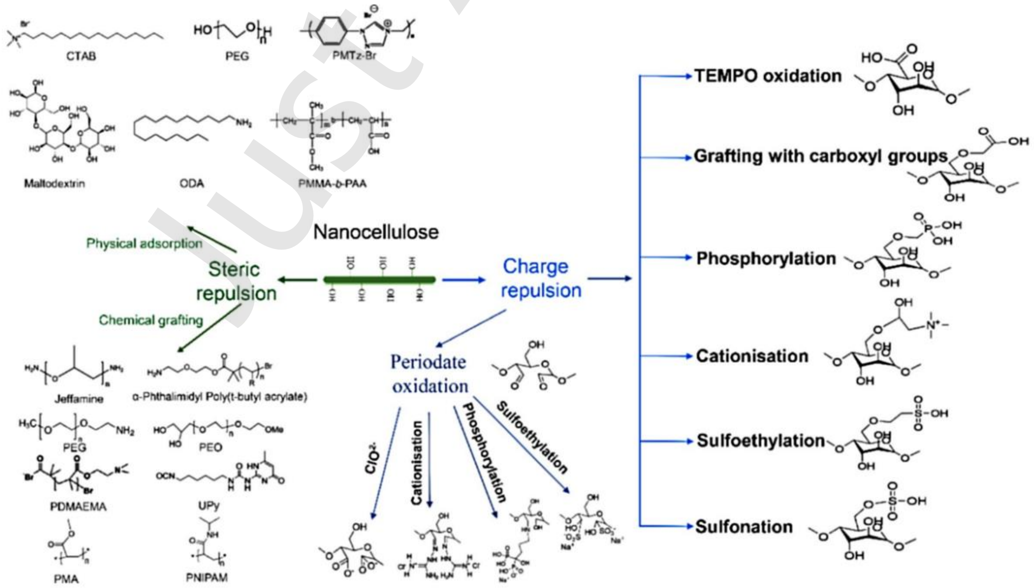

Fig. 4 Surface modification for enhancing nanocellulose dispersion stability through charge and steric repulsion. Reproduced with permission from Ref. [53]. © Carbohydrate Polymers, 2020.

3.1.2 Lubrication performance

Many studies have reported the effectiveness of CNCs as an additive to aqueous lubricants. Zhang et al. doped S-CNC (CNC with sulfonyl groups) and C-CNC (CNC with carboxyl groups), prepared by sulfuric acid hydrolysis and oxidation, as additives in water. S-CNC and C-CNC reduced the coefficient of friction (COF) by 63.5% and 55.84% respectively, due to the micro-rolling effect of CNC particles. Shariatzadeh et al. [62] incorporated 2 wt.% of sulfated CNCs into water and found that it lowered the COF and wear depth by 72% and 54%, respectively. This lubrication improvement was attributed to the mending effect of CNCs on the wear track. Building upon these findings, Zakani et al. [63] made a comprehensive study on the influence of CNC concentration across a wide range from 0.005 wt.% to 5 wt.%.% (Fig. 5). In the dilute regime (CNC concentration <1 wt.%), increasing CNC content effectively reduced both COF and wear in boundary lubrication. In the semi-dilute regime (CNC concentration between 1 wt.% and 2 wt.%), friction and wear reached a minimum. When the CNC concentration was more than 2 wt.%, the excessively high viscosity of suspensions would degrade its lubrication performance. It should be pointed out that the exact critical values of CNC concentration in Fig. 5 may vary by changing the specific material types of CNCs or the operating conditions of the tribological system. However, the revealed correlation of the microstructure, rheological behavior and tribological performance of CNC suspensions is of great significance.

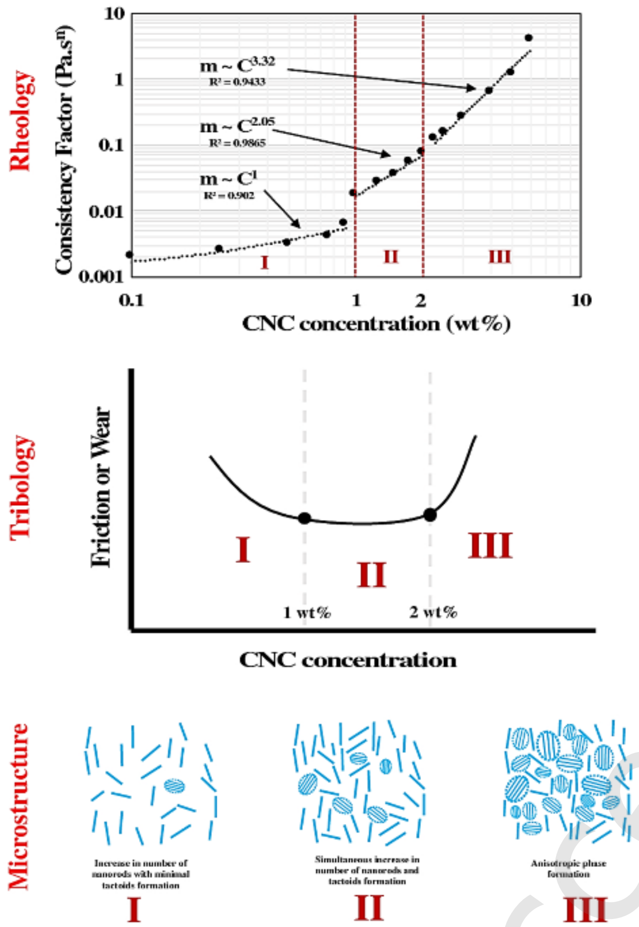

Fig. 5 The evolution of the consistency factor, friction properties (schematic), and microstructure (schematic) by increasing the concentration of CNC suspension. Reproduced with permission from Ref. [63]. © Cellulose, 2022.

