

# TRIBOLOGICAL APPLICATIONS OF PROTIC IONIC LIQUIDS: EFFECTS OF IONIC STRUCTURE, CHALLENGES, AND FUTURE PERSPECTIVES

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## ABSTRACT

Protic ionic liquids (PILs) have emerged as promising candidates for advanced lubrication owing to their intrinsic thermal stability, negligible volatility, and ability to form protective boundary layers at sliding inter faces. This review revealed ionic species present with long alkyl group, or specialised functional group that enhanced intermolecular interaction which tend to improves tribological ability, thermal stability and raise viscosity. Additionally, the compatibility of PIL with base oils is critical for effective additive performance, as insufficient miscibility limits ion transport and interfacial coverage. Hydrolytic stability is another critical concern for application in high-moisture environment which are rarely examined in literature. Collectively, the current findings highlight the need for integrated molecular design strategies that simultaneously consider chain length, ion-pair chemistry, oil compatibility and hydrolytic stability, to realize the full potential of PILs as tunable, high-performance lubricants or additives.

## KEYWORDS

Protic ionic liquids (PILs); lubrication; ion moiety; solubility; hydrolytic stability.

## INTRODUCTION

Due to ongoing advances in materials and the sophistication of machines and technology, lubricants play a crucial role as core components, reducing material loss and improving operational efficiency [1]. Globally, tribological energy losses are substantial, accounting for approximately 20%-30% of the world's primary energy consumption [2–4]. Sazzad et al. estimated that the global market for lubrication products is approximately \$150-\$169 billion per year [5].

The current long-term and heavy reliance on conventional lubricants (i.e., mineral and synthetic oils) poses a significant risk to the environment and public health. Extraction and processing activities of fossil fuels exert pressure on environmental health by producing toxic gases and compounds that contaminate essential biological ecosystems [6]. An ecotoxicology review conducted by Berríos-Rolón et al. shows the widespread presence of carcinogenic compounds (i.e., polycyclic aromatic hydrocarbons) in all fundamental natural resources, of which fossil fuel activities are a major source [7].

Another epidemiology review by Onyije et. al. reveals the elevated risk of contracting cancerous disease, including mesothelioma, lung cancer and leukaemia in oil industry workers and nearby residents [8]. On the other hand, the production of synthetic oils (API Group III-V) does not directly release harmful substances, and the end-product provides extended drain intervals compared to mineral oils. However, the synthesis phase often requires energy-intensive chemical reactions and

heavy metal catalysts, leading to an increase in carbon footprint and a decline in sustainability performance [9–11].

Apart from environmental and toxicity concerns, conventional lubricants exhibit several limitations when operating under demanding conditions. These typically include limited thermal stability, susceptibility to oxidative degradation, poor viscosity-temperature characteristic, inadequate boundary lubrication, and challenges in additive solubility and compatibility [12–14]. Therefore, various specialised additives are introduced into the base oil to address each additive's inherent limitations [15, 16].

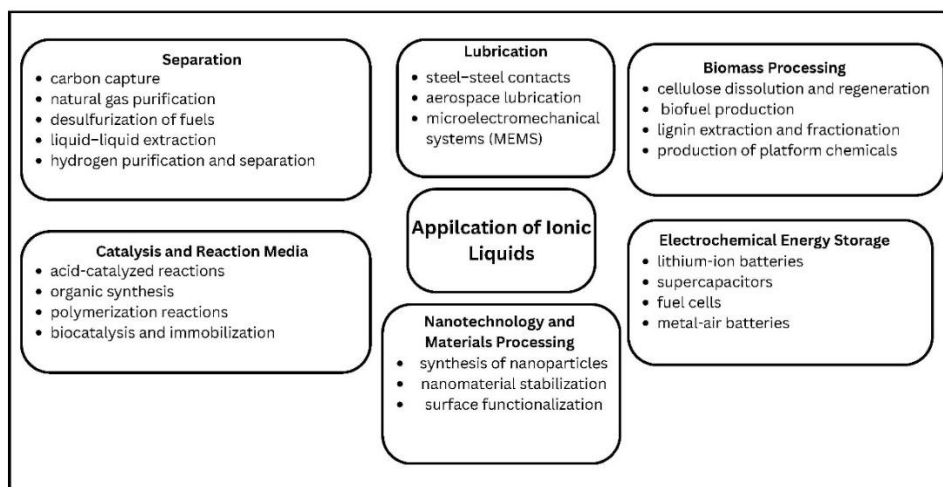
Recent studies show that elevated SAPS contents in conventional lubricant additives have been associated with detrimental effects on engine components and adverse health outcomes. [17–19]. Sulphated ash, originating from metal-based detergents, such as calcium or magnesium sulfonates, clogs the exhaust filter as they are inorganic and non-combustible materials that cannot be burned off during “regeneration” in the diesel particulate filter (DPF) [20–22]. Phosphorus in Zinc dialkyldithiophosphate (ZDDP) coats the catalytic converter metals when engine oil burns or mist escapes through the positive crankcase ventilation (PCV) system, hindering heterogeneous catalysis, leading to increased emissions and accelerated degradation of the emission system [23]. Sulphur present in extreme-pressure (EP) agents and certain antioxidants produces sulfuric acid when burned and combined with moisture, resulting in internal engine corrosion [24]. In a nutshell, long-term reliance on conventional lubricants is unsustainable, and developing alternatives is necessary.

