Research Progress of Ionic Liquids as Lubricants

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Cite This: ACS Omega 2021, 6, 29345−29349

ABSTRACT: Emerging ionic liquid lubricants were discovered only 20 years ago. The superior performance of ionic liquids over traditional lubricating fluids have been reviewed, including low-temperature fluidity, viscosity—temperature properties, thermal oxidation stability, ultralow volatility, incompressibility, electrical conductivity, friction coefficient under elastohydrodynamic lubrication conditions, friction reduction and antiwear performance under boundary lubrication conditions, environmental friendliness, etc. The applications where ionic liquids are superior to traditional lubricant fluids are presented, including hydrogen compressor lubricating fluids and liquid pistons, oxygen compressor lubricating fluids, hydraulic fluids, space lubricants, vehicle engine oils, industrial gear oils, metalworking fluids, industrial coolant, micro/nano electromechanical system applications, electrical conductive lubricants, etc. The ability of ionic liquids to replace ZDDP, a key additive of lubricating oil, is introduced. The future development prospects of ionic liquid lubricants are analyzed.

1. INTRODUCTION

Ionic compounds/salts tend to have high melting points, for example, sodium chloride melts at 801 °C. In 1888, Gabriel and Weiner discovered a low melting point salt (HOC2H5)NH3+NO3− (52−55 °C), and in 1914, Walden discovered another low melting point salt (C2H5)NH3+NO3− (12 °C). Today, ionic liquids usually refer to molten salts with a melting point below 100 °C; the arbitrary temperature division does not have a specific meaning. These ionic liquids all have organic cations whose large organic structure can disperse charges, reduce electrostatic attraction, and lower the melting point of ionic liquids. Some structures of cations and anions of common ionic liquids are shown in Figure 1.

In 2001, Ye et al. observed that two ionic liquids [C6C1im][BF4] and [C6C2im][BF4] have excellent lubricity in various friction systems. Compared with the aviation lubricant X-1P and the aerospace lubricant PFPE, they win out with a lower coefficient of friction, lower wear, and much higher load-carrying capacity. Moreover, they have excellent low-temperature fluidity (pour point below −55 °C) and can withstand high temperatures of 320 °C. Since then, ionic liquid lubricants have attracted a lot of research interest. So far, more than 400 ionic liquids have been investigated for tribological performance in various friction systems.

2. PERFORMANCE ADVANTAGES

There are many types of traditional lubricating fluids, including petroleum-based mineral oil, synthetic hydrocarbons, polyol esters (POE), polyalkylene glycol (PAG), polysiloxanes, perfluoropolyethers (PFPE), and other organic compounds. Each type of traditional lubricating fluid has one or more advantages. Nowadays, ionic liquids are appearing in a subversive manner, challenging the best lubricating fluids in almost every performance, including low-temperature fluidity, viscosity—temperature properties, thermal oxidation stability, ultralow volatility, incompressibility, electrical conductivity, elastohydrodynamic (EHD) lubricity, boundary lubricicity, environmental friendliness, etc.

A good lubricating fluid needs a variety of performances to meet the needs of various applications, such as low volatility to reduce risk to users and consumption of lubricants; better low-temperature fluidity for operation in cold climates and being handled; higher viscosity index to ensure that the lubricating film is thick enough at high temperatures to avoid dry friction; stronger interfacial adsorption to facilitate lower friction coefficients under EHD and boundary lubrication conditions; higher reactivity and lubrication film generation capacity for friction-reduction and antiwear performance under boundary lubrication conditions; higher thermal and oxidation stability to delay the degradation of lubricants for extended service life; lower corrosion to be compatible with relevant materials; better environmental friendliness to protect human and environment for sustainable development.

2.1. Low-Temperature Fluidity. Many ultralow temperature ionic liquids have been found, some of them do not freeze even at −100 °C, as shown in Table 1. The relationship...
Impurities also have an effect on viscosity. For example, the order of viscosity from small to large is $[\text{im}]^+ < [\text{py}]^+ < [\text{pyr}]^+ < [\text{ox}]^+ < [\text{pip}]^+ < [\text{mo}]^+$, phosphonium $<$ ammonium, and $[\text{DCA}]^-$.

