Conversion of Plastic Waste into High-Value Lubricants: Techno-Economic Analysis and Life Cycle Assessment

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Conversion of Plastic Waste into High-Value Lubricants: Techno-Economic Analysis and Life Cycle Assessment

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Given the low recycling rate of the plastic waste in the United States due to low economic incentive, it is of great interest to develop a technology to upgrade plastic waste with favorable economics. Plastic upcycling to valuable chemicals could ensure a circular economy for plastics and reduce the environmental burden caused by their end use cycle and disposal. A conceptual facility to convert 250 metric ton (MT) per day of plastic waste was modeled; the main product was a high-quality liquid (HQL) with similar performance of polyalphaolefin (PAO) lubricants. The modeled process had a lubricant yield up to 90% based on experimental results at the laboratory scale. Techno-economic analysis (TEA) and life cycle assessment (LCA) were also performed to evaluate the process economics and its environmental impact. By using a mix of colored and natural high-density polyethylene (HDPE), the production cost was in the range $0.6–$1.98 per kg of lubricant, depending on the operating conditions. The life cycle emissions were in the range of 0.48–1.2 kg$_{CO2e}$ kg$_{Lub}^{-1}$ showing, for the best case scenario, a 52% reduction relative to the emissions for petroleum lubricants, and a 74% reduction relative to the emissions for PAO lubricants. The impacts of the lubricant yield, the catalyst amount, and reaction time were evaluated, and their effect on the final production cost discussed.

Introduction

Use of plastic products is ubiquitous and important for everyday life as they are convenient, sturdy, and cheap. Prior to the COVID-19 pandemic, the global plastic production was 368 million metric ton (MMT) in 2019.[1] The global production is forecasted to top 900 MMT per year by 2050.[2] A direct consequence of the increase in plastic production is the generation of plastic waste, which has shown a negative impact on the environment and health due to the high chemical resistance of plastics, particularly polyolefins.

Plastic wastes have been a persisting global threat by polluting soil and ocean,[3] harming land and marine organisms,[4] and jeopardizing human health by entering in the drinking water cycle and food chain, as well as by air pollution.[5] These plastic wastes are sourced not only from developing countries that do not have sufficient infrastructure for solid waste management, but also from developed countries, which are characterized by a much higher waste generation per capita.[6] For example, the United States generated in 2016 the largest amount of plastic waste in the world, and it was estimated to have generated up to five times the amount in 2010.[5,6] In 2018, EPA reported that the U.S. generated 32.4 MMT of plastic waste.[7] Of these, only 8.6% were recycled, 5.1 MMT were combusted in municipal solid waste (MSW) facilities, with the remaining 75% of plastic wastes ending their life cycle in landfills. Therefore, it is a national and global priority and urgency to address issues linked to the plastic waste pollution by improving its management, particularly in an economic and impactful way.

The plastic waste management pathway can be divided in three categories, as shown in Figure : downcycle to lower value products (e.g. electricity, recycled plastics for carpets), recycle back to monomer, and upcycle to higher value products (e.g. BTX, wax). In order to tackle the challenges related to plastic waste, it is important to develop technologies or practices that are economical, scalable, and impactful. Particularly, the economics of plastic conversion technology is the key element, within the context of environmental sustainability.

Extensive research has been conducted to increase the value of plastic waste, given that plastics are also high-density energy carriers mainly made of hydrocarbons. For example, in the U.S., the average non-recycled plastic waste has a low heating value

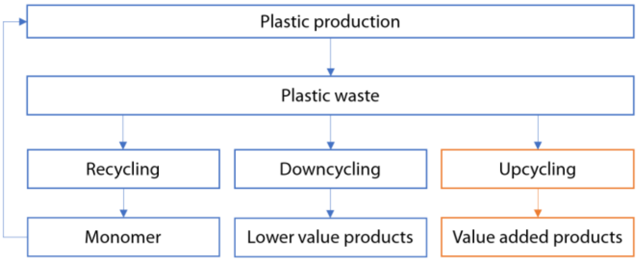

Figure 1 - The pathways of plastic waste management.

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(LHV) of 35.7 MJ kg^{-1},[8] much higher than 26 MJ kg^{-1} for coal and 31 MJ kg^{-1} for petroleum coke.[9] Thus, implementing technology aimed at recovering the energy embedded in discarded plastics could provide a viable and low-cost approach to reduce fossil energy utilization and significantly reduce waste disposal in landfills and oceans. The substantial environmental strain linked to waste plastics, as well as the economic potential in waste recovering technologies, are key drivers that warrant scientific research and technology development focused to convert plastic wastes into value-added products. Several ways to valorize waste plastics have been proposed, with the most common being municipal solid waste combustion for power generation,[10,11] plastic pyrolysis to synthetic crude oil or liquid transportation fuels,[12–15] plastic gasification followed by Fischer-Tropsch (FT) process to produce automotive fuels,[16–18] and plastic conversion to monomers.[19–22] However, the profitability of these technologies is somewhat limited due to the capped sales value of the products (i.e. electricity, liquid fuels) within the framework of the oil market downturn. For the relative lower valued plastic mixture (e.g. thermoplastic polyolefins), it is often not economically convenient to convert them back to monomers (ethylene or propylene). Instead, there is a general interest to convert such waste plastics to higher value products like wax or BTX (benzen, toluene, xylenes), which have a higher market price than crude oil and fuels.[23]

In the present study, we investigated a potential plastic waste management technology which would upgrade waste polyethylenes—high-density polyethylene (HDPE), low-density polyethylene (LDPE), and linear low-density polyethylene (LLDPE)—to high performing lubricant-base oils via catalytic hydrogenolysis.[24,25] The reaction scheme is presented in Figure . The catalyst consists of platinum (Pt) nanoparticles supported on a well-defined $SrTiO_3$ (STO) nanocuboid perovskite surface (Pt/STO). Hydrogenolysis of pre- and post-consumer polyethylenes catalyzed by Pt/STO at 300 °C under H_2 at 11.7 bar in the absence of a solvent yield a high-quality C_{35}-centered liquid product with a narrow dispersity (Đ = 1.06) while producing <1 wt.% of undesired C_1–C_8 alkanes.[26] In contrast, a commercial Pt/Al_2O_3 catalyst produced a liquid with a broader molecular weight range and substantial amounts of C_1–C_8 alkanes.

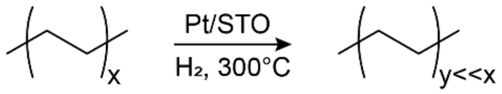

Figure 2 - Reaction scheme for the hydrogenolysis of plastics. STO = $SrTiO_3$.