The recent emergence of Protic ionic liquids (PILs) has shown high potential to replace conventional oil blends as both a neat lubricant and additives [25]. The potential of PILs has been actively studied, as they possess unique multifunctional properties and offer additional sustainability [25, 26]. Various studies on PIL have shown enhanced or superior tribological performance as both a neat lubricant and an additive compared to standard oil [26–29]. However, there is a lack of information on the structure–performance relationships. Therefore, in this paper, the influence of the ion moiety and the major challenges are summarised and reviewed to identify future research prospects for the application of PILs as lubricant alternatives.

Unlike previous reviews that broadly cover ionic liquids, this work focuses specifically on PILs, emphasising structure–tribology relationships, solubility limitations, hydrolytic stability, thermal stability and corrosivity—an area that remains underexplored despite its importance in real-world lubrication systems.

### Fundamentals of Protic Ionic Liquids

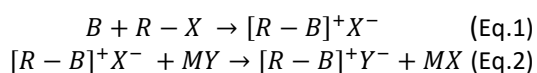
Protic ionic liquid is a subset of ionic liquid (IL), typically defined as compounds composed of entire ions that are in a liquid state below 100 °C. Their unique characteristics, including low volatility, excellent thermal durability, high-voltage stability window, high ion diffusivity, simple preparation, and tunability, contribute to their practicality as a compound for wide-ranging applications (see Figure 1) [30–32].



**Figure 1:** Major applications of ionic liquids, including lubrication, catalysis, biomass processing, energy storage, and carbon capture, summarized from previous studies [33–49].

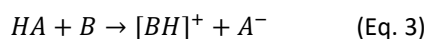
ILs can be classified by proticity into protic ionic liquid (PIL) and aprotic ionic liquid (APIL), distinguished by the presence of transferable proton ( $H^+$ ) on the cation of PIL [50]. Beyond their ability to form hydrogen-bonded networks, PIL differ from APIL in their synthesis methods, which represent another major distinction between the two classes.

Methods of APILs synthesis include direct alkylation of symmetrical Lewis Bases or, more commonly, a two-step reaction involving quaternization to form an organic halide salt, followed by an anion-exchange reaction (metathesis or acid-base reaction) to achieve the desired physical and chemical properties [50–52]. The equations below represent a general example of APIL synthesis via the typical quaternization (Eq. 1) and metathesis (Eq. 2) route:



where B denotes a base (e.g., amine, imidazole, pyridine), R-X represents an alkyl halide in which R is an alkyl group and X is a halide (Cl, Br, or I),  $[R-B]^+$  is the quaternized cation,  $X^-$  is a halide anion,  $Y^-$  is the desired ionic liquid anion, and  $M^+$  is a metal cation (e.g.,  $K^+$ ,  $Na^+$ ,  $Ag^+$ ).

On the other hand, PILs offered a simpler synthesis pathway, requiring only a stoichiometric neutralisation between a Brønsted acid and a Brønsted base (Eq. 3).



where HA represents a Brønsted acid, B denotes a Brønsted base,  $[BH]^+$  is the protonated base forming the cation, and  $A^-$  is the conjugate base forming the anion.

