Future potential framework for synthetic lubricants

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With an ever-expanding international economy reliant on mechanical systems and operations, the importance of proper lubrication only grows as new technologies, demands, and applications are developed. A proper lubrication system is key to maximising mechanical performance and efficiency, two factors sought after by both market demands and govern¬ment/ environmental regulators. As a result, the annual global consumption of finished lubricants in the last decade alone ranged between 38-42 million tons with an expected growth to 48 million tons by 2035 [1], in relation to today´s 4,500 million tons of crude oil [2]. However, ongoing pressure from climate change, sustainability concerns, and the continuing pursuit of higher performance by original equipment manufacturers (OEMs) in automobiles, industrial vehicles, and commercial industries has set a need for improved lubricants which has led to the current trend of implementing synthetic lubricant formulations. The capabilities of lubricants to reduce friction during the use phase, which transduces in energy savings within scope 3, category 11, downstream emissions, has never been in such a demand. Although market penetration and demand of synthetic lubricants were limited due to high cost in the past, synthetic lubrication now promises to be a strong candidate for meeting the needs of the future as continued research and development improves performance even further and stricter regulations/ demands for sustainability outweigh the cost difference with conventional lubricants.

The half-life time of lubricant specifications

Since the beginning of base oil/lubricant classification, all specifications have been and will continue to be replaced by new ones that account for higher demands for performance and longer life by OEMs and market trends. This continuous demand for higher performance, further exacerbated by environmental regulations and societal drivers lead to spiraling development costs that influence the internal specifications of OEMs and thus the choice of lubricants. As such, regulatory mandates such as the ILSAC GF-6 released in 2020 are rolled out periodically every few years to balance government environmental regulation and the industry's capability to develop lubricants [3]. Internationally, similar mandates would include Euro VI standards and BS VI (Euro VI)/Standard V in China and India. Such mandates influence lubricant properties deemed acceptable such as the metal/ash content in addition to greater performance in lubricity and lifetime.

This progressing fragmentation of standardised oil specifications between Europe, Asia and U.S. results in an associated diversification in OEM specifications that poses a challenge for lubricant manufacturers to meet. Second, new classes of functional fluids, such as thermal management fluids and electric vehicle lubricants, have appeared. Further, a rising choice in propulsion technologies and energy carriers (fuels) are on the horizon [4]. On top of that, more fuel grades that include blends with various kinds of biofuels will be released. This adds additional, non-technical layers to the products that lubricant manufacturers will produce and each one will have optimal use scenarios in which the downsides of one lubricant base stock are not as relevant as the significant improvement in other areas, mainly driven to societal drivers. This creates a simultaneous push for higher performance lubricant base stocks while enabling a wider variety of lubricants to be commercially viable due to the advantages that certain base stocks may have in one area over another. The challenge for future lubricants is summarised as follows: more investments and tests due to shorter shelf lives of specs enhanced by a multitude of societal drivers and uncertainties.

Base oil classifications

At the heart of every lubricant is a base oil derived from a mineral, synthetic, or other source. The American Petroleum Institute (API) categorise base oils based on composition, level of refinement, and physical properties such as viscosity and pour point. The base oil classifications I to II are related to mineral oils and supply 90% of the finished lubricant market, while class III oils represent a higher level of refined mineral oil and class IV and V represent synthetic (polyalphaolefins) and any other base oil source (silicones, glycols, etc.) respectively [5]. Figures 1 and 2 depict the shifting utilisation rates of each of the base oil categories within the past couple of decades. Evidently, industries have shifted their demand from group I to the more refined group II and group III oils.

The five API base oil group categories (API 1509, Appendix E (09-2019)) give only a loose definition for the term "synthetic" since only group IV contains a truly 100% synthetic lubricant: poly-alpha-olefins (PAOs). All other base oils not meeting Groups I to IV, including silicones, phosphate esters, polyalkylene glycols (PAG), esters, polyolesters, biolubes, etc. are classified as Group V, even though most of the aforementioned base oils are synthesised and can fit the definition.