Given the molecular feature of PE plastic, it would be advantageous to convert it to refinery products with similar C:H ratio. Among the possible products, lubricants attract significant interest as they are one of the most valuable refinery products, as shown in Figure . For 2019, crude oil and transportation fuels were priced \$1.5 gal^{-1} and \$1.3–\$1.7 gal^{-1}, respectively. More valuable chemicals such as BTXs ranged between \$2.0–\$3.2 gal^{-1};[26–28] in contrast, group III and PAO lubricant market price was about \$6–\$10 gal^{-1}.[29] Lubricant is not only a value-added product, but also has a large market size. For example, U.S. refineries produced 65 million barrels of lubricant oil in 2017,[30] with a market greater than 20 billion dollars, and an expected growth of 40–50% by 2035.[31] The much higher price of lubricant products can be related to the complex production process and intensive energy demand. Lubricant mainly consists of base oil and a small amount of additives, commonly in the range of 10–20%. Conventionally, the lubricant base oil is either produced from refining processes (group I, II and III lubricants) or from petrochemical processes, leading to synthetic polyalphaolefins (PAOs, or group IV lubricants). In refineries, lubricants are commonly produced from vacuum distillation bottom cut of the crude oil, which requires extensive energy. The vacuum distillation residue is then processed to lubricant via four steps: solvent deasphalting, solvent extraction to remove aromatics, dewaxing via hydrocracking or solvent extraction, then finally hydrofinishing to remove remaining impurities and aromatics.[32] These

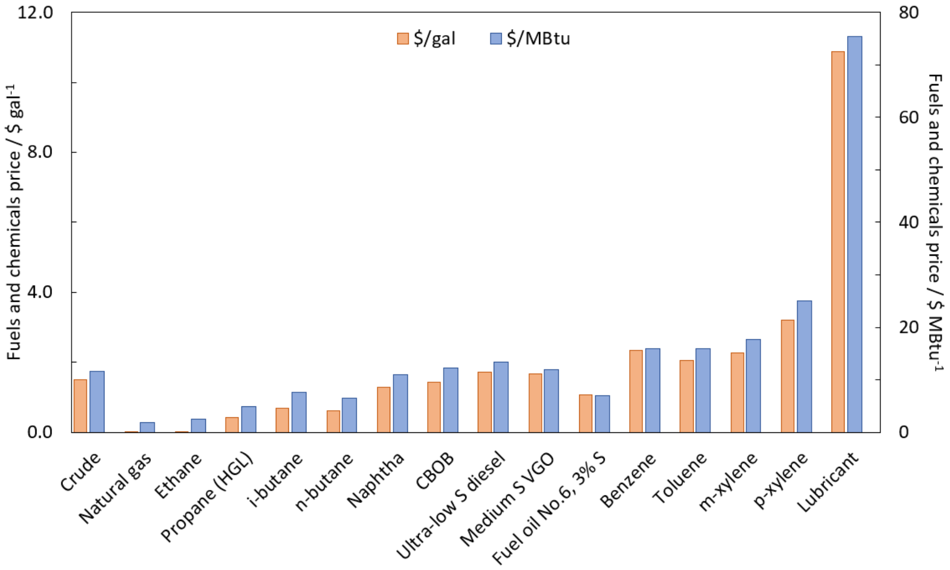

Figure 3 - Price of some U.S. refinery products in 2019.

processes involve intensive energy demand, hydrogen and solvents use. Consequently, for the refinery operation stage, the lubricant production has been estimated to have the highest onsite greenhouse gas (GHG) emissions with about 11.6 g_{CO2e} MJ_{Lub}^{-1}, much higher than the refinery onsite GHG emissions allocated for the production of gasoline (i.e. 7.3 g_{CO2e} $MJ_{Gasoline}^{-1}$) and diesel blendstock (i.e. 4.7 g_{CO2e} $MJ_{Gasoline}^{-1}$).[33] The life cycle GHG emissions are estimated to be 0.98 kg_{CO2e} kg_{Lub}^{-1} for mineral lubricant oil.[34] Meanwhile, the higher performing lubricant, PAO, is produced from petrochemical process using ethylene as feedstock. The high energy consumption associated with ethylene and PAO production processes leads to a high carbon burden, with 1.88 kg_{CO2e} kg_{PAO}^{-1}.[34] Thus, to reduce GHG emissions, it would be of great interest to produce lubricants from a less energy intensive pathway.

Given the potential impact of lubricant production from plastic wastes, using the experimental results tested in laboratories, the present study carried out a techno-economic analysis (TEA) to evaluate the cost of converting waste plastic materials to lubricant products. Additionally, a life cycle assessment (LCA) was conducted to evaluate the life cycle GHG emissions, comparing the results with lubricant production processes from petroleum refinery and petrochemical industry.

Methodology

Laboratory test runs

Hydrogenolysis of polyethylene was performed in a Parr autoclave reactor using 3 g of polymer and varying amounts of Pt/STO catalyst, along with variations in H_2 pressure, temperature, and reaction run time (Figure S3–S5). Detailed information about the reaction condition is reported in the Electronic Supplementary Information (ESI).

Process modelling overview

A conceptual facility to convert 250 MT per day of post-consumer PE waste into lubricant-range molecules was designed, based on lab-scale experimental results.[24,35] The process was modelled with the software Aspen Plus v12 (Aspen Technology Inc.)[36] Two case studies were considered: a high-yield case (HY) based on the best up-to-date test results, and a conservative case characterized by a low-yield (LY). The operating conditions for both cases are listed in Table . The simulated process to convert recovered plastic wastes to high-quality liquids (HQL) consisted of four main process areas. The process scheme is depicted in Figure .

To characterize the plastic feedstock, HDPE (Mw = 35,000 Da) was considered. In Area 100 (A100), four pretreatment lines consisting of washer, dryer, and crusher handled the bales of mixed HDPE before conveying the pelletized feedstock to the reactor. In A200, the hydrogenolysis was carried out in an agitated reactor at 300 °C and 11.7 bar. In the presence of Pt/STO catalyst, the pelletized HDPE was fully converted to HQL and other lighter hydrocarbon products based on the following chemical equations for the low-yield and high-yield cases, respectively:

$$\begin{aligned}(C_2H_4)_{1250} + 528.5H_2 \\ &= 270CH_4 + 90C_2H_6 + 43.3C_3H_8 + 40C_4H_{10} \\ &\quad + 14C_5H_{12} + 13.3C_6H_{14} + 10C_7H_{16} + 5C_8H_{18} \\ &\quad + 42.9C_{35}H_{72} \qquad \text{(Eq. 1)}\end{aligned}$$

$$\begin{aligned}(C_2H_4)_{1250} + 185.7H_2 \\ &= 67.5CH_4 + 22.5C_2H_6 + 10.8C_3H_8 \\ &\quad + 10C_4H_{10} + 3.5C_5H_{12} + 3.3C_6H_{14} \\ &\quad + 2.5C_7H_{16} + 1.3C_8H_{18} + 64.3C_{35}H_{72} \quad \text{(Eq. 2)}\end{aligned}$$

Table 1 - Operating conditions for the low-yield (LY) and high-yield (HY) case studies.