While providing simplicity and cost-effectiveness in production, their structural simplicity also provides endless opportunities to develop a “tailor-made” formulation for purpose-oriented application. Variation in ionic composition as a function of ion size, identity, and structural configuration can adjust physical and chemical properties [26]. Commonly, the cation is formed from amines, heterocycles, or other basic organic molecules, while the anion can range from inorganic species to organic sulfonates or carboxylates. Figure 2 illustrates some of the common ions of PILs [33, 53].

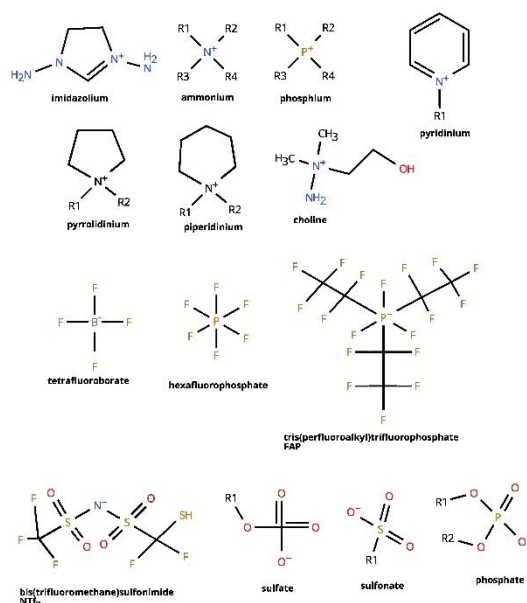


Figure 2: Common cations and anions apply in PILs.

### Influence of Ion Size

A recurring theme in the literature is that molecular size and alkyl chain length strongly modulate lubrication performance through three coupled effects: (i) changes in bulk viscosity and polarity; (ii) altered surface adsorption/ordering and boundary-layer structure; and (iii) differences in tribochemical reactivity at the contact interface [62–64]. Experimentally, several papers that systematically vary chain length report that short additions ( $\approx 1$  wt.%) of PILs to conventional base oils can substantially reduce the coefficient of friction (COF) and wear volume.

Amzad et al. [54] studied four lauryl sulphate-based anion ILs as 1.5 wt.% additives in hydrocarbon mineral oil (SN 150) and polyethylene glycol (PEG 200) base oil under steel contact and observed consistent COF reductions. The best-performing variant (long-chain ammonium anion) reduced COF, WSD, and wear volume by 28%, 48%, and 91%, respectively, relative to PEG-200. In contrast, similar studies by Gusain et al. [55] observed marked degradation in friction-reducing and anti-wear properties with a longer fatty-acid-based anion coupled with a tetrabutylammonium cation pair as additive at 1 wt% in SN-150. The increased internal friction is suggested to be driven by the stronger van der Waals interaction of longer alkyl chain length and correlates positively to viscosity of the ILs.

Guo et al. [56] investigated the effect of ammonium-based cation size paired with an identical anion, both a neat lubricant and as an

additive (1-2 wt%) in BIOTELEX 46 base oil under steel contact. IL with the smallest cation achieved the highest wear reduction, whereas the largest provided comparable performance only when used as an additive in base oil. The corresponding EDX spectrum revealed tribocorrosive properties associated with ILs containing larger cations (high conductivity), and this effect was significantly mitigated when these ILs were introduced in small amounts as additives. Guo et al. [57] further this study by replacing the previous anion with two sulfonate-based anions with distinct alkyl chains. Anions with longer hydrocarbon group length show a tendency to form a protective tribolayer with enhanced tribology performance and a narrower carbon-oxygen-rich wear track when introduced to base oil at 1 wt%.

A straightforward comparison between two fatty acid anions (C1 and C5) IL by Vega et al. [58] supports the idea that larger ions are in favour of tribofilm formation. Demonstrated by negligible material transfer between the aluminium-steel tribopair and achieving COF and wear comparable to the tested commercial lubricant. Minami et al. [59] explored the potential of trifluorotris(pentafluoroethyl) phosphate (FAP) anion, known for its superior hydrophobicity and hydrolytic stability, as a neat lubricant under steel contact with various cations. Similarly, the cation with a longer alkyl chain showed superior wear reduction.