To further enhance the performance of CNC additive, Cheng et al. [64] prepared semi-carbonized CNCs (scCNC) via an ionic liquid-thermal method. By partially removing surface hydroxyl groups, the thermal stability and chemical inertness of CNCs were improved. In the friction test, scCNC produced a large number of carbon dots, which entangled and formed micron-sized, net-like thin films. These thin films not only reduced direct contact between tribopairs but also limited the generation of wear debris (Fig. 6). As a result, scCNC reduced the COF by 22% and the wear volume by 49% compared to the case using pristine CNCs.

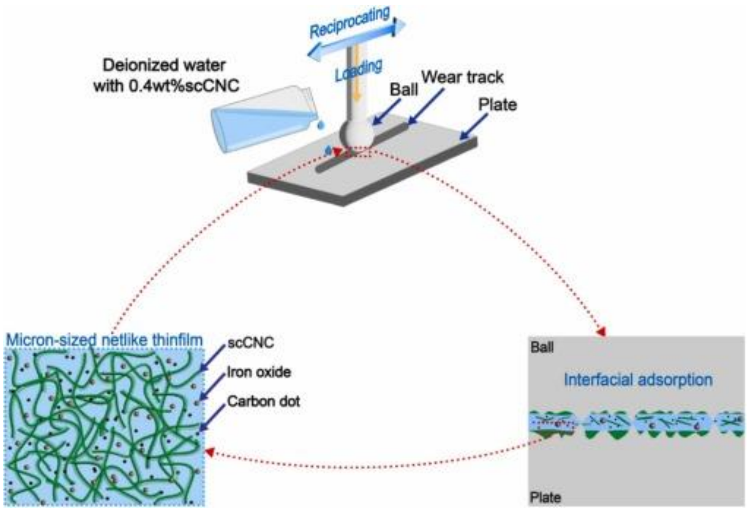

Fig. 6. Schematic diagram of the lubrication mechanism for scCNC as a water-based additive. Reproduced with permission from Ref. [64]. © Tribology International, 2025.

Compared with CNCs, CNFs possess much higher aspect ratios and tend to form entangled micrometer-scale networks. Therefore, rather than the micro-rolling or mending effect of CNCs at the sliding surface, CNFs facilitate the lubrication by adjusting the rheological behavior of the lubricating fluids in the bulk. From quince seeds, Hakala et al. [65] extracted mucilage, which consists of CNF and charged hemicellulose. Tribological pin-on-disc tests showed that the incorporation of mucilage to water was able to reduce the COF of polyethylene/stainless steel contact to values below 0.03 (Fig. 7a). The viscosity of the lubricant showed an exponential increase as a function of the mucilage (Fig. 7b). Thus, a substantial increase in viscosity and reduction of COF occurred in the same concentration range.

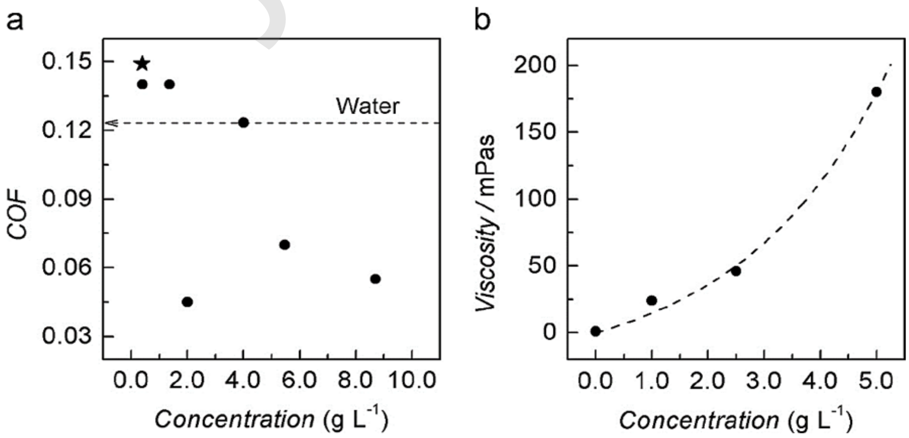

Fig. 7 (a) Friction coefficient of as-extracted quince mucilage as a function of concentration (4.4 MPa nominal contact pressure); (b) Exponential increase of viscosity of as-extracted quince mucilage as a function of concentration at 15 s^{-1}. Reproduced with permission from Ref. [65]. © Tribology International, 2014.

3.2 Applied as additives in lubricating oils

3.2.1 Dispersion behavior

In lubricating oils, the dispersion of nanocellulose presents greater challenges than in aqueous liquids. For instance, Awang et al. [66] added CNCs extracted from acetate-grade dissolving pulp to SAE 40 engine oil at various doping levels ranging from 0.1% to 0.9%. The dispersion of CNCs in the hybrid oils was recorded using photographs at different time intervals (Fig. 8), revealing clear sedimentation of CNCs, even in samples with low CNC concentrations. It should be noted that, as the small phase separation was difficult to observe just using photographs, the actual dispersibility of CNCs in lubricating oils might be worse than the time shown in Fig. 8. Therefore, it is necessary to improve the dispersion of nanocellulose via surface modifications.