In contrast to the API, the National Advertising Division (NAD) of the Council of Better Business Bureaus ruled in April 1999, that hydroprocessed base oils (Group III) can be marketed as "synthetic" base oils, like PAOs. Castrol and Mobil had a disagreement on the use of the word synthetic as a description of certain lubricants. Mobil objected (despite allegedly having marketed hydroisomerised API Group III base oils as "synthetic" in Europe and elsewhere) that Castrol's hydroprocessed Sintec was not synthetic. According to the NAD, a mineral oil is therefore a "natural" product, made by separation processes; a synthetic oil is a "man-made" product, made by building big molecules from small ones. Although some synthetic oils will contain molecules that are very similar in structure and properties to those found in mineral oils, the essential difference between them is that a synthetic oil is composed of a narrow range of very similar or nearly identical molecules, while a mineral oil is composed of a wide range of approximately similar molecules. The chemical and physical properties of a synthetic oil are much more uniform and predictable than those of a mineral oil.

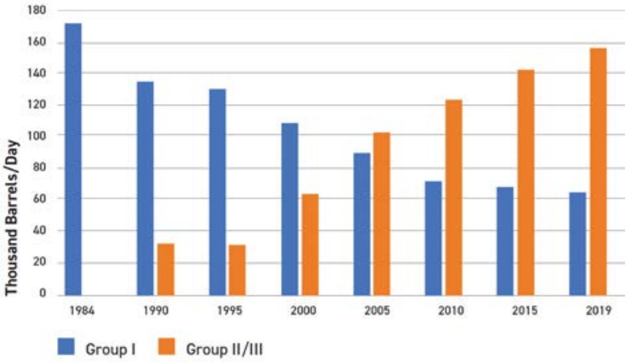

Figure 1: *Shift of global base oil supply in North America from 1984-2019 [6].*

A separate study by Exxon Mobil found that 84% of oil industry professional correspondents agreed that it is a result of the latter being more likely to meet new specifications for sulfur content, fuel efficiency, and viscosity index caused by technological innovation and stricter environmental regulations [3].

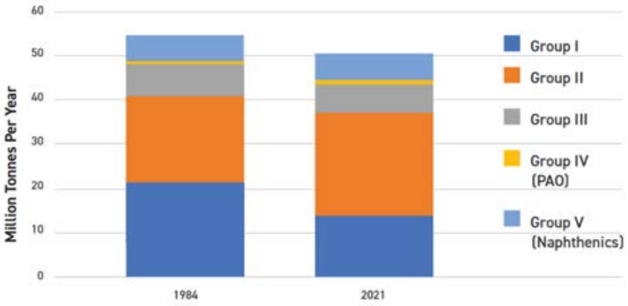

Figure 2: *Shift of global base oil supply in terms of categories from solvent to hydroprocessing [6].*

While chemical modification of crude oil base stocks and additive packages can allow crude oil-based lubricants to match or even exceed the properties of true synthetic base oils, the strong properties of true synthetic base stocks such as PAOs, PAGs, esters, and even bio-lubricants are too attractive to pass on. In particular, the NOACK volatility at low viscosities of true synthetics, such as PAOs, PAGs and esters, are nearly always inherently superior (e.g. lower) to traditional oil base stocks as shown in Figures 3 and 4. With regulations for fleet fuel economy and stringing emission standards increasing demand for low-viscosity (SAE 0W-16/0W-12) and ultralow-viscosity (SAE 0W-8) grade base stocks, the associated stringent NOACK limits will out phase group II-based formulations and push the industry towards higher API base groups. With similar new regulations rolling out every few years such as the newest API Service Category for gasoline engines (SP) having rolled out on 1 May 2020, it is clear that "synthetic" base stocks, derived from crude oil or not, will play a greater role in meeting higher demands for performance and environmental friendliness for future decades to come.