Kaisy et al. [60] studied the performance of a trioctylammonium-based cation in contact with multiple carboxylate-based anions of various structures, providing another useful insight into the structure-tribological relationship. Anions with compact, benzene-based and hydroxyl structures demonstrated superior performance, whereas those with extended alkyl chains or mixed structural features exhibited reduced effectiveness. The study suggested that enhancements are primarily driven by  $\pi$ - $\pi$  intermolecular interactions within the benzene ring, enabling denser packing of the ordered ion-layered protective film. The best-performing salicylate anion improves further due to the additional hydroxyl group, which elevates polarity and aids adsorption.

A study by Fang et al. [61] provides another intuitive insight by controlling anion chain length, with the mechanism deeply explored through MD simulation in LAMMPS. Both COF and wear volume reduce with a shorter alkyl chain in the anion. The combined results suggested that compact ions allow denser ion packing at the interface, thereby providing better lubrication.

In summary, adjusting alkyl chain length or ion structure is a key lever for tuning the lubricating properties of protic ionic liquids (PILs). Larger ions generally enhance frictional performance, thermal stability, and viscosity when used either as neat lubricants or as additives. However, these effects are still strongly dependent on the intrinsic molecular structure and the ionic species, which are ultimately governed by intermolecular interactions. A comparative summary of these effects is presented in Table 1.

## Challenges and Future Recommendations:

### Thermal Oxidative Stability

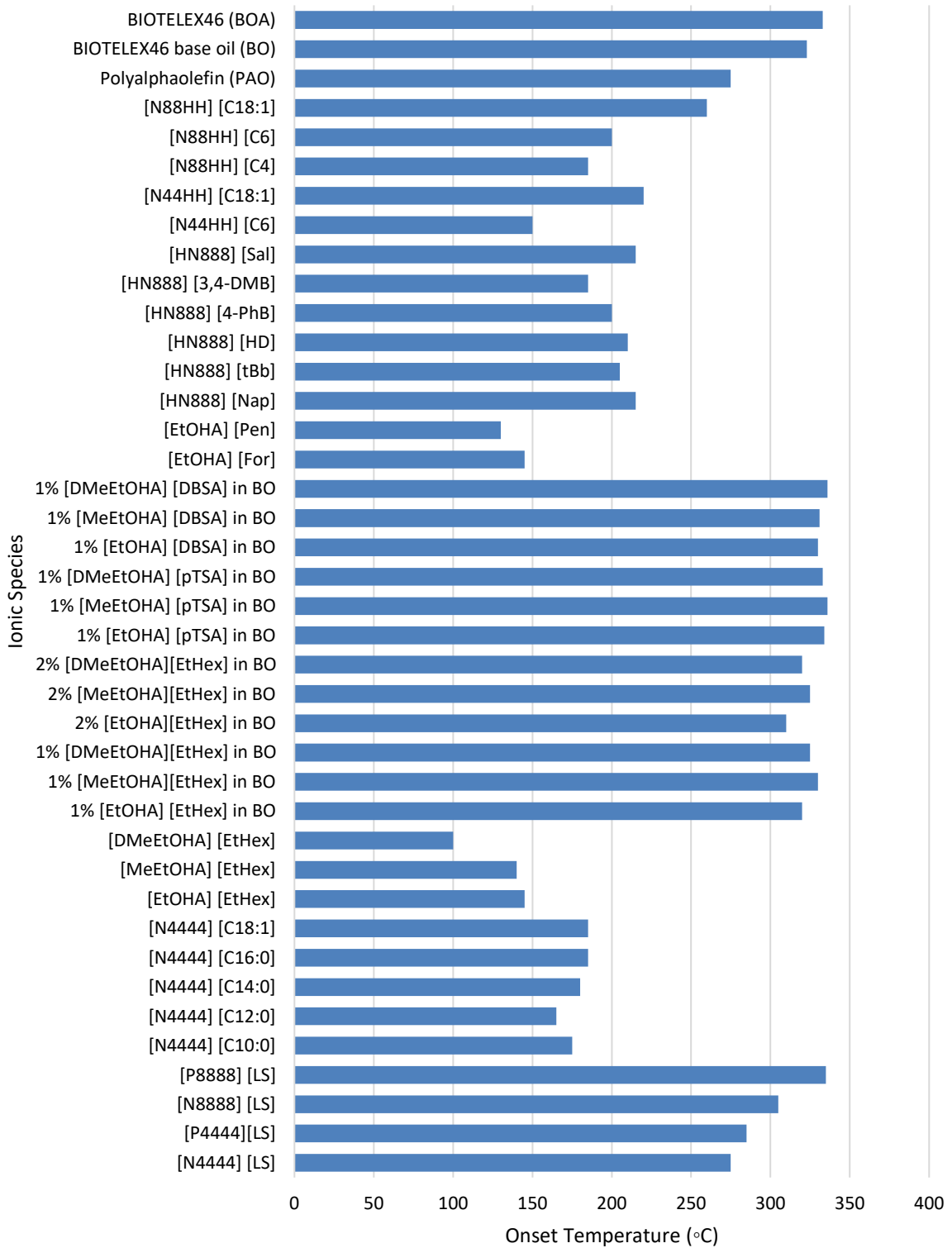
Beyond tribological performance, thermal-oxidative stability is a critical parameter, as it directly dictates a lubricant's long-term usability and durability. A significant portion of the reviewed literature suggests that increasing ionic volume, often achieved through longer molecular chains or bulkier substituents, correlates with enhanced thermal-oxidative resistance. This stability is typically quantified using the onset temperature of oxidation ( $T_{\text{onset}}$ ) via Thermogravimetric Analysis (TGA).  $T_{\text{onset}}$  serves as a benchmark for the initiation of thermal degradation and is determined by the intersection of the extrapolated pre-decomposition baseline and the tangent to the curve at the point of maximum mass loss rate [64].