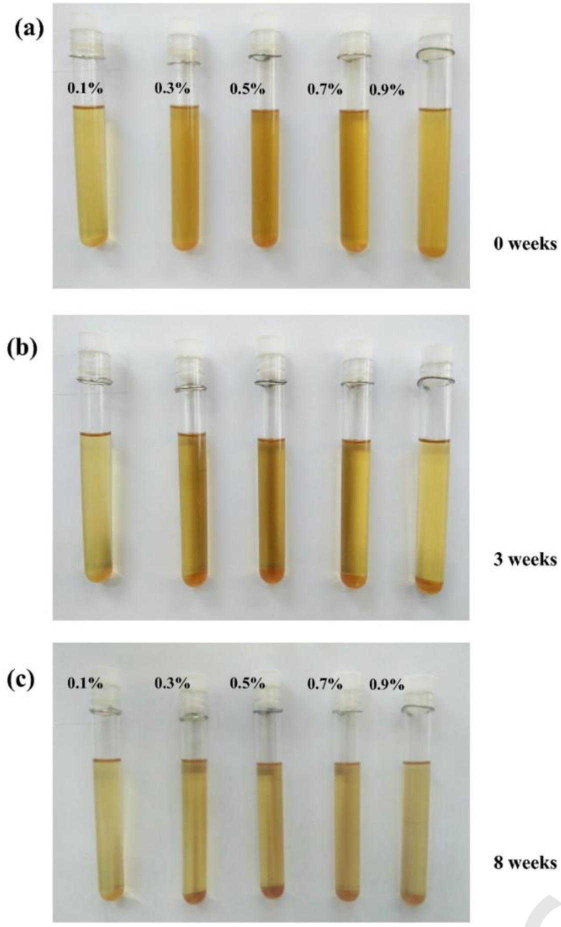

Fig. 8 Sedimentation photographs of CNCs dispersed in engine oil SAE 40. Reproduced with permission from Ref. [66]. © International Journal of Heat and Mass Transfer, 2019.

The primary strategy for surface modification of CNCs is to introduce appropriate segments that are compatible with the corresponding base oils. For example, CNCs prepared from cotton were grafted by stearoyl chains to enhance their lipophilicity [67] (Fig. 9a). These modified CNCs exhibited better dispersibility, anti-friction, and anti-wear ability than the pristine CNCs in polyalphaolefin (PAO) oil. In perfluoropolyether (PFPE) oil, polyhexafuorobutyl methacrylate (PHFMA) was grafted onto CNCs via atom transfer radical polymerization (ATRP) [68] (Fig. 9b). Such a surface functionalization increased the water contact angle of CNCs from 41° to 113°. The obtained CNC-g-PHFMA particles could stably disperse in PFPE oil for more than 60 days [69].

Plant oils attract increasing research attention due to their environmental friendliness. With the expectation of an eco-friendlier lubricant formulation, CNCs have also been incorporated into plant oils. Based on TEMPO-oxidized CNCs, Cai et al. [70] grafted alkylboronic acid through an esterification reaction and added the esterified CNCs to castor oil. Compared with the suspension state of pristine CNCs in castor oil, the esterified CNCs-doped oil formed a typical colloidal solution with a stable dispersion for more than 7 days. In sunflower oil, Shan et al. [71] prepared ionic liquid crystal functionalized nanocellulose (ILC-NC) by integrating 1-hexadecyl-3-methylimidazolium bromide onto nanocellulose (Fig. 9c). The results showed that ILC-NC dispersions (1 wt.%) remained homogeneous in sunflower oil without noticeable sedimentation for at least 30 days, far outperforming pristine nanocellulose.

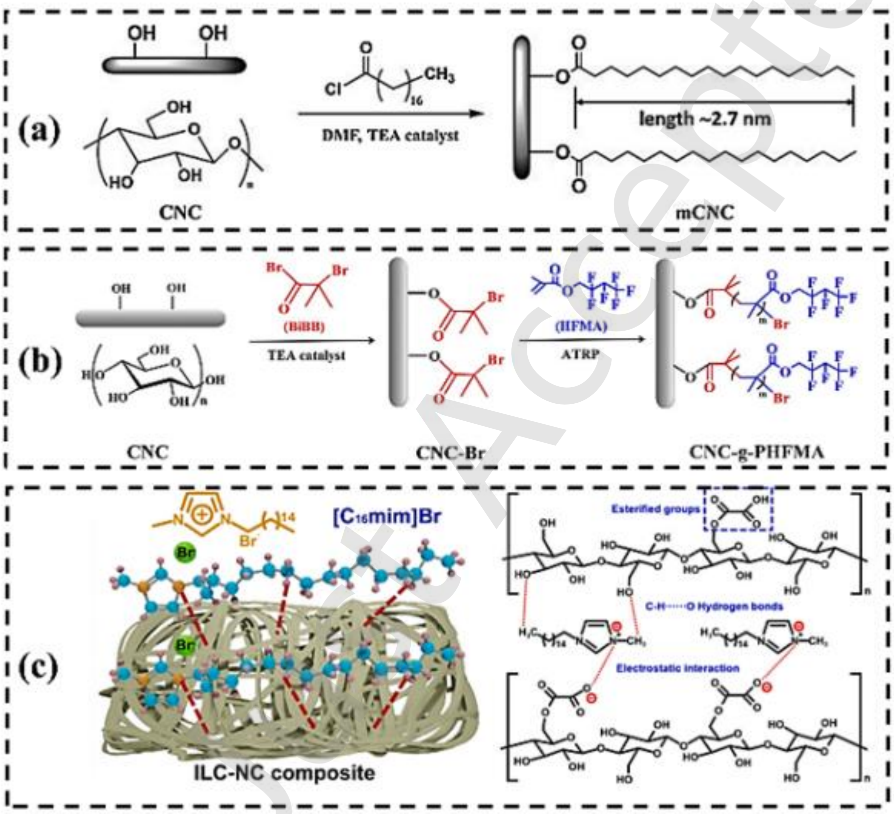

Fig. 9 (a) Surface modification on cellulose nanocrystals by stearoyl chains; (b) Surface modification of cellulose nanocrystals by grafting fluoropolymer; (c) Interactions within ionic liquid crystal functionalized nanocellulose (ILC-NC). (a) Reproduced with permission from Ref. [67]. © Carbohydrate Polymers, 2019. (b) Reproduced with permission from Ref. [68]. © Cellulose, 2023. (c) Reproduced with permission from Ref. [71]. © Carbohydrate Polymers, 2025.