The types of cations and anions affect the viscosity of the ionic liquid; and the greater the number of branches, the greater the viscosity; alkyl $-$OR or ether $-$OR produces lower viscosity than any other substituent.

Viscosity index is an indication of viscosity-temperature dependence. A higher viscosity index means that the viscosity is less susceptible to temperature and the lubricant is more reliable, and vice versa. Ionic liquids have a wide range of viscosity indexes, from less than 0 to more than 250. The relationship between the structure of ionic liquids and their viscosity index is still unclear. Relevant studies are lacking. Some ionic liquids have viscosity indexes that are comparable to that of polyl lubricants, and higher than that of polyyl ester (POE), poly(olefin) (PAO) and hydrocarbon oils.

2.3. Volatility. There are few data points on the vapor pressure of ionic liquids. The reason is that the vapor pressure of ionic liquids is difficult to measure at room temperature. Many ionic liquids have even lower vapor pressures than the best space lubricants. With $[\text{C}_{4}\text{C}_{1}\text{im}]^+[[\text{NTf}_2^-]]$ ionic liquids taken as an example, their vapor pressure is thousands of times lower than that of poly($\alpha$-olefin), as shown in Table 2. Some ionic liquids have relatively higher vapor pressure, such as $[\text{C}_{4}\text{C}_{1}\text{im}]^+[[\text{DCA}]]$ and $[\text{C}_{6}\text{C}_{1}\text{im}]^+[[\text{PF}_6^-]]$, even so they both have vapor pressures $<10^{-18}$ Pa at 298 K.

2.4. Thermal Oxidation Stability. The thermal oxidation stability of many ionic liquids is far better than that of traditional lubricating fluids such as poly($\alpha$-olefin)/s, polyol esters, polysiloxanes, and polyalkylene glycol (PAG), and even significantly better than that of perfluoropolyethers. Containing $[\text{NTf}_2^-]$, $[\text{OAc}^-]$, Methide, $\text{TO}^+$, FAP, $[\text{PF}_6^-]$, and $[\text{BF}_4^-]$ anions often means high thermal oxidation stability. On the contrary, containing dicyanamide, nitrate, acetate, and halide anions means low thermal oxidation stability. The influence of cations on thermal oxidation stability is far less than that of anions. Generally, imidazolium has the highest thermal oxidation stability among the cations.

2.5. Incompressibility. The hydraulic oil or compressor oil used in the high-pressure system should be incompressible. The incompressibility of most ionic liquids is better than that of traditional hydraulic oils such as mineral oil, polyalkylene glycol (PAG), and vegetable oil. The incompressibility of some ionic liquids is even better than that of water which is difficult to compress.

2.6. Electrical Conductivity. Due to their ionic nature, the electrical conductivity of ionic liquids is much better than that of traditional lubricants.
2.7. Friction Coefficient under Elastohydrodynamic (EHD) Lubrication Conditions. Under elastohydrodynamic (EHD) lubrication conditions, the friction coefficient of ionic liquids is lower than that of synthetic esters, polyalkylene glycol (PAG), polyalphaolefins, Group I base oil, Group II base oil, Group III base oil, traction base fluid, polysisobutylene, perfluoropolyether, and vegetable oils. The EHD lubricity of ionic liquid is better than that of traditional lubricating fluids.

2.8. Friction Reduction and Antiwear Performance under Boundary Lubrication Conditions. Under boundary friction conditions, whether in steel–steel friction systems or in steel–aluminum friction systems, or in other friction systems, various kinds of tested ionic liquids exhibited superior boundary lubricity to traditional lubricating fluids, that include not only polyalphaolefins, polyol esters, X-1P, and perfluoropolyethers, but also ZDDP-added mineral oil, fully formulated gear oil, fully formulated engine oil, and commercial hydraulic oil.

It is widely believed that the reason ionic liquids have the lower EHD friction coefficient, as well as the more outstanding boundary friction reduction and antwear performance than traditional lubricating fluids, is that the charge makes the anion and cation adsorb firmly on the friction interface. With the strong Coulometic interactions between ions and surfaces, the boundary film of ILs can resist the squeeze-out even at higher pressures than similar films of uncharged molecules and show much lower friction coefficients than nonpolar molecular liquids, such as hydrocarbon lubricants. By applying and changing interface potential, the composition and structure of the IL boundary films can be tuned, resulting in either increased or decreased friction coefficients. This opens completely new pathways for lubricant design.