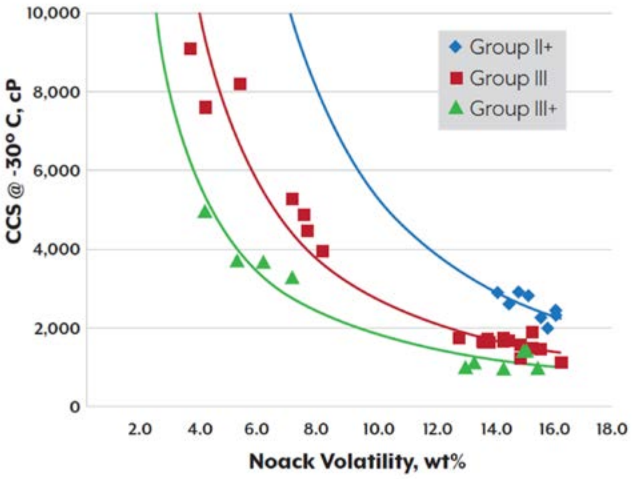

Figure 3: *Relationship between Noack volatility and CCS viscosity [7].*

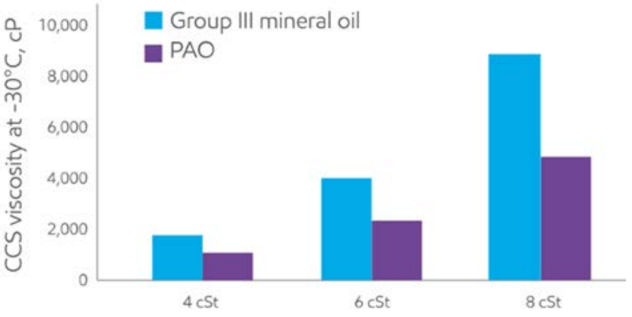

Figure 4: *Cold-cranking simulator (CCS) viscosity for low viscosity API base stocks [8].*

One has to keep in mind, that hybrid (HEV) and plugin hybrid (PHEV) vehicles still operate an internal combustion engine (ICE), but the operating mode differs from full ICE powered vehicles. These ICEs are smaller in displacement and power, but operate at lower oil sump temperatures for HEVs in average of 10°C cooler and in PHEV of ca. 15-20°C, because they operate periodically for charging or while running on battery. The consequent start-stop cycles going sudden to full load and heavy loads at cold start for charging or delivering a spontaneous power surplus representing aggressive operation modes for lubricants leading to an increased acid formation, fuel and water dilution by excessive blow-by as well as sludge formation. Such operating conditions call for adapted "synthetic" or "truly synthetic" base oils.

The birth of synthetics

In 1929, synthetic oil was first developed, but the commercial production gained popularity in aviation during the second world war because of its high efficiency, cold start behavior, and low deposit forming. The commercial development began in:

a. Polymerised olefins from Standard Oil of Indiana [9],
b. Polyalkylene glycols (PAGs) from Union Carbide [10] and I.G. Farben [11],
c. Catalytic polymerisation of ethylene and olefins [12] in different plants in Germany during WWII and
d. Synthetic esters from petrochemical adducts at I.G. Farben [13].

At the time, synthetic oils were considered to be in the early development stage but eventually found themselves in use during WWII as engine oils made from PAGs in the U.S. Air Force and as blend of a diester with polyethylene oil in the German Luftwaffe [14]. As with several other technological developments of the time, societal pressure was high enough that functional benefits overruled the financial costs. Today, a similar phenomenon regarding eco-toxicological properties such as sustainability and biodegradability has influenced the demand for synthetic base oils. Sustainability with a content of renewables and high performances draw a favorable frame for synthetics. With around 800,000 tons in 2017 (or ~2%) [15], synthetics as per API Group V represent still a small portion of the global market.

Types of Synthetic Base Oils

"Synthetic" oils are synthesised from a multitude of processes involving simple hydrocarbon streams such as ethylene and propene, as shown in Figure 5, and can be classified in terms of basic differences in their molecular backbones, specifically, whether they are polar or non-polar base stocks. This polarity can influence the lubricity, NOACK volatility, and viscosity indices of the end lubricant synthesised from the base stock and can be taken advantage of to tailor the lubricant's performance to its specific use-case scenario.