3.2.2 Lubrication performance

The lubrication performance of nanocellulose additives depends not only on the nanocellulose material itself but also on the base oil and operating conditions. Fernández-Silva et al. [72] systematically examined lubricating oils with various compositions in different lubrication regimes. CNCs obtained by sulfuric acid hydrolysis and CNFs obtained by mechanical treatment were introduced into castor oil, high oleic sunflower oil, and their blended oil, respectively. When dispersed in castor oil, the abundant hydroxyl groups of ricinoleic acid molecules formed hydrogen bonds with nanocellulose, thereby facilitating the formation of a dense, stable lubricating film. In contrast, high-oleic sunflower oil exhibited much lower polarity, resulting in weaker interactions with nanocellulose and poorer lubrication performance. When the doped content exceeded 3.3 wt.%, both CNCs and CNFs significantly reduced friction and wear in the boundary and mixed lubrication regimes. However, the high viscosity of the lubricant increased friction. Therefore, the concentration of nanocellulose needed to be appropriately controlled.

Concerning the lubrication mechanism, nanocellulose particles can act as nanoscale fillers to compensate for the roughness and wear track of sliding surfaces. Based on this mending effect, it has been demonstrated that CNCs functionalized with ionic liquid crystals reduced the COF by up to 77.91% in sunflower oil [73]. In addition, nanocellulose could improve lubrication by adjusting oil rheology. When 2 wt.% of surface-modified CNCs (mCNC) was added to PAO oil [67], the grafted stearoyl chains on the surface of mCNC made their dispersion in PAO oil form a stable colloidal solution. The increased viscosity of the hybrid oil facilitated the formation of a lubricant film and the separation of solid contacts in the boundary lubrication regime. The combined effects of CNCs on the sliding surface and the oil fluid are illustrated in Fig. 10.

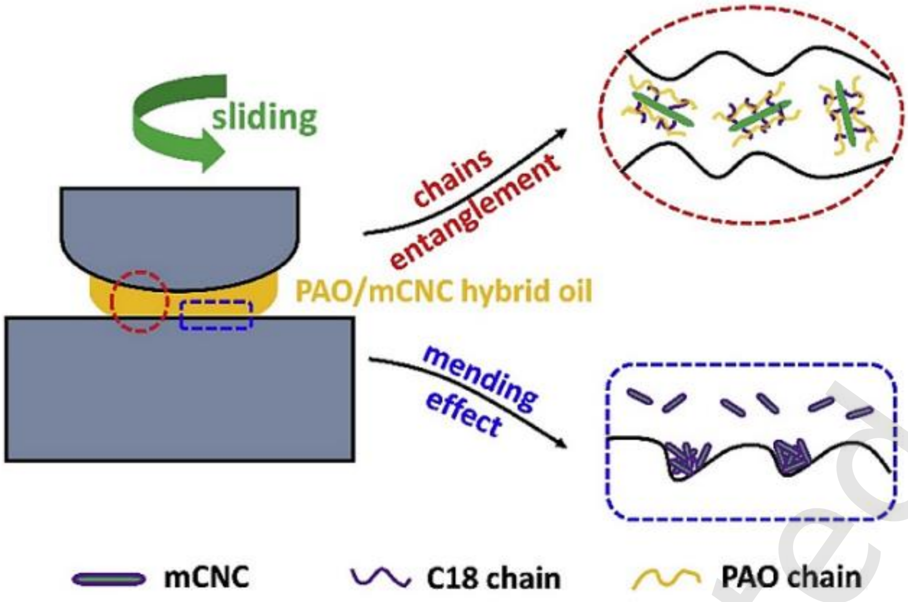

Fig. 10 Schematic of the proposed lubricating mechanisms of PAO/mCNC hybrid oil. Reproduced with permission from Ref. [67]. © Carbohydrate Polymers, 2019.

Moreover, some studies have stated that nanocellulose can form complex tribofilms, providing an enhanced load-carrying capacity and long-term protection. Cai et al. [70] functionalized CNCs using alkylboronic acid and doped them in castor oil (Fig. 11). The esterified CNCs (AEC-5-80) created a "nano-net" structure on steel surfaces and adsorbed oil to form the complete adsorption film. Furthermore, the borate ester groups directly adsorbed on the steel, initiating tribochemical reactions under frictional heat and force, forming a chemical transfer film. A similar result has also been reported in a study of CNCs grafted with fluoropolymer chains (CNC-g-PHFMA) as an additive for PFPE oil [68]. Compared with PTFE, CNC-g-PHFMA formed a more intensive and stable tribofilm due to its high mechanical strength.

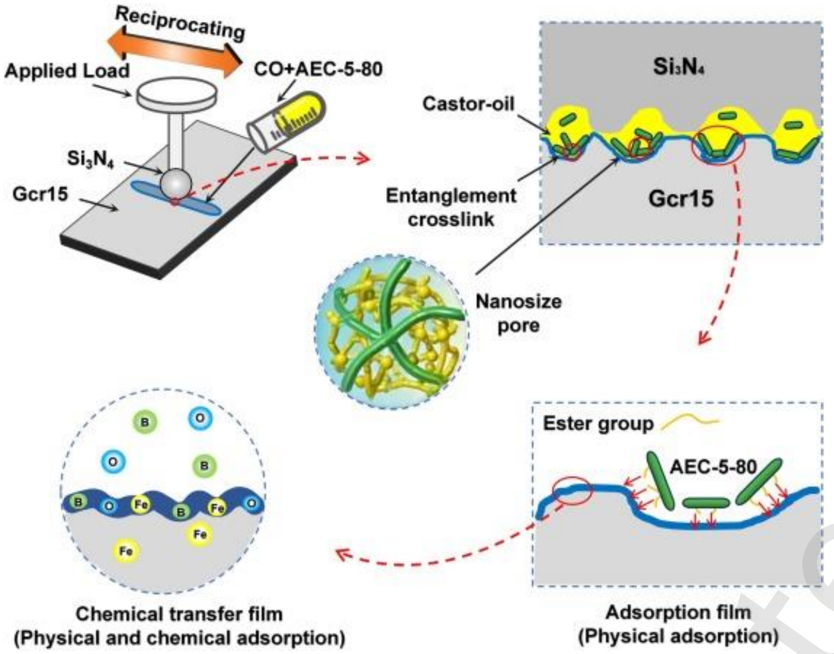

Fig. 11 Schematic illustration of the lubrication mechanism of esterified CNCs (AEC-5-80) in castor oil. Reproduced with permission from Ref. [70]. © Chemical Engineering Journal, 2024.