While TGA is typically included in tribology literature, a common issue is the lack of comparison with a standard lubricant stock. Figure 4 summarises the  $T_{\text{onset}}$  data from publications reviewed in Table 1 to better understand the trend. The result aligns with the present review's previous statement that larger ions tend to improve thermal-oxidative stability, with only the experimental results of Guo et al. [56] and Vega et al. [58] showing an exception.

However, the IL content in  $T_{\text{onset}}$  is lower than that in the standard lubricant (see Figure 4). Among the listed commercial lubricants, their thermal stability is in the order of BOA > BO > PAO. Of 37 ILs, only 41% exhibited greater stability than PAO. The percentage reduced to 27% and 11% when compared with the non-formulated and formulated base oil, respectively. Notably, pure ILs tested with high thermal stability were only lauryl sulphate anion-based ILs from Amzad et al. [54] (refer to Figure 5), whereas the others were presented in additive form in BO as presented in [56].

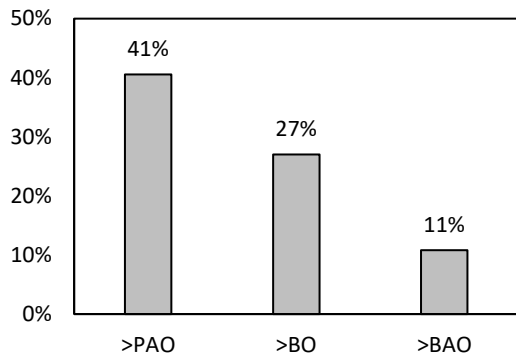
**Table 1: Summary for ionic liquid properties**