Parameter	LY	HY
Lubricant yield	60%	90%
Catalyst : PE	0.1	0.05
Reaction time / h	72	40
Pt : STO ratio	0.1	0.02
Reaction temperature / °C	300	300
Pressure / bar	11.7	11.7
Feedstock / MT day^{-1}	250	250

Because the catalyst produces a narrowly distributed products,[24] n-$C_{35}H_{72}$ component (LHV = 44.26 MJ kg^{-1}, density = 3 kg gal^{-1}) was used to characterize the properties of the HQL. For the low-yield case, the stoichiometry was obtained considering a 60% yield in $C_{35}H_{72}$ and the fraction of lighter gases as reported in the ESI (Table S4). The plastic feedstock was not available in the Aspen Plus database; hence, physical and thermodynamic properties of interest were implemented in the model. The enthalpy of formation was obtained by modelling the combustion of 1 kg of plastic feedstock while varying its value until the heat of reaction matched the gross heat of combustion reported in the literature (i.e. 47.7 MJ kg^{-1}).[37] The rest of the physical property were calculated by Aspen Plus based on the material molecular weight. The plastic was melted and heated up to 300 °C, with heat generated by the combustion of byproduct light gases produced on site. For the heat of fusion, the value 4.14 kJ mol^{-1} was considered.[38] Hydrogen was used in excess (i.e. 2 times the stoichiometric amount) to aid H_2 adsorption on catalyst surface, promote hydrogenolysis, and reduce mass transfer limitations.[39] In the current process design, the reactor was fed with 680 Nm^3 of hydrogen per MT of PE. The unreacted H_2 was recovered by pressure swing adsorption (PSA). The PSA unit was simulated at 9.2 bar to separate hydrogen from hydrocarbons with an overall recovery ratio of 85%.[40,41] The adsorbed gases were then released after depressurization and diverted to a Joule-Thomson (JT) unit (CPS Inc.),[42] while the recovered hydrogen was compressed to 11.7 bar. The JT unit (Figure S2) separated the hydrocarbons in the range C_3–C_8 from the fuel gas to recover light naphtha (boiling range 50–110 °C) that could be sold at $0.33 kg^{-1}; the price of naphtha was estimated based on its composition.[43] The catalyst was separated from the slurry stream via two Fundabac® (DrM Inc.) candle filters in a parallel configuration.[44] Thereafter, the separated catalyst went through a washing process with hexane. The used hexane was purified in a distillation column in A300, from where it was

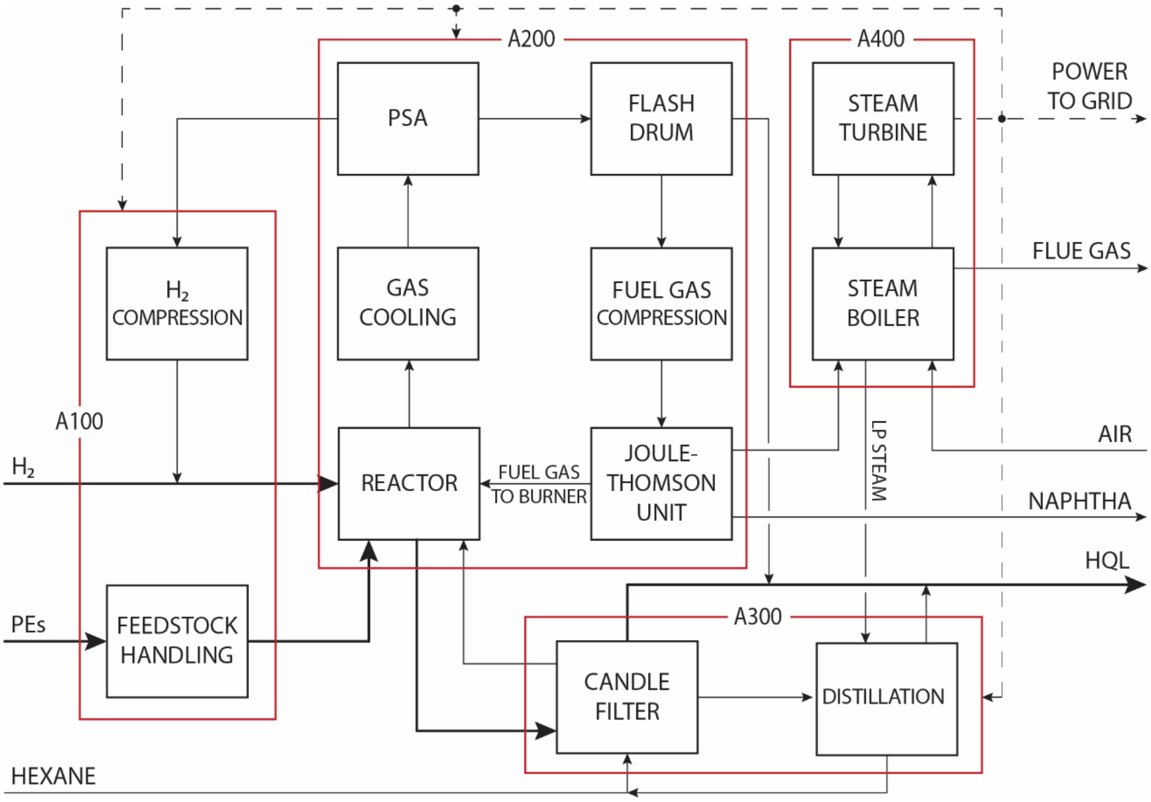

Figure 4 – Block diagram for the conversion process of 250 MT/day of polyolefins. The red boxes highlight the different process areas.

recovered as top product. The hexane loss was 0.5%. Finally, the section A400 contained the steam boiler, turbine, and cooling tower. The temperature of cooling water varied within the range 28–37 °C. The simulation parameters for the main equipment are summarized in Table .

Economic assumptions

The economic analysis was conducted by using the H2A Production Model Framework (version 3.2018), and taking the 2016 U.S. dollar as reference.[45] The major financial input values are listed in Table . The minimum selling price (MSP) was estimated as the price that yields a zero net present value (NPV) when accounting for the total capital investment (TCI), variable cost of production (VCOP), and fixed cost of production (FCOP) during the entire plant construction and operation lifetime. A discount rate of 10.1% was used.

The TCI consisted of total direct capital costs (TDCCs), total depreciable capital costs, and total non-depreciable capital costs. The TDCCs are the sum of the installed equipment costs (converted to the reference year). The installed equipment costs were mostly derived from published equipment costs, which were scaled from the base equipment costs using the following equations:[46]

$$Installed\ cost = Purchase\ cost \times Installation\ factor \qquad (\text{Eq. }3)$$

$$Purchase\ cost = Base\ cost\ \times (Design\ size/Base\ size)^{scaling\ factor} \qquad (\text{Eq. }4)$$

Details on the equipment type, installation factor, and scaling exponent are provided in the ESI (Table S5). The base cost and base size were sourced from the listed references, whereas the new size was calculated from the Aspen Plus model. The total depreciable capital cost included the cost of site preparation (2% of TDCCs), engineering and design (10% of TDCCs), project contingency (15% of TDCCs), and upfront permitting costs (15% of TDCCs).[45] For the total non-depreciable capital cost, the cost of land was estimated as 1.5% of the total depreciable capital costs.[47]

The FCOP accounted for labor cost (LC), general and administrative (G&A) expenses (20% of LC), property taxes and insurance (2% of TCI), and material costs for maintenance and repairs. For the operating labor, 15 operators were considered based on the plant configuration; the detailed calculation of the manning requirement is reported in the ESI. The VCOP consisted of feedstock cost (HDPE and H_2), industrial electricity usage, and non-energy costs (i.e. catalyst, hexane makeup, cooling water, process water).

The prices of the materials are listed in Table . The 2016 price of waste HDPE was estimated at \$323 MT^{-1}, based on the 2019 value for a mixed feedstock of natural (48%, \$0.452 kg^{-1}) and colored HDPE bottles (52%, \$0.284 kg^{-1}), sorted from the material recovery facility (MRF).[48] The price of hexane was assumed to be \$1.1 kg^{-1}, based on commodity price in 2016.[49] The price of H_2, industrial electricity, cooling water, and process water were assumed to be \$1.12 kg^{-1}, \$0.07 kWh^{-1}, \$0.03 m^{-3}, and \$0.63 m^{-3}, respectively, using estimates in the H2A model. Prices for materials and equipment are here presented in USD_{2016}.

