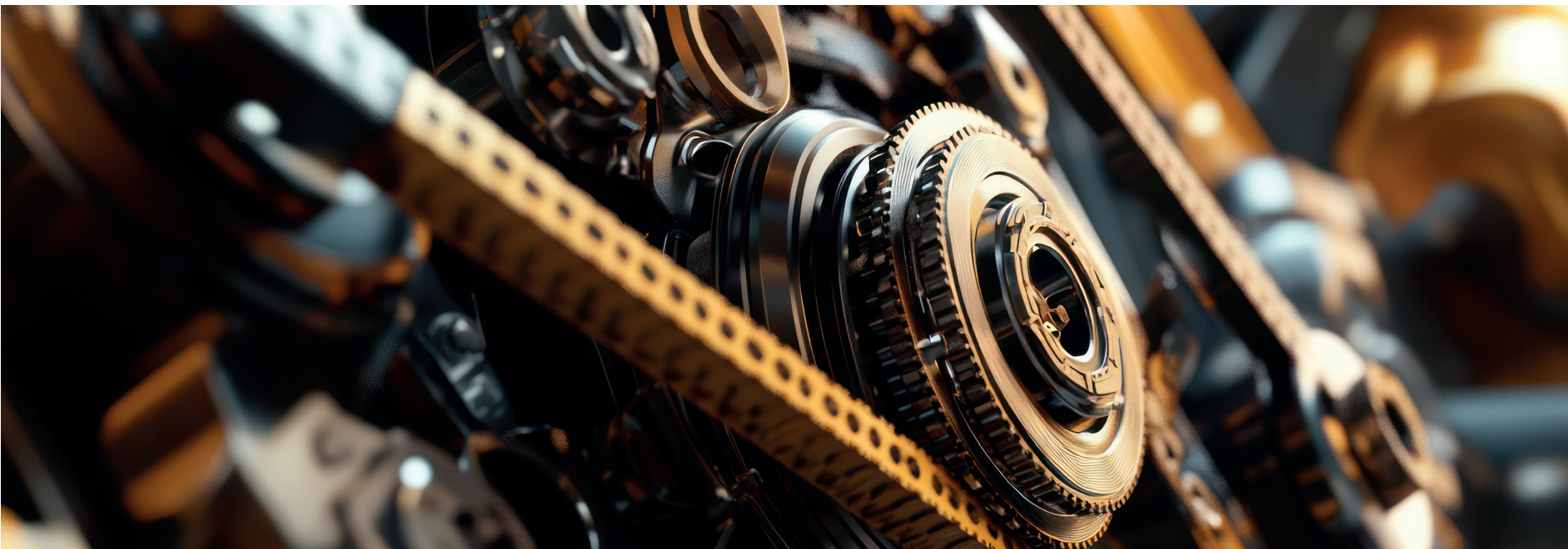




LUBRICATION TECHNOLOGY BREAKTHROUGHS IN INTERNAL COMBUSTION ENGINES THAT USE HYDROGEN AND/OR BIO-BASED FUEL SOURCES

Introduction

As of 2024, transportation is the leading greenhouse gas emitting sector in the United States at 14 million metric tons, a 0.8% increase from 2023 [1]. With over 94% of transportation vehicles relying on petroleum-based fuels, original equipment manufacturers (OEMs) are considering the applications of alternative fuel sources, including hydrogen and biofuels, for sustainable mobility in internal combustion engines [2]. While the concept of hydrogen and biofuels-powered engines is not new and already well-adopted, there are still challenges faced in manufacturing compatible lubricating oils for these advancements and understanding their fuel-oil interactions [3]. As OEMs gradually substitute gasoline and diesel with renewable fuel sources, they must understand the destructive effect green fuels have on the tribological performances of current mineral oil and synthetic oil-based lubricating oils. They then must design lubricant additives that can resolve the issues associated with each fuel type. This paper discusses the most recent discoveries about how hydrogen and bio-based fuels chemically alter lubricating oil properties, recommended commercially available lubricating solutions, and new research trends in developing lubricants for these special fuels.

1. Hydrogen Fuels

Hydrogen is broadly considered to be a substitute for carbon-based fuels in the transportation sector due to its ability to expand domestic production and power fuel cells with only water vapor and warm air emissions. Hydrogen fuel cells are garnering more interest as governments and industries are shifting towards a safe, decarbonizing, and economical approach to sustain fuel cell electric vehicles (FCEVs) and internal combustion engines (ICEs). Additionally, they are longer-lasting power sources compared to electric batteries, making them more suitable for 24-hour operations, long-haul operations, and other transportation demands in locations outside the electric grid. As of now, there are several consumer markets that use hydrogen, such as hydrogen fuel cell-operated light-duty FCEVs, buses, material handling equipment, heavy-duty trucks, and marine vessels. There is the potential to further expand these markets to include the aviation sector [4].

