



Tribological study of imidazolium and phosphonium ionic liquid-based lubricants as additives in carboxylic acid-based natural oil: Advancements in environmentally friendly lubricants

Carlton J. Reeves^a, Arpith Siddaiah^a, Pradeep L. Menezes^{a, b, *}

^a Department of Mechanical Engineering, University of Nevada Reno, Reno, NV, 89557, USA

^b Nevada Institute for Sustainability, University of Nevada Reno, Reno, NV, 89557, USA

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ABSTRACT

This paper highlights the potential of a new family of environmentally sustainable, functional bio-lubricants called room temperature ionic liquids (RTILs). In this investigation, a tribometer was utilized to carry out friction and wear experiments under ambient conditions to investigate the use of imidazolium and phosphonium-based RTIL's. The study investigated the tribological functionality of RTIL's as additives in plant-based avocado oil and as green sustainable fluids. The use of RTIL as additives in natural oil revealed that a higher percentage mix of RTILs can linearly improve the tribological performance. It was found that, depending on the type and percentage mix of RTIL with avocado oil, the coefficient of friction (COF) could be reduced by as much as 68.88% and the wear volume could be reduced by 73.37%. The RTILs were able to reduce the COF and wear volume not only when used as additives but also when used as base lubricating fluids. It was found that this was a result of the more resilient carboxylate acid-based monolayers that influenced the boundary lubrication regime. The present study details the influence of RTILs as additives, and the role of additive concentration in controlling the tribological performance to aid cleaner production research. Further, the study discusses their future as sustainable lubricants, and their underlying lubrication mechanisms.

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1. Introduction

Lubrication of moving components has been an integral part of the mechanical system since the early Roman and Egyptian era. Lubricants had a humble beginning with the use of fluids, such as animal and plant fats, lime, soaps, olive oil and even water which were combined to make the earliest known forms of grease and thick oil slugs (Bartels et al., 2000; Svanberg, 2006). The principal purpose of these lubricants even today is to minimize friction, heat, and wear between mechanical components in contact with each other. The control and minimization of these physical phenomena have always been to provide for an energy efficient system. While the pathway to achieve this is many, the present requirement is for these energy efficient systems to be environmentally benign and

sustainable.

The history of lubricants begins with natural sources, but over the years, human ingenuity powered by technological advancements has led to the development of many forms of lubricating oils whose sources are now primarily based in mineral and synthetic forms. Mineral oils are the most researched forms of oils that are sourced from naturally occurring petroleum or crude oil and are more commonly used due to the easy availability of the raw sources. Synthetic oils are manufactured polyalphaolefins, which are hydrocarbon-based polyglycols or ester oils, manufactured usually to target a specific application and environment which cannot be performed by a mineral oil. But, keeping aside the wide applicability of mineral and synthetic oils, it must be noted that none of these lubricants are environmentally benign. Their production process and post usage footprint on the environment are evident enough to raise concerns globally. It is important now more than ever to weigh the perks of using these sources of oils against the more concerning environmental impacts (Ing, 2009; Luther, 2000; Mang et al., 2011).

* Corresponding author. Department of Mechanical Engineering, University of Nevada Reno, Reno, NV, 89557, USA.

E-mail address: pmenezes@unr.edu (P.L. Menezes).

1.1. Bio-lubricants

For centuries, scientists and engineers have known that bio-based lubricants can also deliver favorable friction and wear performance (Reeves and Menezes, 2016a). Recently, research into the analysis of environmentally friendly oils has gained a substantial importance. The reason for this motivation is that 50% of all oils throughout the world is wasted into the environment through practice, leak, evaporation, or improper dumping. It has been estimated that nearly 95% of the lubricants which enter the environment are a resultant of non-bio-based oils which are harmful to numerous organic ecosystems (Schneider, 2006). Further, over 100 million gallons of contaminated oils' - leak, fall, and leach, annually into our healthy environment in North America alone. In the 1850s, when petroleum-based oils emerged, the utilization of bio-based oils declined significantly. In recent years, a rebirth of bio-based oils has taken place due to increased environmental consciousness to decrease the utilization of non-bio based oils, combat climate change, and curtail greenhouse gas emissions (Deffeyes, 2008; Goodstein, 2004). Researchers are exploring various renewable feedstocks such as protein (Patel, 2016; Su and Lee, 2007; Yu, 2007), tree leaves (Boudrahem et al., 2011; Hameed, 2009), seaweeds (Khan et al., 2009; Mark, 2015), vegetable oils (Issariyakul and Dalai, 2014; Subramaniam et al., 2013), coffee pulps (Gurram et al., 2016), paper mill sludge (Gurram et al., 2015), lignocellulose and other agro-residues (Adekunle et al., 2016; Asgher et al., 2014; Gupta and Verma, 2015) for the synthesis of many bio-based products which include fuels, lubricants, absorbents and stimulants (Panchal et al., 2017).