Table 2 - Simulation parameters of the main equipment.

Process area	Unit	Aspen block	Parameter / unit	Parameter value
A100	H_2 compressor	*Compr*	Discharge pressure / bar	11.7
			Isentropic efficiency	0.86
A200	Plastic heater	*Heater*	Temperature / °C	300
			Duty / cal s^{-1}	81.59[*]
	Reactor	*Rstoic*	Pressure / bar	11.7
			Temperature / °C	300
			HDPE conversion	100%[**]
	Flash separator	*Flash2*	Pressure / bar	9.5
	Gas chiller	*Heater*	Temperature	43
	PSA	*Sep*	H_2 recovery	85%
			CH_4 recovery	0.1%
A200.JT Plant	Gas compressor	*Mcompr*	Number of stages	4
			Intercooler temperature / °C	43
			Discharge pressure / bar	120
			Isentropic efficiency	0.86
	KO drum	*Flash2*	ΔP / bar	0
			Heat / cal·sec^{-1}	0
	Gas-Liq. heater	*HeatX*	Heat transfer coefficient / $W\cdot m^{-2}\cdot K^{-1}$	63.6
			ΔT_{outlet} / K (countercurrent)	10
	Gas-Gas heater	*HeatX*	Heat transfer coefficient / $W\cdot m^{-2}\cdot K^{-1}$	30
			ΔT_{outlet} / K (countercurrent)	10
	JT valve	*Valve*	ΔP / bar	-115
	Cold separator	*Flash2*	ΔP / bar	0
A300	Catalyst filter	*Sep*	Solid recovery	100%
			Liquid entrainment	1%
	Hexane dist. column	*Distl*	Number of stages	15
			Feed stage	2
			Reflux ratio	0.5
			Distillate:Feed	0.99
			Pressure / bar	1
A400	Boiler[†]	*Heater #1*	Water T_{out} / °C	300
		Heater #2	Degree superheating	0
		Heater #3	Steam T_{out} / °C	550
		Heater #4-6	ΔP / bar	0
	Air blower	*Compr*	ΔP / bar	0.3
			Isentropic efficiency	0.86
	Steam turbine	*Compr*	Discharge pressure / bar	0.04[††]
			Isentropic efficiency	0.86
	Water condenser	*Heater*	ΔP / bar	0
			Vapor fraction	0
	Water pump	*Pump*	Discharge pressure / bar	112
			Efficiency	0.78

[*] Based on the heat of fusion of HDPE.

[**] For fractional conversions, consult ***Error! Reference source not found.***.

[†] The boiler was made of an economizer, vaporizer, and superheater, each heater modeled as a couple of heater blocks to describe the water/steam side and the hot gas side.

[††] Obtained from Calculator, based on the water saturation pressure at the outlet conditions (T = 30°C).

Table 3 - Economic assumptions and materials' price.

Financial Values	
Reference year	2016
Plant life / years	40
Analysis period / years	43
Length of construction period / years	3
Capital spent in 1st year of construction	8%
Capital spent in 2nd year of construction	60%
Capital spent in 3rd year of construction	32%
Start-up time / years	1
Depreciation schedule length / years	20
Depreciation type	MACRS*
Equity financing	40%
Interest rate on debt	3.70%
Debt period / years	Constant
FCOP during start-up	75%
Revenues during start-up	50%
VCOP during start-up	75%
Decommissioning costs (% of TDCC)	10%
Salvage value (% of TCI)	10%
Inflation rate	1.9%
After-tax real IRR	8.0%
State taxes (national average)	6.0%
Federal taxes	21.0%
Total tax rate	25.74%
Working capital (% of FCOP)	15%
Materials Prices	
Hydrogen / \$ kg^{-1}	1.13**
Catalyst (2% Pt) / \$ kg^{-1}	834
Catalyst (10% Pt) / \$ kg^{-1}	3470
Waste HDPE / \$ MT^{-1}	323
Hexane / \$ kg^{-1}	1.10
Electricity / \$ kWh^{-1}	0.07
Process water / \$ m^{-3}	0.63
Cooling water / \$ m^{-3}	0.03

*Modified Accelerated Cost Recovery System, as published by IRS.
**For H_2 produced via steam methane reforming (SMR).

Pt/STO price estimation. The price of the catalyst was a major factor impacting the total lubricant production cost. Given the absence of a commercial catalyst as it was prepared in-house,[35] the price of Pt/STO was estimated for two options: purchase and lease, by using the following equations:[50]

$$C_{C,P} = C_M + Pt_{load} \cdot C_{Pt}\,(1 + r_S) \qquad \text{(Eq. 5)}$$

$$C_{C,L} = C_M + Pt_{load} \cdot C_{Pt}\,(r_S + r_L) \qquad \text{(Eq. 6)}$$

Here, $C_{C,P}$ and $C_{C,L}$ are the costs of supported catalyst for purchasing (\$ kg_{Cat}^{-1}) and leasing (\$ kg_{Cat}^{-1}), respectively. C_M is the manufacturer fee (\$ kg_{Cat}^{-1}), Pt_{Load} is the mass ratio of metal to support, and C_{Pt} is the platinum metal cost. Additionally, r_S is the surcharge fee, while r_L is the leasing rate. The manufacture cost and surcharge fee were kindly provided by catalyst vendor Johnson Matthey (London, UK).[51] Note that the manufacturer cost includes processing and support's costs. The latter was based on γ-Al_2O_3, which was used as surrogate to estimate the cost of nano material perovskite STO, that is not yet developed at the commercial scale. Given the supported catalyst cost is led by Pt metal cost, using the cost of Al_2O_3 support was expected to have a small impact on the catalyst cost estimation.

The leasing rate, r_L was considered at 9%, offering a rate-of-return which is risk-adjusted and high enough to justify the opportunity costs of the lender's capital investment. Equation 7 provides the formula to define the components for the leasing rate.[50]

$$r_l = r_{borrow} + r_{rec} + r_{op} + r_{risk} \qquad (Eq. 7)$$

Here, $\boldsymbol{r_{borrow}}$ is the premium accounting for lender's financing cost; $\boldsymbol{r_{rec}}$ corresponds to the premium associated with recovery of spent Pt metal; $\boldsymbol{r_{op}}$ is the premium accounting for lender's overhead expenses, including the operation and credit loss; $\boldsymbol{r_{risk}}$ is the premium associated with price risk incurred by market volatility, this premium functions as a hedging mechanism used by investors to help shield them from the price of owning the metal.

Life Cycle Assessment (LCA) by using GREET model

A LCA based technique was applied to estimate the carbon footprint for lubricant synthesis process from plastic waste, using data from GREET 2020 (Greenhouse gases, Regulated Emissions, and Energy use in Technologies model)[9]. GREET provides the inventory of data for a variety of processes with detailed information of energy use, material consumption and the resulting emissions.

