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Cite this: DOI: 10.1039/d5su00910c

An integrated techno-economic analysis and life cycle assessment of lube oil production from post-use polypropylene and comparison with conventional base oils

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Received 8th December 2025
Accepted 14th March 2026

DOI: 10.1039/d5su00910c

rsc.li/rscsus

This study presents an integrated analysis of the technical, economic, and environmental performance of upcycling post-use polypropylene (PU-PP) through hydrogenolysis, using a platinum strontium titanate ($Pt/SrTiO_3$) catalyst. This process generates lubricating oils from upcycled plastics, also referred to as LOUPs. The refining of Group I, II, and III base oils from crude oil is also evaluated as a conventional process for comparison with LOUPs production. The minimum selling price (MSP) of LOUPs was estimated at \$2.18 per gallon, representing reductions of 16%, 49%, and 61% compared to the MSPs of conventional Group I, II, and III base oils, respectively. The greenhouse gas (GHG) emissions from LOUPs production ranged between 0.6 and 0.7 kg CO_2e per kg. These emissions are up to 40% higher than those of Group I base oil but 62% and 74% lower compared to Group II and Group III base oils, respectively. Hydrogenolysis also resulted in lower GHG emissions compared to conventional end-of-life (EOL) management of PU-PP. According to these findings, the analysis presented showed the cost-effectiveness and environmental benefits of producing LOUPs from PU-PP in comparison to conventional Group II and III base oil refining.

Sustainability spotlight

This study provides a comprehensive assessment of the technoeconomic and life cycle analysis of converting PU-PP into LOUPs *via* hydrogenolysis. The estimated MSP of LOUPs is 16%, 49%, and 61% lower than the MSPs of conventional Group I, Group II, and Group III base oils, respectively. The findings indicate that GHG emissions are up to 40% higher than those of Group I base oil but are 62% and 74% lower than those of Group II and Group III base oils, respectively. The analysis offers key insights into how the implications of the proposed technologies compare to the conventional pathway and highlights the most significant process variables to develop hydrogenolysis recycling technologies for PU-PP, as a basis for upcoming assessments.

Introduction

Globally, the production of plastics has been increasing due to their widespread use in various sectors such as agriculture, construction, packaging, the automotive industry, and electrical equipment manufacturing.[1] Global plastic production reached 367 million tonnes in 2020, representing a 25% rise compared to 2010. If current production and consumption trends continue, plastic production could surpass one billion tonnes by 2050.[1] However, improper disposal of post-use plastic (PUP) poses a serious threat to the environment and human health, leading to soil and groundwater pollution. Currently, only 10–12% of global PUP is properly recycled,[2] while the remainder is either landfilled, incinerated, or left untreated and mismanaged. In the United States, more than 75% of the plastic waste is sent to landfills, with only 8.7% being recycled.[2,3]

The incineration and open burning of PUP have intensified environmental issues by releasing air and waterborne pollutants,[4] while mechanical recycling processes are constrained by the production of recycled resin that exhibit limited performance and applicability, due to the accumulation of additives and the reduction of physicochemical properties over multiple recycling cycles.[5,6] These limitations reduce the reintegration of recycled resins into the supply chain, which enforces a linear economy. Therefore, transitioning from the current linear framework to a supply chain that fully valorizes PUP products requires the promotion of advanced recycling technologies alongside conventional mechanical recycling. Advanced recycling involves technologies such as pyrolysis, gasification, methanolysis, and enzymatic hydrolysis. These solutions

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represent an opportunity to address PUP management, especially for post-use polyolefins (PU-POs) such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP). By leveraging these resources with high energy and carbon content, advanced recycling can convert PUP into high-value products, which could create new economic incentives that are incorporated into existing distributed waste management infrastructure and workflows.

Among polyolefins, PP is consistently used in various products in daily life, with applications in packaging, textiles, automotive components, and various consumer products.[7] PP follows a linear production flow, starting with the raw material extraction and finishing with waste generation and disposal, with only 1% being recycled. Approximately 50 million tonnes of PP were made in 2015, a number that continues to rise due to its versatility.[2] Although this growing production raises concerns about the increased generation of PU-PP, recycling of this material presents opportunities for lessening these environmental challenges.[7] Utilizing PU-PP to produce valuable products appears to offer an economically attractive solution for leveraging this material.[7]

The key approach to reduce plastic contamination is the chemical conversion of PUP into monomers and liquid products.[8] Many studies have explored the pyrolysis of virgin PP and waste PP, either as a single stream or in plastic blends.[7–15] Pyrolysis is a traditional chemical recycling and upcycling method to produce gaseous and liquid products from PU-POs.[16] However, this method involves high temperatures (>400 °C), and the reaction is generally endothermic, which increases the process energy costs and lowers the product selectivity. In contrast, the catalytic hydrogenolysis reaction is mostly exothermic, operates at a significantly lower reaction temperature of approximately 300 °C, and converts PU-POs to obtain a narrower product distribution of desirable linear alkane products, such as light alkanes, diesel fuel, and wax, compared to the pyrolysis process.[15,17]