The use of renewable feedstocks, natural organic substances and/or synthetic esters to produce lubricants that are non-toxic to humans and the environment, can be categorized as bio-lubricants. These bio-lubricants have varying grades of biodegradability based on their inherent properties, composition and biological factors (Duzcukoglu and Şahin, 2010; Erdemir, 1990; Lovell et al., 2006). The revival of bio-based oils was a consequence of the rise in demand for sustainable materials which are less harmful to the ecosystem, renewable and offering viable-inexpensive replacements to toxic petroleum-based oils. From a research perspective, the research on bio-lubricants was ignited and fueled mostly by plant sources of oils, such as vegetable oils having a wide range of applicability as described in Table 1 (Karaosmanoğlu et al., 1999; Kumar and Sharma, 2011; Li et al., 2012; Mofijur et al., 2012; Sharma and Singh, 2010; Singh and Singh, 2010; Usta et al., 2011; Wang et al., 2011, 2012). The interest on liquid oils extracted from vegetable oils was due to their designed chemical structure where triacylglycerol molecules were created from long chains of polar fatty acids and esters, derived from glycerol (Menezes et al., 2013). Interest in bio-based oils has focused on analyzing the basics of saturated and unsaturated fatty acids with the primary focus on the use of naturally-based lubricants (Fox and Stachowiak, 2007; Lovell et al., 2010; Menezes et al., 2012; Reeves et al., 2015). Still, bio-lubricants are finding new applications such as carrier fluids for lamellar structure based powder additives particles in sliding contacts (Grushcow and Smith, 2005; Lovell et al., 2010).

Bio-lubricants tend to have many benefits over traditional petroleum-based oils, such as maintaining a higher lubricating property, shear stability, viscosity properties, load carrying capacity and a lower volatility as well as superior dispersancy and detergency (Bennion and Scheule, 2009; Menezes et al., 2012; Reeves et al., 2012, 2017a). Although these lubricants have favorable advantages, there are many drawbacks, such as their poor thermal-oxidative stability, high pour points, poor viscosity index, poor low temperature properties and inconsistent chemical composition (Nagendramma and Kaul, 2012). These drawbacks have led to the

Table 1

Specific applications of various vegetable oil (Panchal et al., 2017; Liew Yun Hsien, 2015; Shashidhara and Jayaram, 2010).

Vegetable oil	Applications
Canola oil	Hydraulic oils, tractor transmission fluids, metal working fluids, food grade lubes, penetrating oils, chain bar lubes
Castor oil	Gear lubricants, greases
Coconut oil	Gas engine oils
Olive oil	Automotive lubricants
Palm oil	Rolling lubricant, steel industry, grease
Rapeseed oil	Chain saw bar lubricants, air compressor-farm equipment, Biodegradable greases.
Safflower oil	Light-colored paints, diesel fuel, resins, enamels
Linseed oil	Coating, paints, lacquers, varnishes, stains,
Soybean oil	Lubricants, biodiesel fuel, metal casting/working, printing inks, paints, coatings, soaps, shampoos, detergents, pesticides, disinfectants, plasticizers, hydraulic oil
Jojoba oil	Grease, cosmetic industry, lubricant applications
Crambe oil	Grease, intermediate chemicals, surfactants
Sunflower oil	Grease, diesel fuel substitutes
Cuphea oil	Cosmetics and motor oil
Tallow oil	Steam cylinder oils, soaps, cosmetics, lubricants, plastics

invention of chemically adapted synthetic bio-based lubricants, stabilizing additives, and most recently ionic liquid (IL) lubricants (Kumar and Sharma, 2008; Reeves et al., 2013a, 2017b). In the current study, a natural plant-based liquid lubricant, avocado oil was employed as the base oil due to its proven high oleic acid and fatty acid contents which results in better tribological performance especially with additives, when compared to other natural plant-based lubricants as can be seen in Fig. 1 (Mannekote et al., 2013; Reeves and Menezes, 2016b; Reeves et al., 2015; Reeves et al., 2017a).

1.2. Room temperature ionic liquid (RTIL) lubricants

The lubrication industry today is more ecologically focused on achieving increased energy efficiency and sustainability through manufacturing of a new family of environmentally friendly oils. The ILs, mainly those that are liquid at ambient condition (called, RTILs), represent a promising new family of bio-based lubricants. They show a good potential to enhance the tribological properties and counter the drawbacks related to petroleum-based oils, bio-based oils, and lamellar solid powder additives (Reeves et al., 2017b; Yao et al., 2009; Zhou et al., 2009). The low friction properties of ILs was demonstrated by Ye et al. (2001), against synthetic oils for high load conditions (200–600 N) where the IL showed a coefficient of friction <0.060 under all loads (Ye et al., 2001).

RTILs' are molten salts that are typically made of combinations of a bulky, asymmetric organic cation and a suitable organic anion with melting points less than 100 °C and a liquid range of over 300 °C (Freemantle, 2010; Matlack, 2010). The molecular structure of an IL is as represented in Fig. 2. This molecular structure is known as a liquid lamellar crystal structure (Suisse et al., 2005) since it resembles a solid lamellar-like crystal structure similar to hBN. The only difference being that the anion and cationic form of ionic bonds that make-up the ionic bonded atomic planes are held together by the weak van der Waals forces (Manahan, 2011). This liquidus lamellar crystal structure of RTILs' exhibit a number of unique and useful properties that make them well suited as a new family of environmentally friendly lubricants and researchers have been investigating their properties extensively (Hernandez Battez et al., 2011; Liu et al., 2006; Reeves et al., 2017b; Xia et al., 2007; Yao et al., 2008, 2009).

The physical and tribological characteristics have been reviewed by Zhou et al. (2009) where they indicate that for ILs with the same

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