The system boundary used for the LCA is shown in Figure , which encompasses the process scheme shown in Figure plus the

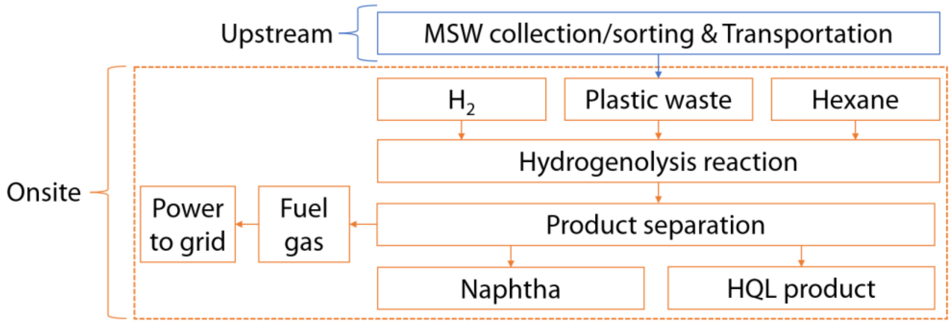

Figure 5 – Boundaries for the HQL process.

materials consumption, energy use, and emissions for collection, separation, and transportation of MSW to an MRF, thereby constituting a cradle-to-gate LCA. The emissions profile reported in this study was generated on a 1 kg of HQL basis. To quantify the potential environmental benefit of converting plastic waste to HQL lubricants, the conventional processes for group III and PAO lubricants were taken as comparative baselines.[34]

Furthermore, it was important to assess the total reduction in solid waste materials as a part of the environmental analysis. For this estimation, the net amount of solid waste reduction was calculated accounting for the plastic used as raw material and the average catalyst support material disposed, which was calculated from the total amount of supported catalyst used over the lifetime of the plant.

Results and discussions

Process modeling results

The total material and energy streams are listed in Table . More detailed information on the major material and energy flows are listed in Table S1, S2. The scenario characterized by a lubricant yield of 60% led to a HQL production of 153 MT day^{-1} and about 46 MT day^{-1} of naphtha. Increasing the lubricant yield from 60% to 90% led to a 50% increase in lubricant production. The case with 90% yield was also characterized by a lower amount of hydrogen consumption per kg of HQL, due to the lower production of light hydrocarbons, the formation of which requires more H_2 than liquid products. Moreover, the on-site CO_2 emissions decreased by 78% due to the lower amount of fuel gas burnt for power generation. The amount of co-product decreased to about 8 MT day^{-1}, as well as the electric power produced, that decreased by 67%.

The energy balance is summarized in Table . The process energy efficiency was calculated based on the energy of the product, co-product, and feedstock materials:

$$Efficiency = \frac{E_{HDPE} + E_{H_2} + W_{e,IN}}{E_{HQL} + E_{naphtha} + W_{e,OUT}} \qquad (\text{Eq. 8})$$

Here, *E* is the energy content of feedstock and products based on the low heating value (LHV), while W_e is the electric power needed (*IN*) and generated (*OUT*). For the 90% yield case, both the increase in HQL productivity and decrease in hydrogen demand led to a more efficient process overall, with an energy efficiency of 89%.

Economic analysis results

Catalyst cost from purchase and leasing. For purchasing Pt/STO, the estimated cost would be \$3474 kg_{Cat}^{-1} and \$834 kg_{Cat}^{-1} for a Pt content of 10% and 2%, respectively. In the purchase scenario, the first batch of catalyst was accounted for as capital expenditure, while the future purchase of catalyst throughout the plant life was counted as variable operating costs.

Table 4 - Summary stream table for the two scenarios. The catalyst consumption rate was calculated based on the full replacement of the catalyst every three years during the total life of the plant (i.e. 40 years).

		LY	HY	unit
Input	HDPE	1.64	1.10	Kg kg_{HQL}^{-1}
	H_2 makeup	0.0562	0.02	Kg kg_{HQL}^{-1}
	Air	7.80	1.92	Kg kg_{HQL}^{-1}
	Hexane makeup	0.004	0.002	Kg kg_{HQL}^{-1}
Output	HQL	153	227	MT day^{-1}
	Naphtha	0.304	0.034	Kg kg_{HQL}^{-1}
	Flue gas	8.19	2.01	Kg kg_{HQL}^{-1}
CO_2 in flue gas		1.08	0.24	Kg kg_{HQL}^{-1}
Cooling water		378	117	Kg kg_{HQL}^{-1}
Process water		0.71	0.20	Kg kg_{HQL}^{-1}
Catalyst consumption rate		4.70 10^{-4}	8.81 10^{-5}	Kg MT_{HQL}^{-1}

Table 5 - Energy balance and process efficiency for both low-yield and high-yield cases.

		LY	HY	unit
Input	HDPE	128	128	MW
	H_2	11.9	6.13	MW
	Power	5.83	4.29	MW
Output	HQL	78.3	116.2	MW
	Naphtha	24.6	4.06	MW
	Power	11.2	3.74	MW
Efficiency		78%	89%	

For the leasing option, the cost would be \$587 kg_{Cat}^{-1} and \$257 kg_{Cat}^{-1}, for a Pt content of 10% and 2%, respectively. In the cash flow analysis, the lease cost was accounted for as fixed operating cost.

Lubricant production cost. The normalized cost breakdown is shown in Figure . For each scenario, two options were analyzed. In one case, the catalyst needed throughout the life plant was purchased (LY-P, HY-P); the second option was based on the leasing of catalyst (LY-L, HY-L). The production cost for the LY-P case was \$1.98 kg^{-1} (\$5.96 gal^{-1}), i.e., with 60% lubricant yield, catalyst:HDPE = 1:10, and a reaction time of 72 h. For this case, the major expenses were the feedstock cost and variable operating costs, mainly due to the catalyst which accounted for the 34% of the final production cost. Leasing the catalyst proved

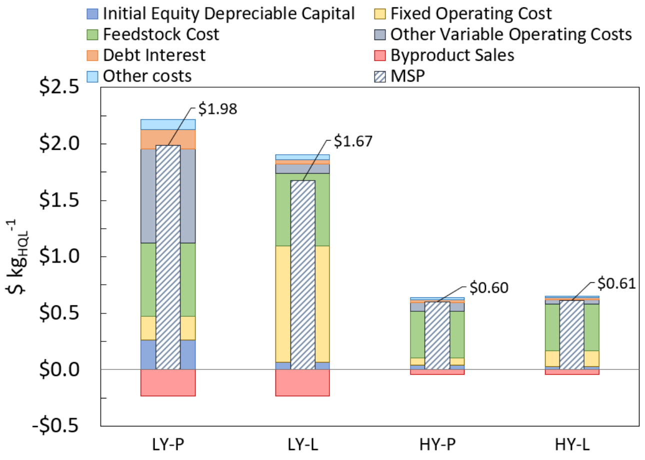

Figure 6 - Cost breakdown for low-yield and high-yield scenarios.

to be a more profitable option at the lower yield (LY-L case), leading to a 16% decrease in the MSP. In this scenario, the leasing expenses were accounted for as FCOP, which constituted the 54% of the total cost of production.

The last two case studies at higher yield were characterized by a lower MSP. Lowering the catalyst-to-feedstock ratio from 1:10 to 1:20 and reducing the Pt load cut the cost by 70%, when LY-P and HY-P were compared. At higher HQL yield, less hydrogen is needed per amount of PE converted, which led to lower feedstock cost (-36%). Furthermore, lower gas streams led also to overall cheaper equipment, especially in the energy generation section of the plant (see Table for equipment costs). At high yield, leasing the catalyst did not improve the economics of the process by assuming a 9% leasing rate, which varies with the borrower's background and financials. A further decrease in the catalyst leasing rate could make the leasing option more attractive, and with a 2.1% leasing rate the two options would result in the same MSP. Capital expenditures and operating costs are listed in Table Table .