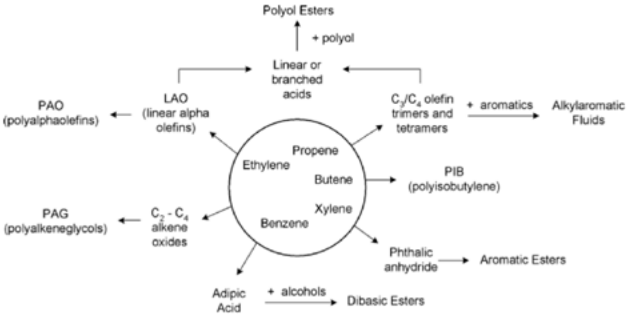

Figure 5: *Diagram of synthesis routes for common lubricant base stocks [16].*

A. Non-polar synthetic base stocks

- Polyalphaolefins (PAO)
  PAO are hydrocarbons manufactured by a process of catalytic oligomerisation or polymerisation to low molecular weight linear-olefins, particularly ten (C10) or twelve (C12) carbon atoms. The resulting oligomers consist of dimers, trimers tetramers and so on. PAOs are classified as API group IV and commercially possess viscosities between 2 and 100 cSt @ 100°C with low viscosities between 4 and 8 cSt representing 80% of the total volume [17]. It has a higher temperature stability of approximately 320°F to 520°F, which is higher than the mineral oils. Lack of wax content allows it to function at low temperatures too (-50°C to-60°C). It is also the most cost-effective synthetic.

  Polyinternal olefins (PIO)
- PIOs are synthesised from the oligomerisation of n-olefins (C_{15}-C_{16}), commonly with a BF3 catalyst, and are composed mainly of dimers and trimers. They were classified as group VI (ATIEL, Europe only) base oils, and have comparable results in bench and engine tests along with PAO [17].

- *Gas-to-liquid (GTL)*
  The formation of syngas, a mixture of carbon monoxide and hydrogen, with the subsequent Fischer–Tropsch type synthesis is not limited to the use of natural gas (case of GTL). Renewable raw materials, or any kind of biomass[1], can be used as feedstock and converted by Fischer–Tropsch synthesis to "Green Crudes". Both are often used to synthesise Iso-paraffins, which possess qualities on par with PAO such as high viscosity index, flash point, and low volatility and performed well under ILSAC GF-3 Sequence IIIF and Sequence VIB engine tests.

- *Bio-Olefins*
  Biological processes form hydrocarbons by synthase instead of synthesis and use biomass and renewables as feedstocks. As such, most biological olefins are bran¬ched and unsaturated requiring a hydrogenation process in order to elimi¬nate the double bonds and to obtain properties suitable for finished lubricants [18].

- *Coal-to-liquid (CTL)*
  In a similar fashion to GTL, Coal-to-liquid (CTL) base stocks include Fischer-Tropsch processes to synthesise syngas, which are then converted to crudes. Although more direct methods such as coal pyrolysis exist, they are not yet as commercialised [19]. However, Fischer-Tropsch can also be used to process biomass in order to synthesised renewable hydrocarbons.

B. Polar synthetic base stock [20]

- Esters
  Esters are a wide class of synthetic base stock and include mono, dibasic, polyol, and aromatic esters that each have different molecular structures and properties. They are often synthesised from mono- or polyvalent alcohols and mono- or polyvalent acids, which can be fully sourced from renewables. In general, ester fluids possess extremely attractive properties such as high solvency for additives (which other synthetic stocks, such as PAO, do not possess), superior lubricity and volatility to PAOs, and potential for high biodegradability through proper choice of molecular composition. However, esters suffer from low hydrolytic stability at elevated temperatures that cause the ester to decompose into acids that can harm mechanical components. With proper additive packages, the hydrolytic stability can be increased to minimise this decomposition. The most common esters in lubricants, turbine jet and hydraulic oils are:

  a. Di-isotridecyladipate (iC_{13}-C_6-iC_{13}),
  b. Trimethylolpropane ester (TMP-(C_8-C_{10})$_3$)
  c. Pentaerythritol tetraester (PE-(C_5-C_{10})$_4$)