Ref	Author and Year	Ionic Species	Independent Variable	Application	TriboPair-Material	Properties with increasing ion size		
						Thermal Stability	Viscosity at 40°C	Tribological
[54]	Amzad et al., 2020	lauryl sulphate anion, ammonium and phosphonium-based cation	Cation chain length	Additive (1.5 wt% in PEG-200 and SN-150)	100Cr6 steel ball and 316LN stainless steel disc	Improve	Higher	Improve
[55]	Gusain et al., 2019	Tetrabutylammonium cation and fatty acid-based anion	Anion chain length	Additive (1 wt% in SN-150)	100Cr6 steel ball and 6061-T6 aluminium alloy disc	Improve	Higher	Degrade
[56]	Guo et al., 2019	Ammonium-based cation and carboxylate-based anion	Cation size	Additive (1 wt% and 2 wt% in BIOTELEX 46 base oil) and neat lubricant	AISI52100 steel ball and disc	Degrade as a neat lubricant, but showed comparable performance when used as an additive	Lower	Improve at 1 wt%, but degraded at 2 wt% and when used as a neat lubricant
[57]	Guo et al., 2023	Ammonium-based cations and sulfonate-based anions	Anion size and cation chain length	Additive (1 wt% in biodegradable polar base oil)	Steel ball and 2024 aluminium alloy flat	Improve	Lower	Improve
[58]	Vega et al., 2017	Ammonium-based cation and carboxylate-based anion	Anion chain length	Neat lubricant	Aluminium 1100 plate and AISI 1050 steel ball	Degrade	Increase	Improve
[59]	Minami et al., 2008	Trifluorotris (pentafluoroethyl) phosphate-based anion	Cation type and size	Neat lubricant	SUJ2 (JIS) ball and flat	NA	Increase	Improve
[60]	Kaisy et al., 2021	Trioctylammonium-based cation	Anion structure (benzene and alkyl)	Neat lubricant	AISI 52100 steel ball (four-ball tester)	Significant trend not observed	Higher viscosity when presented with benzene ring and hydroxyl group anion, lower in carboxylate-based long chain anion	Anions with benzene ring showed superior lubricating performance as opposed to alkyl chain carboxylate anion
[61]	Fang et al., 2022	ammonium-based cation and carboxylate-based anion	Anion chain length	Additive (2 wt% in PAO 10)	AISI 52100 steel ball and disc	Improve	Higher	Degrade

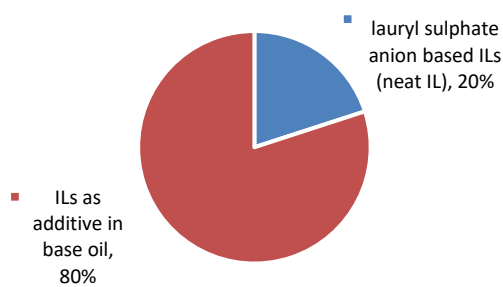


**Figure 5:** Overall comparison on thermal stability of ionic species, ionic liquid additised base lubricant, commercial lubricant (BOA), and commercial base oil (BO, PA)

This behaviour might be attributed to the equilibrium nature of the proton transfer reaction, in which the PIL can undergo back-proton transfer, dissociating into its neutral acid and base precursors. These molecular precursors typically possess significantly higher vapour pressures than the parent ionic species, facilitating mass loss via evaporation and subsequent thermal degradation under prolonged thermal stress [65].



**Figure 4:** Summary of comparison of ionic liquids with standard lubricant.



**Figure 5:** Proportion of ionic liquids exhibiting higher thermal-oxidative stability than PAO.

### Solubility, Hydrolytic Stability and Corrosivity

Results from applying PIL as a lubricant additive typically demonstrate superior properties compared to conventional oils. However, insolubility between the PIL and the base oil is very common. Viesca et al. [66] studied the miscibility of six hexanoate- and sulfonate-based PILs in mineral oil, ester and polyalphaolefin (PAO) by 1 h visual monitoring for physical separation, followed by a subsequent 12 h multiple light scattering measurement in the Turbiscan station. The results show that only one formulation exhibited appreciable miscibility with ester oil. Guo et al. [57, 67] extensively investigated the tribological properties of various PIL as lubricant additives, with few reporting any signs of insolubility.

Mixture stability was investigated by visual inspection at 24 h, 168 h, and 720 h after a 6500 rpm, 10 min centrifugation step upon complete mixing. Visible phase separation was observed at the first 24 h in both mineral oil and polar biodegradable base oil incorporated with PIL.

PILs generally exhibit poor solubility due to their strong hydrogen-bonding networks and ionic interactions, which hinder their compatibility with apolar environments [68, 69]. However, certain structural or molecular modifications, such as the presence of long alkyl groups and fatty acid anions, can improve miscibility with base oil, especially in non-polar oils, by reducing hydrogen bonding, charge delocalisation and increasing apolar domain, enhancing compatibility with oil molecules [70, 71]. For example, PILs with alkylammonium cations and nitrate anions show better solubility for hydrocarbons compared to those with hydrogen sulphate anions, as the latter exhibited a stronger hydrogen bonding network [69]. Apart from structural modification, solubility in oils can be enhanced by combining PILs with complementary properties (i.e., good solubility with poor solubility), creating a binary system referred to as a binary ionic liquid (BIL). Fang et al. [72] synthesised two BILs composed of oil-insoluble and oil-soluble PILs, and achieved a 1-2 wt% solubility in PAO.