Sensitivity analysis. To assess some of the uncertainties related to the assumptions made or to the plant capacity fluctuations, a sensitivity analysis was done to monitor the variation of MSP.[52] In Figure , the findings from the sensitivity analysis are shown for the HY-P scenario. At higher yield and lower catalyst amount, the feedstock cost becomes a major element in the HQL MSP (65%, as reported in Figure); therefore, fluctuations in the plastic waste price would cause considerable fluctuations in the MSP.

For the operating conditions considered in this study, hydrogen had a very low impact on the HQL production cost. This is because of the relativately low flow rate of fresh hydrogen needed—especially for the high yield scenario. Furthermore, the cost of H_2 from conventional steam methane reforming was taken into account. If low-carbon hydrogen had been used, the MSP would have been higher, as it can be seen in Figure . By using green hydrogen, the cost of production would increase up to 13%; however, the conversion of plastic waste to HQL would still be profitable given the high market price of lubricants with comparable properties.

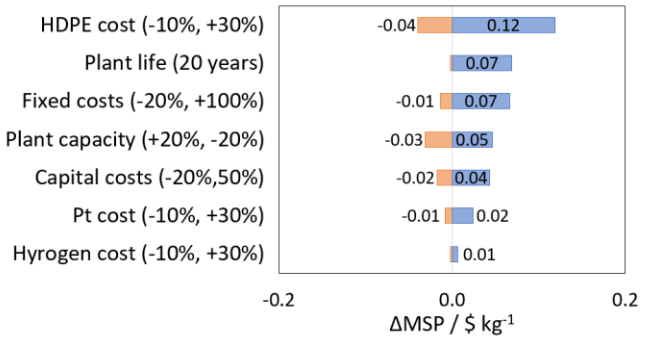

Figure 7 - MSP variation based on the HY-P case study, for which MSP = \$0.60 kg_{HQL}^{-1}.

Comparison with MRF and other conversion technologies

The designed facility to convert plastic waste to lubricant product can be standalone or integrated with an MRF. For the former, the analysis here presented has demonstrated that the facility is likely to be profitable given the high product selling price. For the latter, a case study was conducted to estimate the additional profit for an MRF to upgrade sorted plastic waste to lubricant product. The bottom line of the decision to upgrade the waste plastic is whether the margin of converting plastic waste to lubricant is higher than the gross margin from directly selling the sorted plastics. In a recent feasibility study for a single stream MRF in Iowa, operating costs were estimated to be around \$230 MT^{-1} of plastic waste.[53] Considering a feedstock with the same composition as in our case study (i.e. 48% natural HDPE, 52% colored HDPE, sold at \$323 MT^{-1}), and assuming all HDPE were sold as sorted plastic bottles, the gross profit would be \$93 MT_{PE}^{-1} (i.e. \$323 MT^{-1} – \$230 MT^{-1}).

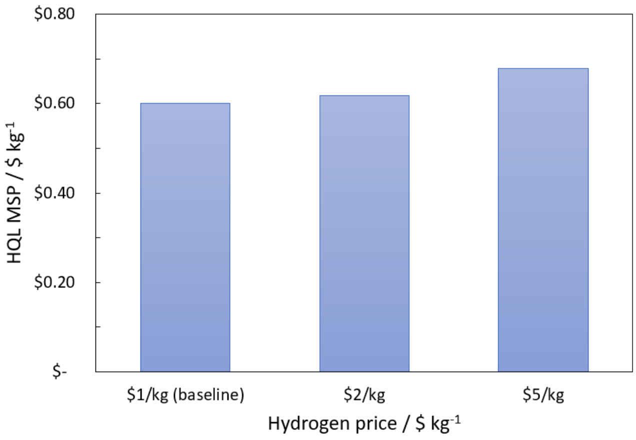

Figure 8 – Cost of production of HQL as a function of the hydrogen price.

The HQL process presented in this study was characterized by a production cost of \$600 MT_{HQL}^{-1} with a productivity of 0.90 MT_{HQL} MT_{HDPE}^{-1} for the HY case. Therefore, by considering a market price in the range \$1.56–\$3.12 kg_{HQL}^{-1} (i.e. \$4.7–\$9.4 gal_{HQL}^{-1}), the gross profit would vary between \$870 MT_{HDPE}^{-1} and \$2290 MT_{HDPE}^{-1}, making the plastic upgrade a more profitable process. The profit was calculated as:

$$Gross\ profit\ /\ \$\ MT_{PE}^{-1} = (HQL\ price - MSP) \cdot MT_{HQL}/MT_{PE} \qquad (Eq.9)$$

All the prices were reported in USD_{2016}. The effect of the lubricant price on the HQL process profitability can be seen in the Figure . For a lubricant price higher than \$0.7 kg^{-1}, upgrading the plastic waste to HQL would be characterized by a higher gross profit than selling the sorted plastic waste.

Similarly, conversion of waste plastics to HQL would be more advantageous even compared to conversion to fuel products. For example, a recent study reported a MSP of \$0.56/gal for the fuel produced from plastic waste pyrolysis.[15] For a pyrolysis plant with a capacity of 66 gal of fuel per MT of plastic waste, the gross profit would then be around \$115 per MT of plastic waste. The 2016 average price for diesel (i.e. \$2.3/gal) was considered for the calculation.

Given its tribological properties close to PAOs, the HQL from PEs waste could likely be sold at prices higher than \$0.7 kg^{-1}; therefore, upgrading the plastic feedstock to non-fuel chemicals has a much higher economic potential than just sorting the plastic feedstock or converting it to fuel.

Table 6 - Direct and indirect capital investment for the different scenarios. All costs are reported in USD_{2016}.

	LY-P	LY-L	HY-P	HY-L
A100	$ 10,212,770	$ 10,212,770	$ 10,332,680	$ 10,332,680
A200	$ 24,378,110	$ 24,378,110	$ 14,783,670	$ 14,783,670
A300	$ 2,339,140	$ 2,339,140	$ 2,570,420	$ 2,570,420
A400	$ 23,804,950	$ 23,804,950	$ 14,047,920	$ 14,047,920
Total direct capital cost	$ 60,734,970	$ 60,734,970	$ 41,734,690	$ 41,734,690
Site Preparation	$ 1,214,700	$ 1,214,700	$ 834,690	$ 834,690
Engineering & design	$ 6,073,500	$ 6,073,500	$ 4,173,470	$ 4,173,470
Project contingency	$ 9,110,250	$ 9,110,250	$ 6,260,200	$ 6,260,200
Catalyst first fill fee	$ 271,470,690	-	$ 18,102,900	-
Up-Front Permitting Costs	$ 9,110,250	$ 9,110,250	$ 6,260,200	$ 6,260,200
Land Cost	$ 3,577,140	$ 862,440	$ 773,660	$ 592,630
Total indirect capital costs	$ 300,556,520	$ 26,371,120	$ 36,405,140	$ 18,121,200
Total capital costs	$ 361,291,490	$ 87,106,090	$ 78,139,830	$ 59,855,890

Table 7 - Operating costs (USD_{2016}/year) for the different scenarios.