- *Polyalkylene glycols (PAG)*
  Polyalkylene glycols are a family of synthetic polymers derived from petrochemical-based monomers of ethylene oxide (EO) and/or propylene oxide (PO). These monomers can be obtained from renewables, like EO from bio-ethanol and PO from bio-glycerin, except the starting molecule, like bio-butanol. Depending on the backbone, they can be water soluble or insoluble and hydrocarbon tolerant as well as oil soluble. PAGs are available in large volumes due to its use in polyurethane applications and generally excellent lubricity, strong solvency for additives, and are non-varnishing/low ash (leaves little to no residue) [21]. They also tend to have high intrinsic viscosity of 160 to 250 cSt, in addition to good low and high temperature abilities. Lean burning without forming particulates is also seen as a benefit for these engine oils [22].

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[1] *Main feedstocks: energy crops and trees, agricultural food and feed crops, agricultural crop wastes, wood wastes and residues*

- *Estolides*

Unsaturated or hydroxycarboxylic fatty acids, as a renewable (plant) resource, possess strong lubrication properties that are difficult to apply in finished lubricants due to inadequate low temperature performance and oxidative stability. However, when converted to estolides, they become fully saturated secondary esters (as estolides) with mostly hydrocarbon characteristics that imparts stronger oxidative/low temperature performance [23]. There are two options for synthesising estolides from vegetable oils (oleic):

a. Polymerisation involving the epoxidation of unsaturations (double bonds) of fatty acids
b. Reactions of hydroxy fatty acids, such as ricinoleic acid (C18:1-0H) from castor oil or lesquerolic acids (C20:1-0H).

Future evolution of base oils

Today, lubricants meet demanding technical-oriented specifications by OEMs and private/government regulatory bodies such as the API. In the future, more non-technical criteria, like eco-toxicological properties and/or sustainability, will need to be added into their functionality on top of the technical requirements. With the large presence of environmental regulations from organisations, like the ECHA or EPA, the growing concern for sustainability and the future of the lubricant industry from the next five to the next fifty years is called into question. As such, life-cycle analysis of the entire production processes will become more important as regulatory bodies push for minimising CO_2 emissions. It would be remiss to discuss the near future of lubricants for both automotive and industrial applications without mentioning the shift towards sustainability and "green chemistry" to derive new base oils. This can include existing backbone chemistries with known functional profiles, but using renewable resources to derive them in total or in part.

- Environmental acceptable lubricants

Around 1990, the first environmental acceptable lubricants, or "EALs", appeared in Europe, which met eco-toxicological properties. Today, the most important schemes for EALs are:

a. European ecolabel as per EC/2018/1702 (3rd revision),
b. Second issuance of U.S. Vessel General Permit (VGP), now in force as VIDA, and
c. Biolubricants as per EN16807.

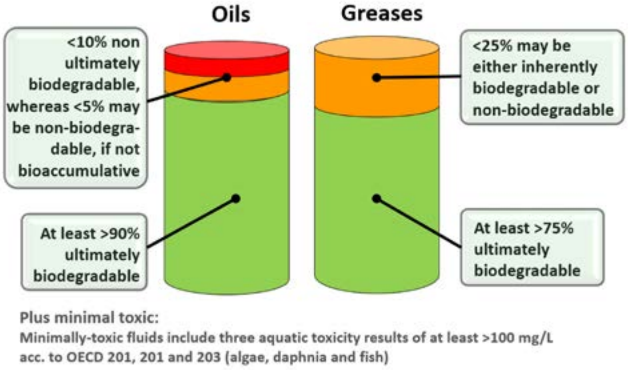

Figure 6: *Diagram representing key criteria for environmentally acceptable lubricant composition as according to the US VGP 2013 [Courtesy: MATRILUB].*

Today, "Biolubes" that fulfill EAL requirements are available for all classes of lubricants and operate safely but are fit for purpose. Most esters and PAGs used as base oils may also meet the ecotoxicity requirements for EALs through the use of additive packages. In general, EALs have varying contents of renewables defined by the 14C content (as per ASTM D6866 or EN16640) and are generally defined by the requirement for biodegradation (ready or ultimate as full mineralisation) or short persistence in the environ¬ment when spilled. They have very limited toxicities for aquatic species, like fish, daphnia, and algae, and different models for evaluating the potential for bioaccumulation. For example, the second issue of the US VGP 2013, the acceptable limits for oil are at least 90% biodegradable and at least 75% biodegradable as shown in Figure 6. This issue of environmental friendliness is defined by legislations and guidelines as well as verifiable test methods, which often include numerous ASTM testing.