Commercial lubricants are comprised of base oil, which provides most of the lubricating properties. However, for specific applications, additives are used to extend the stability and longevity, as well as to regulate the friction and wear performance of the lubricant.[18] Globally, nearly 35 million tonnes of lubricants are used every year, the bulk of which is produced from petroleum-based distillates, and a growing percentage is synthetic hydrocarbons.[17,19] In 2024, the global lubricants market was valued at 144.4 billion U.S. dollars (USD), which is projected to reach 180.2 billion USD by 2030, expanding at a compound annual growth rate (CAGR) of 3.8% from 2025 to 2030.[20] This market is primarily driven by automotive engine oils, industrial metalworking fluids, and hydraulic oils.[21] These molecules can be obtained by the conversion of polyolefins, offering a promising upcycling strategy to capitalize on the expanding market and demand for lubricants, which hold higher value than fuels and most plastics. Although pyrolysis could be used to convert plastic waste into lubricanting oils, commercial-scale operation will be difficult to attain, as solid waste is subsequently produced along with the main product.[22] Therefore, the hydrogenolysis of PU-PP has gained interest as a promising technology for producing base oil and other fuel oils.[23] The combination of reactive separation and catalytic hydrogenolysis was discovered as an approach for separating the liquid product from polymer deconstruction.[24] Reactive separation integrates a reaction and a separation process into one unit operation, including precipitation, distillation, membrane filtration, or extraction. The separation process removes a product from the reactant mixture, thus easing equilibrium limitations.[24,25]

Numerous studies have discussed the use of hydrogenolysis. For instance, Celik *et al.*[26] investigated hydrogenolysis using a well-dispersed platinum catalyst supported on strontium titanate (Pt/STO) for the depolymerization of polyethylene (PE) at 170 psi of hydrogen (H_2) and 300 °C for 96 hours, obtaining high yields of liquid hydrocarbons. Hernández *et al.*[23] investigated the pyrolysis of LDPE to olefins, followed by conversion to lubricanting oil. These authors demonstrated the production of Group III lubricanting oils from LDPE through hydrogenolysis over a ruthenium/tungstated zirconia catalyst at 250 °C and 30 bar for 2 hours. Although this method yields a small fraction (15–20 wt%) of lubricanting oil compared to pyrolysis technologies (48%), it generates the lowest carbon dioxide (CO_2) emissions among them.[23] Nakaji *et al.*[27] studied the hydrogenolysis of PP on CeO_2-supported Ru (Ru/CeO_2) at 240 °C to produce various liquid products and waxes. Rorrer *et al.*[28] investigated the hydrogenolysis of PP waste over Ru/C under mild conditions (200–250 °C and 20–50 bar H_2) to produce liquid alkanes. Dufaud *et al.*[29] conducted the catalytic hydrogenolysis of PP over a zirconium hydride supported on silica–alumina at moderate activity under mild conditions (190 °C), producing diesel or lower alkanes. Du *et al.*[30] studied the catalytic hydrogenolysis–isomerization of waste polyolefin plastics over a Rh/Nb_2O_5 catalyst at 300 °C and 3 MPa to high-value gasoline, diesel, and light lubricants. Kots *et al.*[31] studied the conversion of PP waste over a Ru/TiO_2 catalyst to lubricants *via* hydrogenolysis.

Some studies have compared the economic and environmental impacts of various advanced recycling technologies for converting PU-PP into value-added products.[1,32] Zabaniotou *et al.*[1] studied the economic feasibility of PP plastic waste employing the pyrolysis method and obtained oil yields varying from 82 wt% to 92.3 wt%, respectively, for operating temperatures from 450 °C to 550 °C. Khoury *et al.*[32] studied the techno-economic and environmental assessment of PP waste conversion using pyrolysis and found that the product retrieval from waste PP could reach 94%. This can play a substantial role in lessening raw material extraction and emissions. However, very few studies have discussed the economic feasibility and environmental impact of the hydrogenolysis of PU-PP into lubricating oils within a reactive separation process. To this end, this study presents the conversion of PU-PP into lubricating oils from upcycled plastics (LOUPs) from economic and environmental perspectives. Process modeling, techno-economic analysis (TEA), and life-cycle assessment (LCA) are used to understand the trade-offs between the economic and environmental performance of the proposed technology. Due to a lack of publicly available models of lubricant base oil production

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from crude oil, a comprehensive evaluation was performed as a baseline assessment. TEA and LCA results of the hydrogenolysis of PU-PP conversion to LOUPs were discussed and compared with the conventional technologies for producing base oil from crude oil. Sensitivity analyses were performed to compare process options to identify variations in the economic and environmental impacts that changing conditions can have on the system.