3.2.3 Hybrid additives

Some synergistic effects have been reported when nanocellulose is combined with other nanoparticles as a hybrid additive. A primary motivation is that nanocellulose can serve as a structural scaffold to enhance the interfacial compatibility and dispersion stability of other nanoparticles. Shan et al. [74] prepared a hybrid additive of nanocellulose (NC) and fluorinated graphite (FG). FG nanocomponents were domain-limited within the NC-constructed interwoven layers, achieved via mechanical ball-milling and freeze-induced self-assembly. When the NC@FG additive was introduced in sunflower oil, a nanocellulose network was formed via van der Waals forces and mechanical locking. By wrapping oil molecules and FG particles within the network, the hybrid oil remained stable and homogeneous for up to 30 days. Similarly, nanocellulose formed a protective "armor" when it was coated on carbon nanotubes (CNTs), resulting in highly improved dispersibility of CNTs in sunflower oil [75].

Another motivation of hybrid additives is to improve the thermophysical properties of lubricating oils. For example, CNC and copper oxide powders were physically blended through dry mixing [76]. The obtained CNC-CuO composite nanoparticles increased the viscosity index of SAE 40 oil from 129 to 154, thereby stabilizing viscosity and maintaining the lubricant film at high temperatures. Additionally, the addition of CNC-CuO increased the lubricant's thermal conductivity by 180%, improving heat dissipation during rubbing.

In terms of tribological properties, nanocellulose primarily exhibits synergistic effects with other nanoparticles to enhance lubrication performance. For example, Shan et al. [77] coated polytetrafluoro-wax (PFW) with polydopamine (PDA) first, and then hydrothermally reacted it with MXene to obtain MXene/PFW@PDA. Through a process of ultrasonic dispersion and lyophilization with nanocellulose (NC), the NC-MXene/PFW@PDA additive was prepared (Fig. 12a). As a hybrid additive in PAO oil, NC-MXene/ PFW@PDA entered the gap of the friction pairs under the shear stress. With the confinement effect of NC, MXene/PFW@PDA was evenly dispersed on the sliding surface, forming a transfer film. The synergistic effect of the hybrid additive greatly reduced the wear (Fig. 12b).

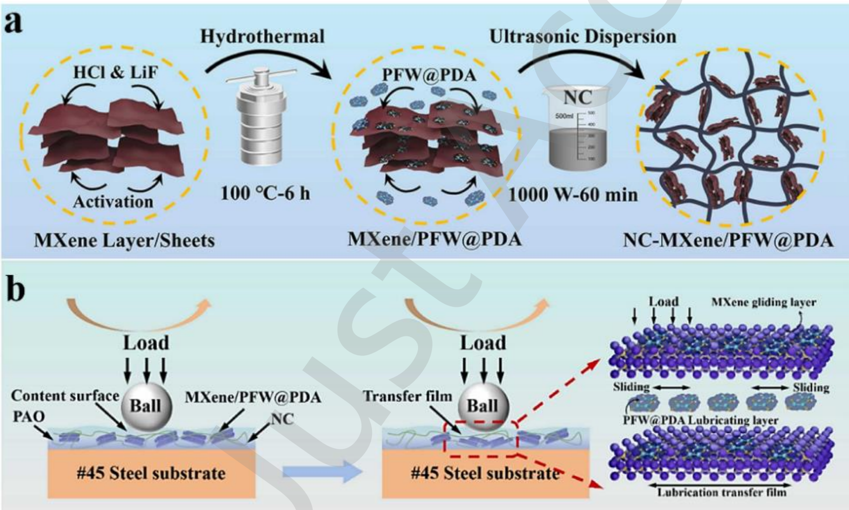

Fig. 12 (a) Preparation process and (b) lubrication mechanism of NC-MXene/PFW@PDA. Reproduced with permission from Ref. [77]. © Tribology International, 2023.

3.2.4 Electric response

Recently, the electrorheological (ER) behavior of nanocellulose has garnered growing interest in tribology, particularly for the design of smart lubricants [78,79]. García-Morales et al. [80] investigated the ER effect of CNCs and CNFs in castor oil (Fig. 13). Without an electric field, the nanocellulose dispersions behaved as a Newtonian fluid. When an electric field was applied, nanocellulose became polarized and aligned along the field direction, forming chain-like structures. As the shear rate increased, the chain-like structure established by the electric field was gradually destroyed, and the flow properties approached those of a Newtonian fluid. The critical shear rates shifted to higher values with increasing electric field strength, because higher hydrodynamic forces are required to overcome electrostatic interactions. To provide direct evidence of these structuring processes of nanocellulose caused by the electric field, Fernández-Silva et al [81] validated the electro-induced assembly of CNCs and CNFs through an in-situ optical microscopy visualization.

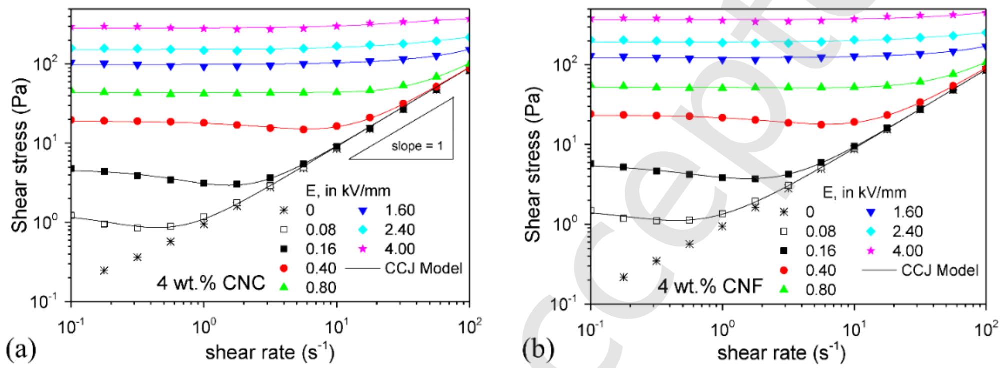

Fig. 13 Steady state flow curves of castor oils doped with 4 wt.% (a) CNC and (b) CNF under electric fields. Reproduced with permission from Ref. [80]. © Processes, 2020.