- Sustainability of lubricants

The 17 Sustainable Development Goals (SDG) adopted in fall 2015 by the United Nations draw wide frames for different aspects of sustainability, but are non-mandatory. They teach about the complexity of sustainability. However, the international consensus for a new class of lubricants called "sustainable lubricants" is blurred and possess attributes different from EALs. EALs focus on human and environmental toxicology, whereas sustainable lubricants will consider:

a. Sourcing sustainable and renewable bio-components (CO_2 neutral) for base oils and additives (in part also used for EALs).
b. Waste reduction by longer drains.
c. Circular economy by full waste collection of used oils (re-refining or cycled resources (plastics to base oils).

d. The carbon dioxide emissions during their whole lifecycle (low friction) of lubricated machineries as scope 3, category 11, downstream emissions.
e. The consumption of resources with respect to their lifetime and the generation of individual carbon dioxide emissions as a result.

Even the third revision of the European Ecolabel for lubricants EC/2018/1702 no longer contains a content of renewables, "sustainably sourced bio-based and other raw materials" are an element in article 13(1a) of the new European Taxonomie EU/2020/852. A roll back in the fourth revision to renewables is likely.

Explicitly, article 2(17) of EC/2018/1702 highlights energy efficiency, which is related to CO_2 emissions via friction reduction:" 'energy efficiency', a more efficient use of energy along the entire energy supply chain from generation to final consumption;' ". Further, according to article 10(1b), an economic activity makes a significant contribution to climate protection, if it increases energy efficiency, what indisputably low-friction lubricants do. Friction occurs everywhere along the value chain. Carbon pricing will be the tail wind for synthetics and/or truly low friction lubricants. Friction reduction transduces directly in CO_2 savings and becomes as such a quantifiable economic value to open a business case for synthetics and truly low friction lubricants. Toyota has applied life cycle analysis (LCA) tools and evaluated the CO_2 emissions ratio of their vehicles. They have found that the use phase of their vehicles accounted to 81.8 % (FY2019) and 80.5% (FY2020) [24] of all indirect emissions (scope 3). The fuel consump¬tion of vehicles motorised with internal combustion engines during the use phase con¬tributed to the largest portion of greenhouse gas emissions. Consequentially, low friction engine and gear oils improving some percent in fuel economy exert a direct impact on CO_2 emissions times the carbon pricing calculates an economic value of synthetics and/or low friction lubricants. Carbon pricing revenues from low friction lubricants counteract the additional costs of synthetics.

Conclusion

The traceability of renewable or biogenic raw materials in life-cycle analysis processes is a critical issue as the content of these resources determines the lubricant's sustainability and CO_2 footprint, but CO_2 savings through significant reductions in friction in the use phase overrule all other parameters in LCAs. Criteria for sustainability can be found in the seventeen sustainable development goals (SDG) of the United Nations from October 2015 and are critical to determining the true sustainability aspect of synthetic lubricants and their processes. Such legislation related to EALs and similar concepts for sustainable lubrication has been and will continue to be of utmost importance as research and industry shifts away from a heavy reliance on fossil resources. In addition to this ongoing chase for sustainability, Gen Z (born after 2000) have shown a high sustainability and environmental awareness and are willing to pay more for brands that are more sustainable.
As a result, future low friction lubricants will favor synthetics synthesised from renewable or biogenic raw materials processed by chemical synthesis. Sustainable lubricants from biomass and other renewable sources made of esters, PAGs and bio-olefins have continued to show very promising low friction performances and are key to addressing the issue of eco-toxicity and sustainability. As the new decade progresses, such synthetic base stocks, whether truly "synthetic" or not, will continue to replace conventional petroleum lubricants to achieve a more sustainable future of lubricants.

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