2.9. Corrosion. Ionic liquids containing [BF₄] or [PF₆] anions can hydrolyze corrosive products in humid environments, and other halogen-containing anions, such as [TNF₆], [FAP], and TIO anions, have no hydrolytic corrosion problems. At present, a large number of ionic liquids that are noncorrosive to metals used in friction system have been synthesized.

2.10. Environmental Friendliness. By simply combining the anions and cations of different environmentally friendly raw materials, an environmentally friendly ionic liquid is obtained. These raw materials are often cheap foods, medicines, and low-toxicity compounds, such as oleic acid in edible vegetable oils, the nutrients choline and amino acids, the sweetener sodium saccharinate, the sweetener potassium aceturate, the laxative phosphoric acid, saccharinate, the sweetener potassium acesulfamate, the laxative

3. APPLICATION ADVANTAGES

Different application scenarios have different requirements for lubricating fluid performance. With excellent performance, ionic liquids outperform traditional lubricants in the following application scenarios.

3.1. Hydrogen Compressor Lubricating Fluid and Liquid Piston. Kermani proposed the performance requirements for the liquid piston and lubricant of hydrogen compressor and screened out 5 ionic liquids. Among them, [C₆C₁im][CF₃SO₂], [C₆C₁im][NTF₂], and [N₁₁₄][NTF₂] are suitable for use as liquid pistons for reciprocating hydrogen compressors; [P₆₆₆₆₆₆][NTF₂] and [N₁₈₈₈][NTF₂] are suitable for use as lubricating fluid for various types of hydrogen compressors. [C₆C₁im][NTF₂] was selected as the best liquid piston for hydrogen compressors.

Linde successfully commercialized ionic liquid lubricants. Relying on the technical advantages of ionic liquid compressors, the world’s first fuel cell passenger train hydrogen refueling station was built, and hydrogen refueling facilities were provided for the world’s first fuel cell marine ship. The Linde ionic liquid compressor uses ionic liquid as piston, lubricating fluid, and coolant. The market share of this technology for 70 MPa hydrogen fueling stations is >50%, so it can be labeled as state-of-the-art. This is due to the negligible volatility of ionic liquids, which avoid the degradation of hydrogen purity and fuel cell performance caused by the volatilization of traditional lubricating oil. At the end of 2020, there were 432 hydrogen refueling stations worldwide, nearly 200 hydrogen refueling stations used Linde technology.

3.2. Oxygen Compressor Lubricating Fluid. High-pressure pure oxygen has extremely high reactivity with traditional lubricating oil. In order to prevent explosion, the labyrinth piston oxygen compressor works in an oil-free manner, which is not only more expensive than an ordinary compressor, but also has higher wear due to the lack of lubrication. Its sealing effect is not as good as oil sealing, resulting in lower compression efficiency.

To use ionic liquids as lubricant for pure oxygen compressors, Predel et al. screened out ionic liquids through a series of performance tests, including 96 h 120 °C metal corrosion test, compressibility test, 5 × 72 h high-temperature high-pressure pure oxygen reactivity test, oxygen solubility investigation, and tribological test under high temperature and high load conditions. With ionic liquid lubrication, the pure oxygen compressor worked for 3000 h with pressure to 30 bar and displacement to 200 N m³/h. Temperature, pressure, and exhaust gas monitoring proved that the ionic liquid lubricated pure oxygen compressor is safe and reliable.

3.3. Hydraulic Fluid. Compared with traditional hydraulic oil, ionic liquids have the advantage of incompressibility, which is actually manifested as a significant improvement in hydraulic efficiency and an improvement in hydraulic response sensitivity. In addition, the metal corrosivity of ionic liquids is significantly better than that of mineral hydraulic oils; their antiwear performance, bearing capacity, and viscosity index are all better than mineral hydraulic oils. Ionic liquid hydraulic fluids are readily available on the market, and pilot projects using ionic liquids as flame-retardant hydraulic oils have been implemented.

3.4. Space Lubricant. The existing space lubricants are not flawless; the issues involve the boundary lubricity of silicone fluids, the volatility of synthetic ester oils and ultrarefinned mineral oils in vacuum, the boundary lubricity and degradation of perfluoropolyether (PFPE), low-temperature fluidity and load-bearing capacity of polyalkylcyclpentane (MAC), and so on. Ionic liquids are expected to be the next generation of space lubricants.