	LY-P	LY-L	HY-P	HY-L
Catalyst leasing	-	$ 45,830,490	-	$ 5,567,460
Labor	$ 1,801,490	$ 1,801,490	$ 1,801,490	$ 1,801,490
G&A	$ 360,300	$ 360,300	$ 360,300	$ 360,300
Property taxes and insurance	$ 7,225,830	$ 1,742,120	$ 1,562,800	$ 1,197,120
Maintenance and repairs	$ 1,049,010	$ 1,049,010	$ 1,049,010	$ 1,049,010
Total fixed operating costs	$ 10,436,630	$ 50,783,410	$ 4,773,600	$ 9,975,370
Feedstock	$ 31,853,270	$ 31,853,270	$ 30,257,600	$ 30,257,600
Electricity	$ 3,215,470	$ 3,215,470	$ 2,369,450	$ 2,369,450
By-products credits	$ (11,279,570)	$ (11,279,570)	$ (2,900,090)	$ (2,900,090)
Catalyst recovery	$ 36,974,960	-	$ 3,027,700	-
Water (process, cooling)	$ 577,030	$ 577,030	$ 257,380	$ 257,380
Total variable operating costs	$ 61,341,150	$ 24,366,190	$ 33,012,030	$ 29,984,330
Total operating costs	$ 71,777,780	$ 75,149,600	$ 37,785,620	$ 39,959,700

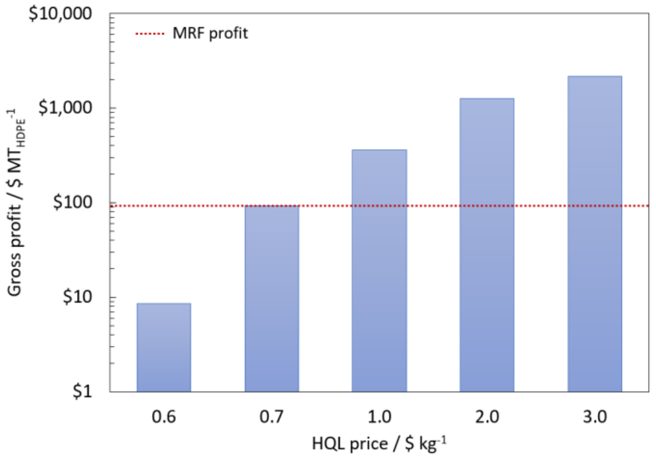

Figure 9 – Effect of the HQL price to the gross profit for waste plastics upgrading to HQL.

LCA results

The CO_2 emissions associated with the production of plastic-derived lubricants are a vital parameter to ensure the environmental benefit synergy between plastic waste and air emission reductions. This section provides results for the emissions profile of the HQL process, a discussion on petroleum lubricant emissions as a baseline, and the sensitivity case of conventional PAO lubricant production process.

Plastic waste reduction by upcycling. The process for conversion of plastics to HQL product used 91,250 MT year^{-1} of HDPE feedstock. From a solid material balance viewpoint this corresponds to the total input solid HDPE that is converted to HQL. In this process, the only solid waste generated over the lifetime of the plant comes from catalyst support that is disposed, since the metal, Pt, is recovered owing to its high cost and value. Therefore, based on the amount of support catalyst generating the sole source of solid waste, on a yearly average of 6.63 MT year^{-1}, this process is likely to bring about a reduction in solid waste by 99.9%.

A conventional process for mineral lube production is estimated to yield an average of 40 MT of solid waste, in the form of clay.[54] Compared to this value, the current process accounts for over 83% reduction in plastic waste. It is important to point here that along with this reduction in plastic waste, the lubricant product obtained from the process in this research is of a higher quality than mineral lube.[35]

Emissions from the plastic waste derived lubricant. Table provides the onsite CO_2 emissions produced from the combustion of fuel gas. In the LY case study, the combustion of fuel gas generated a surplus of power, which was accounted as power injected into the grid. The HY case produced nearly 80% less CO_2 from power generation, due to the lower amount of light hydrocarbons produced from the HDPE hydrogenolysis.

Table 8 - Carbon emissions generated by the HQL process from combustion of light gases.

LY	HY	Unit
1.08	0.24	$kg_{CO2}\ kg_{HQL}^{-1}$
3.25	0.72	$kg_{CO2}\ gal_{HQL}^{-1}$

The cradle-to-gate GHG emissions of the lubricant production from plastic waste was calculated by accounting for the upstream burdens of feedstocks and other material and energy inputs. In Figure , the total GHG emissions for each scenario are reported. The lubricant yield has a significant impact not only on process economics, but also on GHG emissions. The use of H_2 from the emission-intensive industrial steam methane reforming process has a significant impact on GHG emissions, for both L-Y and H-Y case. Therefore, we also considered additional scenarios with green H_2 (from solar electrolysis) and low carbon H_2 (from nuclear electrolysis). Herein, “Onsite” refers to onsite emission due to combustion; “Naphtha” refers to a naphtha to naphtha displacement credit based on the naphtha wells-to-gate (WTG) burden as obtained from GREET; “Electricity” refers to electricity consumption or displacement credit which varies between different cases; “PE waste” refers to the emissions associated with collection and transportation of PE waste as the PE burden (associated with PE production) is set to zero owing to its waste feature. Upstream emissions of hexane and the catalyst were omitted from the chart as they accounted for less than 0.2% of the total emissions. The HQL product was characterized by well-to-gate (WTG) emissions equal to 1.17 $kg_{CO2e}\ kg_{HQL}^{-1}$ (3.52 $kg_{CO2e}\ gal_{HQL}^{-1}$) and 0.48 $kg_{CO2e}\ kg_{HQL}^{-1}$ (1.44 $kg_{CO2e}\ gal_{HQL}^{-1}$) for the LY and HY cases, respectively. For both scenarios, most of the emissions came from H_2 upstream emissions (35% for LY and 42% for HY) and CO_2 from gas combustion onsite (63% and 48%). If low-carbon H_2 were used instead of gray H_2, a further decrease of 34% and 35% (compared to the HY case) would be achieved for nuclear and solar H_2, respectively.

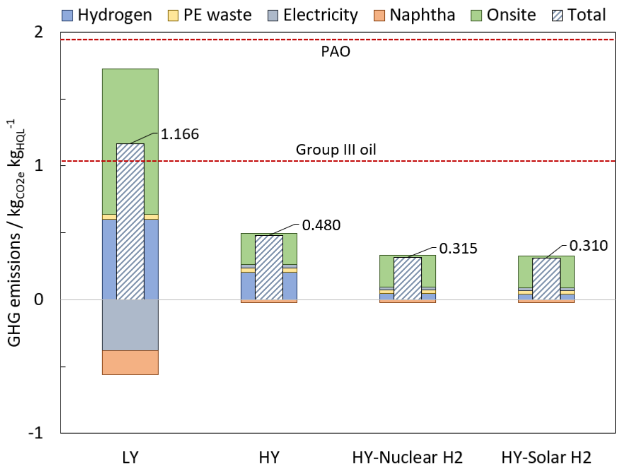

Figure 10 - Breakdown of the total GHG emissions of the HQL process for LY and HY cases. For the latter, the GHG emissions were also evaluated in the case of H_2 from nuclear and solar energy.