Apart from oil solubility, the hydrolytic stability of PIL is another underexplored physicochemical property. Most experiments evaluate thermal-oxidation stability, biodegradability, toxicity, tribology behaviour, but often overlook the potential of the water-ingress system in real-life applications [73–75]. It is noted by Minami et al. [59] that their study on hydrophobic FAP anions is driven by the hydrolytic instability of the conventional hydrophilic BF<sub>4</sub> and PF<sub>6</sub>-derived anions and would undergo hydrolysis to produce highly toxic and corrosive hydrogen fluoride.

Steudte et al. [76] exposed six PILs to buffer solutions at different pH values to investigate their hydrolytic behaviour. High-Performance Liquid Chromatography (HPLC) and Ion Chromatography (IC) were employed to monitor the decrease in the concentration of ionic liquid anions, while the formation of degradation products was identified by liquid chromatography–mass spectrometry (LC–MS). The results show that dicyanamide and tricyanmethanide undergo hydrolysis to form amides, acids, and nitrogen-containing compounds. This finding is particularly significant, as most lubrication systems exhibit a gradual increase in acidity over their service life due to the formation of oxidative by-products [77]. Consequently, protic ionic liquid (PIL) additives

must demonstrate strong resistance to hydrolysis under acidic conditions, as susceptibility to such degradation could further accelerate lubricant deterioration and compromise system performance.

Protic ionic liquids (PILs) also present notable challenges in terms of corrosivity, primarily due to their intrinsic Brønsted acidity, residual water content, and the presence of equilibrium molecular species arising from incomplete proton transfer. These factors can promote metal corrosion and accelerate material degradation, particularly under elevated temperatures and in open systems. Furthermore, thermal exposure may shift the acid–base equilibrium and generate reactive species (e.g., amides or free acids), exacerbating corrosive behaviour [65]. To mitigate these issues, careful selection of cation–anion combinations with higher  $\Delta pK_a$  (i.e., greater ionicity), rigorous control of water and impurity levels, and operation under sealed or inert conditions are recommended. Additionally, incorporating corrosion inhibitors or using corrosion-resistant materials can further enhance the practical applicability of PIL-based systems in industrial environments.

## CONCLUSION

In conclusion, the potential of applying Protic Ionic Liquids (PILs) in lubrication systems has been widely explored; however, significant challenges remain for their industrial implementation. PILs offer a broad spectrum of functionalities and distinct advantages over conventional lubricants and additives, yet the optimal formulation for specific applications has not been fully established.

Ionic liquids with larger or longer molecular functional groups generally exhibit enhanced thermal stability and increased viscosity, which can influence frictional behaviour. Nevertheless, their overall performance remains highly dependent on the precise chemical structure, which governs intermolecular interactions and surface affinity.

Thermal stability remains critical in the ionic system as the majority of the neat ILs exhibited poor performance relative to commercial lubricants. Applying ILs as oil additives significantly mitigates the issue.

Solubility remains a major limitation for the use of PILs as lubricant additives. Their ionic nature and extensive hydrogen-bonding networks promote strong self-association, leading to poor compatibility with nonpolar base oils and formulation difficulties. In addition, hydrolytic

stability represents a critical but overlooked failure pathway in real lubrication systems, particularly under humid or hydrogen-rich environments, and should be treated as a primary design constraint in future PIL development. In addition, the potential corrosivity of protic ionic liquids, arising from their Brønsted acidity, water content and equilibrium molecular species, should be carefully considered for practical applications, particularly under prolonged thermal exposure, where compositional changes may occur

Future research should therefore focus on the design and evaluation of advanced PIL systems, including binary protic ionic liquids (BILs), with particular emphasis on improving solubility, examining hydrolytic and thermal stability, and carefully selecting ionic pairs. Incorporating these considerations into lubricant formulation strategies will be essential for advancing PIL technology toward viable industrial applications.

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