Delgado-Canto et al. [82] investigated the relationship between the ER effect and tribological performance of the same lubricating system (Fig. 14). The leak current intensity increased with increasing voltage, indicating that the CNF structure evolved toward more ordered columnar formations. In the boundary lubrication regime (velocity < 0.2 mm/s), the polarized nanoparticles electroadhered near the contact region, thereby increasing the COF at high electric field voltages of 10 V and 40 V. However, at a low electric field of 1 V, the attracted particles served as a filling/separating layer at the micro-contact, reducing the actual contact area and consequently lowering friction. In the mixed lubrication regime (i.e., the velocity was between 0.2 and 8 mm/s), the lubricant at all electric voltages achieved a smaller COF than in the case without an electric field. The reason is that the field-induced structural change enhanced the lubricant's bulk viscosity and improved its load-carrying capacity, thereby reducing friction. However, at velocities above 8 mm/s, the hydrodynamic effect of the lubricant fluid dominated the friction behavior, so that the electric field no longer improved tribological performance in the fluid lubrication regime.

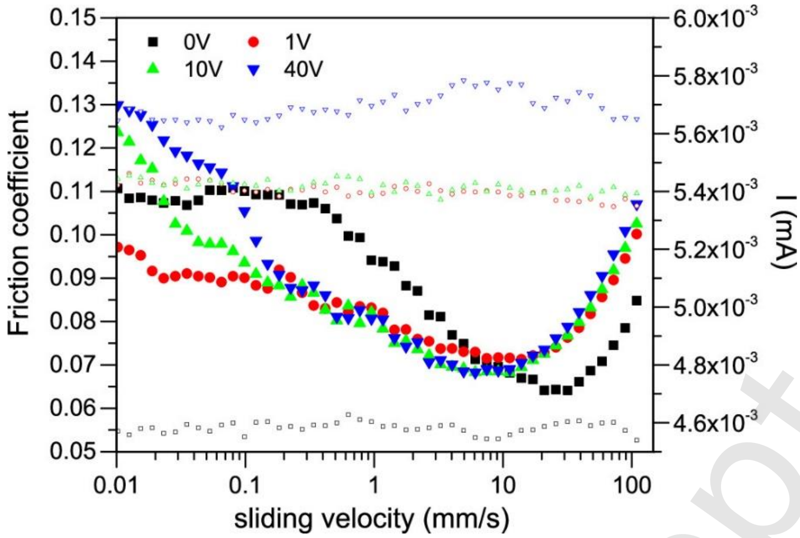

Fig. 14 The friction coefficient and leak current intensity (I) of castor oil doped with 4 wt.% CNF at varied sliding velocities and electric voltages. Reproduced with permission from Ref. [82]. © ACS Applied Materials & Interfaces, 2020.

3.3 Applications in lubricating greases

3.3.1 Applied as additives

In addition to being used as an additive for fluid lubricants, nanocellulose can also be applied in lubricating greases, where the nanoparticles can be stably trapped in the three-dimensional matrix of soap fibers, maintaining a uniform dispersion. In a series of lithium-based greases with different consistencies (NLGI grade 1 and NLGI grade 2) and apparent viscosities, CNC particles prepared through sulfuric acid hydrolysis were incorporated at varied concentrations as the lubricating additive [83]. The friction results showed that in the pristine grease with lower consistency (i.e., the sample 421 in Fig. 15), the incorporated CNC could be more effectively transported through the fibrous network and deposited on the sliding surface. It is worth noting that the doping content of CNCs was relatively high (1% to 10%) compared with other nano-additives reported in most studies, because the pristine grease used in this study contained no friction modifier or anti-wear additive, which required greater assistance from the CNC additive. When more CNCs were incorporated (15%), they would strongly influence the viscoelastic behavior of the pristine grease, potentially altering its consistency grade. Since consistency grade is a significant indicator of grease selection across different applications, the concentration of doped CNC should be limited to minimize excessive variation in the grease's basic physical properties.

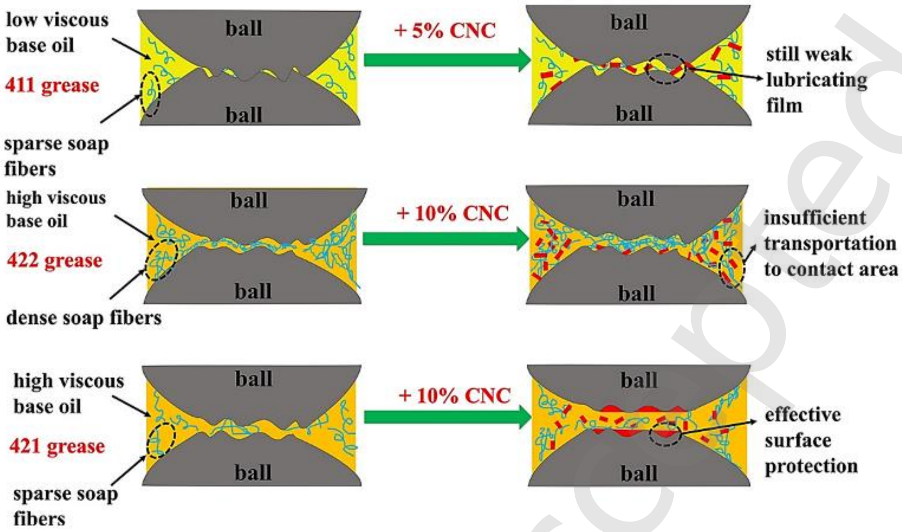

Fig. 15 Schematic of the lubrication mechanisms of CNCs additive in the greases with various base oil viscosities and consistencies. Reproduced with permission from Ref. [83]. © Carbohydrate Polymers, 2022.