Okaniwa et al. screened out an ionic liquid whose low-temperature fluidity is equivalent to that of PFPE and better than that of MAC. The friction-reduction and antwear performance of the ionic liquid surpass MAC and PFPE, and it can resist the radiation in the geosynchronous orbit for 100 years. Ionic liquid [C₆C₁im][NTF₂] outperforms space lubricant MAC in many aspects, such as thermal oxidation stability, nonvolatility under high vacuum and heat conditions, friction-...
reduction and antiewear to steel–steel contact under high vacuum conditions, resistance to the radiation of atomic oxygen, UV rays and high-energy protons, etc. The service lifetime of an ionic liquid \([C_{6}C_{1}pyr][NTf_{2}]\) is at least 23 times longer than that of space lubricants; in addition, its vacuum volatility, corrosion resistance, friction reduction, and antiewear properties all surpassed those of PFPE space lubricant. The vapor pressure of ionic liquid \([C_{6}C_{1}im][C_{5}SO_{2}]\) is as low as that of space lubricants, and it has a lower coefficient of friction and a longer service life.

3.5. Automotive Engine Oil and Industrial Gear Oil. Compared with fully formulated engine oils, ionic liquids \([C_{6}C_{1}im][NTf_{2}], [C_{6}C_{2}im][NTf_{2}], \text{ and } [C_{6}H_{17}]NH.NTF_{2}\) have a series of performance advantages, such as lower friction coefficient, less wear, and higher decomposition temperature. Not only can the engine be better protected, but the service life of lubricating fluid will also be greatly extended. No need for ZDDP addition means that the ionic liquid has a lower ash content, which is more beneficial to the automobile exhaust treatment system. Theoretically, the benefits of replacing traditional engine oil with ionic liquids also include the following: extremely low volatility will reduce the loss of oil during use, additives for cleaning carbon deposits and dispersing sludge are no longer necessary, because ionic liquids themselves are excellent solvents.

In terms of industrial gear oils, ionic liquids \([\text{Oley}][\text{Oleic}]\) have outstanding lubricity under extreme pressure conditions (Hertzian contact pressure up to 3.61 GPa), and its friction-reduction and antiewear effects are better than those of commercially available gear oils.

3.6. Other Lubrication Applications. Compared with traditional metalworking fluids, ionic liquids can reduce the damage of the cutter head and improve the surface finish of the workpiece in metal processing. In addition, the ultralow volatility of ionic liquids can greatly inhibit the generation of oil fume; ionic liquids are more environmentally friendly than traditional metalworking fluids.

In the metallurgical industry, ionic liquid has become a more advanced and safer coolant than water. Since 2016, SMS Group, a leading metallurgical equipment company, has applied ionic liquid coolants to several metallurgical installations.

In the field of micro/nanoelectromechanical systems (MEMS/NEMS), the nanotribological properties and durability of ionic liquids are comparable to those of perfluoropolyether. Ionic liquids are expected to be applied to microfluidic devices in the future (also known as lab-on-a-chip or micrototal analysis system).

The conductivity of traditional lubricants mostly relies on the addition of graphite, but graphite particles are not suitable for being introduced into bearings. With intrinsic conductivity, ionic liquid lubricants that prevent electro-erosion in roller bearings are already on the market. Conventional high-end lubricants, PFPEs, are electrical insulators and subject to decomposition under an applied potential; however, ILs are suitable for various electrical applications.

3.7. Substitute for Antiewear Additive ZDDP. Major automobile manufacturers and related organizations have formulated increasingly stringent standards to limit the content of sulfur, ash, and phosphorus (SAPs) in internal combustion engine oil. For this reason, the task of finding alternative additives to ZDDP is becoming more urgent. Ionic liquids have brought optimistic results. Compared with ZDDP, ionic liquids \([P_{666,14}][\text{BTMPP}], [P_{666,14}][\text{DEHP}], [P_{888}][\text{DEHP}], \text{ and } [P_{442}][C_{6}C_{1}PO_{4}]\) has better friction reduction and antiewear performance, lower SAPs content, higher thermal oxidation stability, and higher solubility in polar and nonpolar base oils. Engine testing proved that replacing ZDDP with ionic liquid in engine oil will cause a reduction in friction coefficient and wear, as well as a significant improvement in fuel economy.