1.1 Effects of Hydrogen on Lubricating Properties

Hydrogen internal combustion engines (H2ICEs) offer promising environmental sustainability that can help reach a net carbon neutralization worldwide, but this field presents major challenges to researchers and lubricant formulators. One major challenge with H2ICEs is the regulation of hydrogen and generated water vapor contact with lubricant oils. When hydrogen contacts common lubricants, like conventional mineral oils and Group IV synthetic oils, an emulsification reaction occurs that can degrade lubricating oil films strength and lower their life expectancy

[5]. During emulsification, hydrogen hydrolyzes lubricating oil molecules and additives to produce acidic by-products. These acidic by-products then weaken lubricity, promote foaming behavior, corrode metal parts, and cause sediment or sludge buildup [6], [7]. The water vapor produced by hydrogen combustion also leads to high water accumulation of 1% to 5% in engine oils that can increase oxidation by tenfold, cause the oil to pre-maturely age, degrade engine performance, and further emulsion formations [7],[8]. Furthermore, in H2ICEs, lubricating oils contaminate combustion chambers through piston-rings system leakages or oil droplets carried by blowby gas streams. Lubricating oil contamination forms highly reactive spots in combustion chambers that can lead to pre-ignition, detonation, and engine backfire phenomena across all engine types. However, the low ionization energy of fuels like hydrogen are at the highest risk for exacerbated ignition and combustion issues [9]. Consequently, lubricant formulators should consider base oil and additives qualities for pre-ignition risks and set volatility requirements to ensure sufficient lubricant oil loss and durability. While there are already numerous concerns regarding hydrogen fuel interactions with lubricant oils in the combustion chamber, their reactions to other engine components, including seals, gaskets, and hardware metals, are still unknown with further research needed to promote the reliability of H2ICEs [8].

1.2 Hydrogen Internal Combustion Engines: New Industrial Lubricants and Research Trends

Many companies have successfully engineered specialized lubricants that address hydrogen fuel concerns on oil stability and performance efficiency in H2ICEs. One such company is Lubrizol Corporation, a New Jersey-based chemical research company with a patented H2ICE lubricant using a synthetic based oil. Their unique addition of specific additives, like nitrogen-containing dispersants, magnesium detergents, antioxidants, and friction modifiers, optimizes H2ICE efficiencies by regulating water accumulation in engines and inhibiting corrosions from water vapors.[10] The Chemours Company, a Delaware-based chemical company, is another successful H2ICE lubricant developer who has formulated synthetic perfluoropolyether fluid (PFPE fluid), an inert lubricating oil that is nonflammable at extreme temperatures with high compatibility with highly reactive elements, like hydrogen, oxygen, and chlorine [11].

Other research pathways explore coupling biolubricants with H2ICEs to enhance environmental sustainability and reduce toxicity. In 2020, Cheah et al. produced a microalgae oil biolubricant through the esterification of crude oil obtained from dried Chlorella biomass. Upon adding 10% of microalgae oil into pure poly-alpha-olefin (PAO) synthetic oil to create a modified microalgae oil (MMO), the MMO demonstrated superior anti-friction and anti-wear properties via a high frequency pin-on-plate test. The coefficient of friction was reduced by up to 10.1% with noticeable less wear loss, decreased surface roughness, and

improved heat dissipation compared to that obtained by pure PAO [12]. Microalgae oil's excellent tribological traits and renewable feedstock introduces itself as a new, competitive lubricant alongside readily available synthetic lubricants for specialized H2ICE applications that may inspire biolubricant researchers into this field and encourage wider H2ICE commercialization.

2. Ethanol

Fossil fuel dependence has led to future energy security concerns due to its global non-uniformity and long-term availability. Fortunately, biofuels have been recognized as immediate alternative fuel sources due to their shared heating values and properties with fossil fuels, replenishable nature, aquatic non-toxicity, and agrarian economy stimulation [13]. Biofuels are liquid fuels derived from plant material, animal fats, and algae, such as sugarcane, wood waste, manure, and modified bacteria [14]. As of now, they are the preferred decarbonization route worldwide responsible for a forecasted 7% decrease in transportation sector oil demand by 2028 due to a 23% increase in growth demand since 2023 [15]. Ethanol, created through starch and sugar fermentation or processed cellulose biomass, is the most produced biofuel worldwide for ethanol-fuelled engines, gasoline blends in ICEs, and oxygenated additives in gasoline to reduce emissions from older, high-polluting vehicles [16], [17]. While there is a plethora of biofuels, like biodiesel, renewable diesel, and biogas, present in industries and research studies, this section selectively focuses on ethanol-fuelled engines, the effects of ethanol on lubricating properties, and lubricant trends in this field.