3.3.2 Applied as thickeners

Besides serving as a lubricating additive, nanocellulose can also be utilized as a thickener in both polar and non-polar base oils. For non-polar oils, such as PAO, nanocellulose alone typically cannot maintain a consistent grease structure due to its tendency to aggregate [84,85]. Therefore, another component is needed to assist as a stabilizer for the grease. For example, Saini et al. [86] prepared a hybrid grease comprising a PAO base oil, nanoclays, oleic acid, and TEMPO-oxidized CNFs (10 wt.%). Due to their high aspect ratio and abundant surface functional groups, CNFs acted as efficient thickening agents, forming a continuous fibrillar network that enhanced the oil-holding capacity of the grease. Meanwhile, the addition of nanoclays and oleic acid jointly improved the dispersion stability of the system by preventing CNF aggregation and promoting their homogeneous distribution within the grease matrix. Such a hybrid grease exhibited lower COF values and more stable lubrication performance than the grease that only used nanoclays.

In polar oils, nanocellulose exhibits significantly higher thickening efficiency, even when used as a single thickener. Ilyin et al. [87] prepared a biodegradable grease of NLGI grade 3 by incorporating 3 wt.% microfibrillated cellulose (MFC, which can be regarded as a type of CNFs with a wider diameter distribution) into triethyl citrate. However, to obtain a grease with similar rheological properties, it needed to apply 13 wt.% lithium soaps as the thickening agent (Fig. 16). The higher thickening efficiency of MFC was attributed to their thinner microfibrils, which could be bent and form a denser structural network compared to the traditional thickeners of metallic soaps. In addition to its thickening effect, MFC also contributed to lubrication via different mechanisms, depending on its concentration. In small quantities (1%), microfibrils smoothed the friction pairs by filling surface defects, resulting in a considerable reduction in wear and a slight decrease in friction. In large quantities (7 %), microfibrils actively interacted with the steel surface during lapping. Although the strong abrasive action of microfibrils induced relatively high wear during the short running-in time, a tribological film formed thereafter, leading to a 25% reduction in COF.

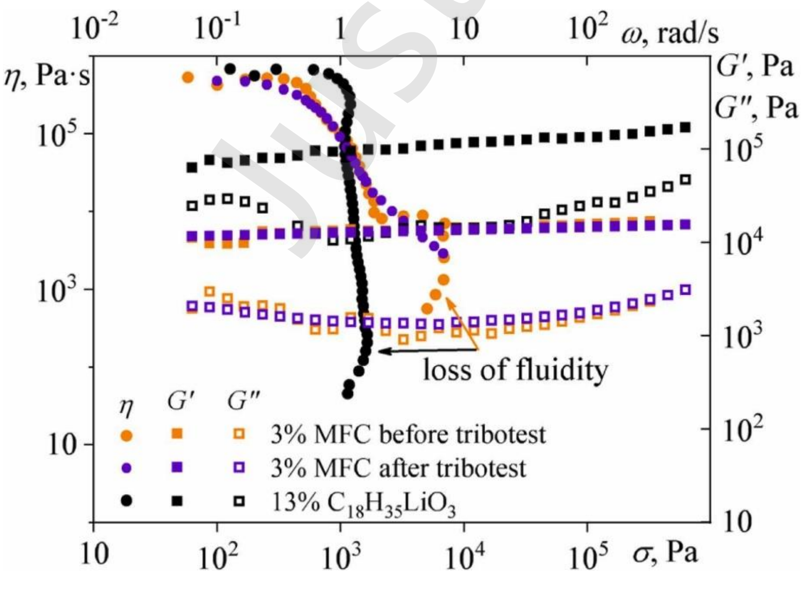

Fig. 16 Effective viscosity, storage, and loss moduli of MFC or lithium-based greases under varied shear stress. Reproduced with permission from Ref. [87]. Tribology International, 2023.

3.4 Lubrication mechanisms of nanocellulose

Based on the literature reviewed above, it is evident that CNCs and CNFs exhibit distinct lubrication mechanisms due to their differences in morphology, rigidity, and the interaction with lubricants. In the boundary and mixed lubrication regimes (Fig. 17), nanocellulose additives mainly function at the solid-liquid interfaces. Specifically, CNC nanorods can transition from isotropic dispersion to liquid-crystalline alignment in lubricants under shear. Depending on the friction pair materials and operating conditions, CNCs can improve lubrication through the micro-rolling effect or the mending effect on the sliding surfaces. In the case of CNFs, flexible fiber layers are more likely to form due to their high aspect ratio, which can protect the substrate and reduce wear.

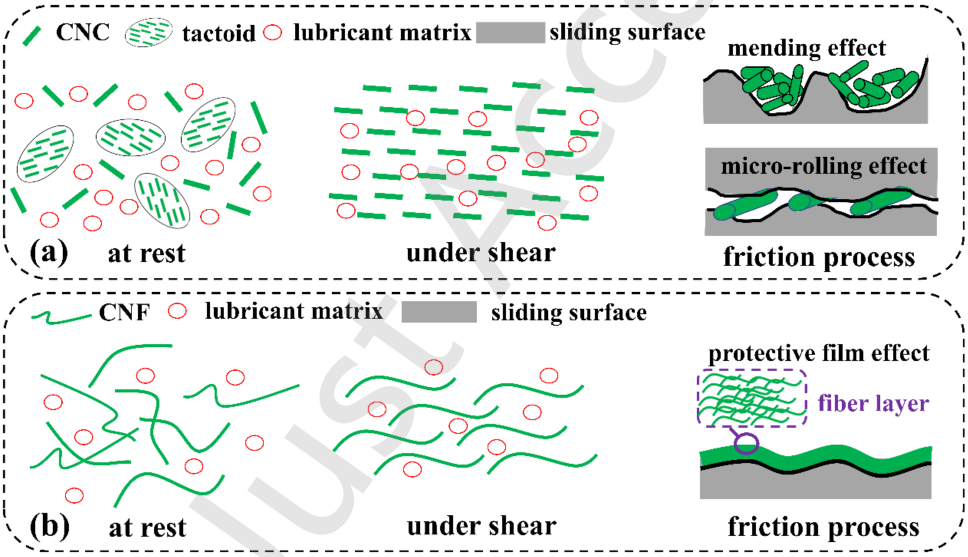

Fig. 17 Schematic diagram of the lubrication mechanisms of CNCs (a) and CNFs (b) in the boundary and mixed lubrication regimes.