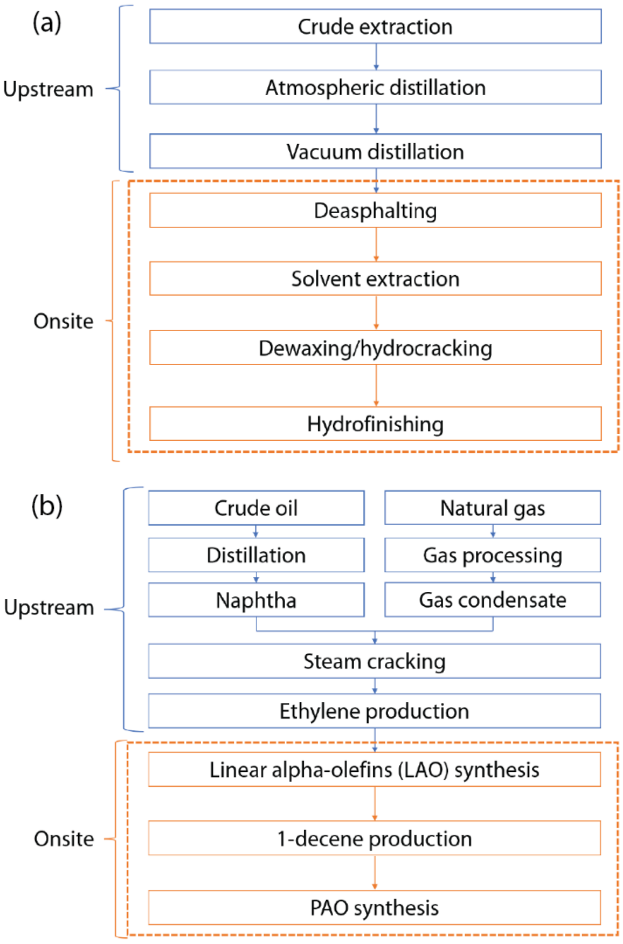

Figure 11 - Block diagram highlighting the LCA boundaries for mineral lubricant (a) and PAO (b) production processes.

Petroleum lubricant well-to-gate CO_2 emissions as a baseline. Figure provides a general scheme for the industrial production of lubricant base oils from crude oil (a) and PAO (b). The production process can be divided into an onsite and an upstream fraction. The processing steps up to vacuum distillation are usually considered upstream of the actual onsite lubricant synthesis process. It is therefore important to point up that the emissions associated with the onsite production of lubricants takes into consideration the burden arising due to the use of heat, electricity and fuel needed for the various onsite processes, as shown in Figure .

Despite the existence of some energy consumption data for lubricant product, the CO_2 emission has been rarely reported, as most studies focused on the refinery emissions allocated to major products, such as gasoline, diesel, jet, naphtha, pet coke, etc. The CO_2 emission is highly depended on refinery crude slates, refinery product slates, and the operating conditions of the refinery. We conducted a unit-based process analysis to obtain a detailed breakdown of energy sources, in order to obtain CO_2 emission by using emission factors of various energy sources. In addition, we developed a detailed input inventory list, to calculate the upstream burden for lubricant production.

The results presented here consolidate a baseline for the lubricant production process calculated from available literature. Table shows sources from the literature which provided a comparison of emissions data for the current technology to literature available for emissions associated with lubricant production. According to the energy and environmental profile for US refineries, the total fuel and electricity consumption for lubricant production is reported to be 11.26 MJ kg_{Lub}^{-1} and 0.5 MJ kg_{Lub}^{-1}, respectively.[55] Other sources reported fuel and electricity use at 11.56 MJ/$kg_{Lub.}$ and 0.06 MJ kg_{Lub}^{-1}, respectively.[33] Best Available Techniques (BAT) for refining of mineral oil and gas, a European Commission report, also provides fuel and electricity use at 6.07 MJ kg_{Lub}^{-1} and 0.21 MJ kg_{Lub}^{-1}, respectively. The corresponding onsite emissions for lubricant production fall in the range of 0.89–1.35 kg_{CO2e} kg_{Lub}^{-1}.[56]

Table 9 - Energy use and GHG emissions for conventional lubricant production. All the values refer to 1 kg of lubricant.

Lubricant type	Fuel / MJ kg^{-1}	Electricity / MJ kg^{-1}	Onsite GHG / kg_{CO2e} kg^{-1}	WTG GHG / kg_{CO2e} kg^{-1}	Ref
Group III	11.3	0.50	0.67	1.10	55
Group III	-	-	-	0.98	34
PAO	-	-	-	1.88	34

Synthetic lubricant (PAO) emissions profile. The total onsite energy input of the process includes power, heat, and steam. The energy consumption for PAO production is 2.37 MJ kg_{Lub}^{-1} and 9.60 MJ kg_{Lub}^{-1}, for power and thermal components, respectively; the total emissions is reported at 1.88 kg_{CO2e} kg_{Lub}^{-1}.[34]

As seen in Figure , the process for HQL production has GHG emissions in the range of 0.48–1.2 kg_{CO2e} kg_{HQL}^{-1}, for a cradle-to-gate basis, considering all onsite and upstream emissions. Therefore, on a cradle-to-gate framework, mineral lube production would lead to nearly twice the amount of CO_{2e} per kg of lubricant produced, and PAO production would emit about 4 times more GHGs.

Waste plastics upcycling is a field with high potential for waste-to-value generation and has as such garnered active interest from researchers. A variety of high-value products such as fuels, construction materials, nanomaterials, hydrogen, and lubricants have been obtained using pre-consumer and waste plastics alike, thereby developing novel waste-to-value routes.[21,57,58] In terms of emissions, with the validated 90% yield, the HQL synthesis from HDPE led to 0.48 kg_{CO2e} kg_{HQL}^{-1}. This represented a 52% reduction in the WTG GHG emissions relative to group III lubricant oil and a 74% reduction respect to PAO, as well as 99.9% of plastic waste reduction. A further decrease in the GHG emissions (-35%) would be reached if low-carbon H_2 were used.

Conclusions

A process to upgrade waste polyolefins to high-quality liquid was described. This process can tackle the plastic management issues while being economically profitable with an estimated MSP of the lubricant-like product of \$0.6 kg_{HQL}^{-1} (\$1.8 gal_{HQL}^{-1}), considering the market price for group III and PAO lubricants in the range \$6–\$10 gal^{-1}. Given the high market price of the final product, the profitability of the process would not be greatly impacted even in the case of H_2 produced by low-carbon sources (e.g. nuclear or solar energy). The main parameters that affected the production costs were the cost of plastic waste and the amount of Pt/STO catalyst needed.

The process for lubricant production would have enormous environmental and national energy security benefits. For the former, our proposed technology has extraordinary benefit in plastic waste reduction in landfill and in waterways, without producing and disposing of solid char. This is especially important for the U.S. as the waste export market continues to dwindle as foreign countries ban the importing of waste. For the latter, the process could potentially reduce GHG emissions significantly compared to refineries by displacing the energy intensive lubricant production process. The LCA showed that lubricant produced from plastic waste had GHG emissions as low as 0.31 kg_{CO2e} kg_{Lub}^{-1} (i.e. when low-carbon H_2 was used) which was significantly lower than the emissions from petroleum lubricant (1–1.88 kg_{CO2e} kg_{Lub}^{-1}). Based on the U.S. lubricant production of almost 61 million barrels in 2019,[59,60] replacing petroleum-derived lubricant with plastic waste-derived lubricant could reduce CO_2 emissions between 3 and 9 million MT per year in the U.S.

Author Contributions

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Guiyan Zang: Process modeling.
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Ryan Hackler: Experimental data.
Hernan E. Delgado: TEA, writing (review).
Massimiliano Delferro: Experimental data, writing (review).
Amgad Elgowainy: LCA review.
Theodore Krause: Process review.

Conflicts of interest

The authors declare the following competing financial interest: a provisional patent application partially based on this work has been filed.

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