2.1 Effects of Ethanol on Lubricating Properties

Ethanol fuel has been used worldwide since the 1970s, but its chemical interactions with lubricant oils and additives have yet to be well-understood as it gains popularity in industrial and transportation sectors. Currently, it is known that ethanol possesses higher heat of evaporation and boiling point than gasoline and diesel fuels, causing uncombusted ethanol to condense on cylinder walls and migrate to the oil sump, an integral component of an engine's lubrication system. In fact, 6% to 25% of ethanol and its combustion products can be found in the crankcase, the protective casing of the whole engine block, that can lead to fuel-oil contamination. Lubricating oil contaminated by ethanol dilution experiences higher oxidation, accelerated thermal aging due to the higher oxygen content in ethanol, acidification that increases friction and engine wear, and viscosity reduction. Furthermore, ethanol-contaminated lubricant oils impede the growth of tribofilms formed from zinc dialkyldithiophosphate (ZDDP), the most common anti-wear additive found in engine lubricants. In fact, most automotive lubricants rely on the presence of tribofilm to protect engines from wear. When ZDDP undergoes shear stress on metallic surfaces, sulfur and phosphorus from ZDDP form sulfides and phosphates that react with zinc to generate long-chain zinc

Figure caption

polyphosphates, protecting the surface from oxygen and debris. Nonetheless, ethanol contamination of lubricating oils inhibits the polymerization and regeneration of long phosphate chains in ZDDP, leading to thinner, irregular tribofilm formation that reduces debris cushioning, higher coefficients of frictions, and increased wear [18].

2.2 Ethanol-fueled Engines: Commercially Available Lubricants and Nanoparticle Additives Research

Commercially available lubricant oils with fuel system stabilizers and vapor rush inhibitors are recommended to mitigate these concerns in ethanol-based engines. Other lubricating oils specially suitable for ethanol fuels are two-stroke oils [19]. Two-stroke oils, typically used in smaller engines, use synthetic or mineral base oils and require fuel-oil mixing for superior lubrication, like excellent heat dissipation, carbon deposit prevention, and corrosion inhibition [20]. Ongoing research directions for ethanol-based engine tailored lubricants include adding greater amounts of oxidation inhibitors, dispersants, and detergents compared to those already in gasoline engines and incorporating antiwear and friction modifiers additives that flourish in fuel contamination [18].

Nanoparticles are among the most promising additives for improving the lubricating properties of biolubricants. Nanoparticle additives, such as graphene, molybdenum disulfide, and metal oxides, utilize their small, spherical shape to create a rolling friction effect as opposed to the typical sliding friction between surfaces, polish rough surfaces, and develop a protective film that can compensate for weight loss. As a result, these unique particles exhibit effective friction and wear reduction, improved load-carrying capacity, energy conservation, and high thermal stability [21].

In 2024, Hamnas and Unnikrishnan conducted a tribological analysis of mustard oil-based biolubricant enriched with calcium carbonate nanoparticles derived from waste eggshells. Samples of this biolubricant presented higher contact angles compared to that of standard mineral oil SAE20W-40, indicating higher sticking characteristics onto metal surfaces. Additionally, the biolubricant samples observed 80oC higher thermal stability at 370oC, 30oC higher flash point, lower wear scar diameter, 20 J less energy consumption, and around 1.7 times higher viscosity index compared to those of mineral oil [22]. The study successfully demonstrates the potency of nanoparticles in biolubricants and their potential substitution of mineral oil as a more favorable, green high-performing lubricating oil. Characteristics like higher flash point and lower energy consumption also point towards the higher thermal stability of nanoparticle biolubricants that can hopefully impede thermal aging and degradation in lubricating oils associated with ethanol-fuelled engines.

In 2020, Singh et al. investigated the application of transesterified julfiflora oil as a biolubricant and evaluated the effect of titanium dioxide nanoparticles additives on its tribological properties. Titanium dioxide nanoparticles at 0.6% concentration exemplified an optimal reduction of pin wear and coefficient of friction under SEM imaging, higher viscosity index and flashpoint, and the low iodine and acid values [23]. High viscosity, low iodine value, and low acid value are particularly important for lubricating oils in ethanol-based engine applications to circumvent existing complications regarding poor tribofilm layer protection and lubricant oxidation or acidification from ethanol dilution. The inclusion of nanoparticles in biolubricants already conveys strong viability that will hopefully be applicable to lubricating oils for ethanol-fuelled engines, a breakthrough that will further green chemistry and sustainability advocacy.

Conclusion

The development of lubrication technology suitable for hydrogen and biofuels, like ethanol, in internal combustion engine applications is a new research trend that would advance the field of tribology. Fuel and oil contamination remain the root cause of lost engine efficiencies due to fuel dilution, pre-ignition risks, reduced tribolayer, and increased friction and wear that should be noted in lubricating oil formulations. Challenges arise in not only effectively incorporating additives to shelter lubricating oil properties but also to compel these changes to satisfy stringent environmental requirements and scale these modifications on an industrial level. Fortunately, modern researchers have proposed the implementation of metallic nanoparticles in biolubricants derived from readily available biological sources, like julfiflora oil, mustard oil, and microalgae, that showcased outperforming anti-friction abilities compared to that in standard lubricating oils. The adoption of nanoparticles and biolubricants in alternative fuel internal combustion engines in the industrial and transportation sectors would be an innovative exploration that will hopefully set us a step closer to carbon neutrality.

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