4. PROSPECT ANALYSIS

The application of ionic liquid lubricating fluid is very promising. Take hydrogen compressors as an example, Sinopec plans to build 1000 hydrogen refueling stations in the next 5 years, and China plans to build 10,000 hydrogen refueling stations by 2050, the demand for hydrogen compressors and their lubricants will increase accordingly, and ionic liquids, as state-of-the-art lubricants of hydrogen compressor, will face huge market demand in China and globally. In other applications, such as oxygen compressors, hydraulic systems, space devices, vehicle engines, metalworking, industrial lubrication, lubricant functional additives, green lubrication, etc., with outstanding performance superior to that of traditional lubricants, ionic liquid lubricants have great potential for development.

Currently, ionic liquids are expensive. With the increase in production and technological progress, the price of ionic liquids is expected to drop significantly in the future.

So far, a total of 2175 ionic liquids are included in the database IL Thermo and a total of 2003 pure ionic liquids are included in the database Dortmund Data Bank. Theoretically, the anions and cations from these ionic liquids can combine with each other to yield more than 10\(^6\) simple ionic liquids and 10\(^{18}\) ternary ionic liquid systems. Venkatraman et al. enumerated more than 8 million ionic liquids that can be synthesized. Currently, only less than 1% of the total amount of ionic liquids has been studied. It is certain that more ionic liquids with better performance will be discovered in the future. The discovery of ionic liquid lubricants is very promising.

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Notes
The author declares no competing financial interest.

ABBREVIATIONS

\[C_{6}C_{1}im, 1\text{-ethyl-3-methylimidazolium}; C_{6}C_{2}im, 1\text{-propyl-3-methylimidazolium}; C_{6}C_{2}im, 1\text{-ethyl-3-ethylimidazolium}; C_{6}C_{3}im, 1\text{-propyl-3-ethylimidazolium}; C_{6}C_{3}im, 1\text{-butyl-3-ethylimidazolium}; C_{6}C_{4}im, 1\text{-hexyl-3-methylimidazolium}; C_{6}C_{4}im, 1\text{-hexyl-3-ethylimidazolium}; C_{7}C_{5}im, 1\text{-octyl-3-methylimidazolium}; C_{7}C_{5}im, 1\text{-decyl-3-methylimidazolium}; C_{8}C_{5}py, 1\text{-propyl-3-methylpyridinium}; C_{8}C_{5}py, 1\text{-butyl-3-methylpyridinium}; C_{8}C_{5}py, 1\text{-octyl-3-methylpyridinium}; C_{8}C_{5}py, 1\text{-propyl-1-methylpyrrolidinium}; C_{8}C_{5}pyr, 1\text{-butyl-1-methylpyrrolidinium}; C_{9}(2o1)pyr, 1\text{-methoxyethyl}-1\text{-methylpyrrolidinium}; P_{666,14}\text{-trihexyltetradecylphosphonium}; methide, tris-(trifluoromethylsulfonyl)methide; FTFSI, fluorosulfonyl-(trifluoromethanesulfonyl)imide; NTF_{2} bis(trifluoromethyl)-]
sulfonyl)imide; DEHP, di(2-ethylhexyl)phosphate; FAP, tris(pentafluoroethyl)trifluorophosphate; Oleic, oleate; TFO, trifluoromethanesulfonate; DCA, dicyanamide; OAC, acetate; DOSS, dioctyl sulfosuccinate; BTMP, Bis(2,4,4-trimethylpentyl) phosphinate; $N_{\text{base}}$ $\text{N,N,N-triocetyl-N-methylammonium}$; Oley, oleylamine; $P$, phosphonium; PAO, poly-alpha-olefin; PAG, polyalkylene glycol; POE, polyol ester; ZDDP, zinc dialkyldithiophosphate; PFPE, perfluoropolyether; MAC, multiply alkylated cyclopentane; EHD, elastohydrodynamic; X-1P, phosphazene; im, imidazolium; py, pyridinium; pyr, pyrrolidinium; ox, oxazolidinium; pip, piperidinium; mo, morpholinium; N, ammonium

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