In the fluid lubrication regime, nanocellulose primarily affects lubrication by influencing the rheological properties of the lubricant. At high shear rates, the alignment of CNCs or the partial disentanglement of CNFs could effectively lower the apparent viscosity of lubricants, i.e., the shearing resistance. When an electric field is applied, the electrorheological response of nanocellulose could optimally adjust the lubricant viscosity to achieve smart lubrication.

It is important to note that the lubrication mechanisms discussed above are based on a general summary of reported studies. The actual mechanism of nanocellulose under specific conditions, such as different friction pair materials, environmental factors, and lubrication regimes, requires separate analysis. Moreover, many proposed mechanisms remain speculative and lack direct experimental evidence. Therefore, future research should integrate advanced microscopic techniques such as atomic force microscopy and quartz crystal microbalance with simulation methods, including molecular dynamics, to gain deeper insights into the interfacial behavior and dynamic interactions of cellulose nanocrystals and nanofibers in lubricating environments.

4 Conclusions and outlooks

In this paper, the feasibility of using CNCs and CNFs as lubricating additives and thickening agents for lubricants is analyzed with respect to environmental friendliness, physical and chemical properties, and economic considerations. Existing studies have shown that the dispersion stability of nanocellulose in lubricating fluids is critical for their lubricating properties. To enhance their dispersion in aqueous or oil-based systems, Appropriate strategies such as physical methods (e.g., ultrasonication, high-pressure homogenization, mechanical shear) and chemical modifications (e.g., surface charge introduction, acylation, silylation, ionic liquid crystal grafting) should be employed. In lubricating fluids, nanocellulose additives can improve lubrication through a mending effect, micro-rolling, or protective film effect. Moreover, enhanced performance or additional functionality of nanocellulose can be achieved by formulating a hybrid additive with other nanoparticles or by applying an electric field. In grease applications, on the one hand, nanocellulose can be used as an additive to reduce friction and wear. On the other hand, owing to its fibrous structure, nanocellulose can form a three-dimensional network and trap oil molecules, serving as a thickening agent.

Although nanocellulose shows promising application potential in eco-friendly lubrication, several issues remain to be further explored, including understanding lubrication mechanisms and enhancing lubrication performance. Therefore, future research on nanocellulose in lubrication should focus on the following aspects:

1) Considering that lubricating oils remain the most widely used lubricants in the industry, the lipophilic modification of nanocellulose is a key issue that must be addressed for its practical application. Native nanocellulose is highly hydrophilic and poorly dispersible in oil-based systems, requiring the introduction of hydrophobic chains or lipophilic functional groups to improve compatibility. Although various surface modification methods have been proposed, the enhancement of lubricating performance lacks consistent patterns, and systematic comparative studies are still limited. In the future, simulations can be used to investigate interactions between modified nanocellulose and base oil molecules with varying polarities and viscosities, thereby helping predict how the type and grafting density of functional groups influence dispersion behavior and guiding the development of more efficient and targeted modification strategies.

2) Another common issue limiting the widespread use of nano-additives is the generally high price of nanomaterials. Although cellulose is widely available, the cost of nanocellulose in extraction and modification processes still needs to be further reduced through more advanced methods. To enhance the economy and protect the environment, breakthroughs should be made with a focus on sustainable materials and recycling. It is expected to develop strategies to efficiently prepare nanocellulose from agricultural and forestry waste, as well as food processing by-products (e.g., straw, bamboo residue, fruit peels), and establish a resource recycling system, which will promote the integration of nanocellulose production into the recycling economy chain, thereby minimizing the cost of raw materials. In addition, simplified or green chemical modification routes, such as bio-enzyme modification or supercritical carbon dioxide-assisted reactions, need to be explored to reduce the use of harmful solvents and energy consumption.

3) "Eco-friendly" is a general term and needs to be conditionally defined. Specific environmental regulations or policies should be analyzed for various applications. For example, the requirements and specific indicators of ecological friendliness differ between marine and food industry applications. Based on different requirements, the optimal configuration of nanocellulose additives and traditional anti-friction and anti-wear additives (e.g., MoDTP, ZDDP) should be investigated to achieve a balance between environmental friendliness and excellent lubrication performance. Future research should focus on clarifying the green index thresholds and safety standards in different scenarios, and systematically study the compatibility and coupling effect between nanocellulose and other additives. To avoid potential antagonistic effects of the mixed additives, a combination of deep learning and experimental studies should be used to optimize the formulation design of lubricating materials.

4) Smart lubrication is very attractive due to its adaptive and self-adjusting nature to the operating conditions. Future research can focus on exploring the response properties of nanocellulose in smart lubrication. On the one hand, it can be considered to regulate the molecular structure of nanocellulose, which can be designed as an intelligent release carrier with other functional additives sealed inside. When the internal temperature or pressure of the lubricant reaches a predetermined threshold, the nanocellulose will open or disintegrate, thereby releasing the internal sealing functional additives and achieving secondary addition. On the other hand, the introduction of environment-sensitive groups on the surface of nanocellulose, such as temperature- and strain-sensitive groups, can endow it with the ability to respond rapidly to environmental variations.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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