Materials and methods

Process design and techno-economic analysis for conventional lube oils and LOUPs

Process description and economic model construction for the conventional lube oil. Due to the limited available data on the environmental and economic analysis of conventional base oil refining from crude oil, simulation models were developed. These models were designed to estimate material and energy consumption for three distinct types of base oils (Groups I, II, and III)[33] and to facilitate subsequent TEA and LCA. Fig. 1 displays a simplified block diagram for the production of base oils from crude oil, based on current conventional pathways.[34] The refining pathways for conventional base oils were modeled in Aspen Plus v12, and the minimum selling price (MSP) for each base oil group was estimated using Aspen Economic Analyzer v12. Detailed process configurations are provided in Fig. S1–S3 (Groups I–III) in the SI. Basrah Light crude oil (API gravity 33.7) was selected as the feedstock; its assay data are reported in Tables S1–S3 of the SI.[34] Light crudes are economically attractive because they yield high-value refined products (*e.g.*, gasoline and diesel) and dominate refinery inputs across most U.S. regions.[33,34] The United States produced an average of 11.1 million barrels per day of light crude in 2022. Accordingly, the process models were parameterized using Basrah Light assays available in the Aspen Plus database.[35]

Most of the base oil manufacturing plants are a combination of many processing steps, including multiple distillation columns, extraction columns, flashers, *etc.*, and depending on the type of base oil (Group I, Group II, and Group III) produced, the different processes, such as solvent extraction, hydrotreatment, and hydrocracking, are used. The unit operations, processing stages, and conditions of the modeled processes were based on the Best Available Techniques (BAT) reference document from the refining of mineral oils and gas from the European Commission[36] and from other literature resources.[37–41] For additional details on the production of Group I, Group II, and Group III, please refer to Section 1 of the SI. It was also assumed that the facility has a plant life of 20 years, operating for 8000 hours each year to consider maintenance and downtime. According to U.S. Energy Information Administration (U.S. EIA) reports, the United States refining capacity reached 18.4 million barrels per calendar day at the beginning of 2024, a 2% increase compared to 2023.[42] To produce base oils, this study considered a flow rate of crude oil between 100 000 and 200 000 barrels per day at a refinery, which is consistent with the operating capacity of refineries in the U.S.,[43,44] with an operating temperature and pressure of ∼93.33 °C and 4.14 bar, respectively. Details of the process description and process configurations, economic model construction, raw materials, and financial assumptions of the TEA analysis for the conventional lube base oils are provided in Sections S1 and S2 of the SI (Tables S1–S17).

Process description and economic model construction for LOUPs. A modular catalytic reactor was modeled for the conversion of PU-PP to LOUPs using experimental data from Sadow *et al.*[45] on PP hydrogenolysis with a platinum strontium titanate (Pt/$SrTiO_3$ or Pt/STO) catalyst. PU-PP was fully converted into LOUPs and other lighter hydrocarbon products in the presence of the Pt/STO catalyst, yielding up to 86% based on experimental results at the laboratory scale.[45] Because catalysts can affect the economics and environmental impacts of LOUPs production, the TEA and LCA of the Pt/STO catalysts employed

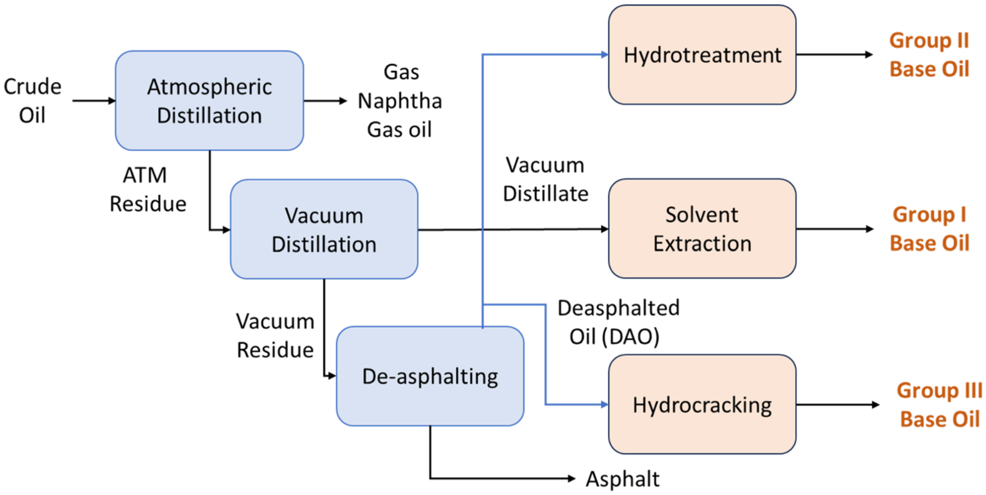

Fig. 1 Simplified block diagram of conventional production of base oils from crude oil, adapted from O'Reily.[34] This figure shows a simplified flow diagram to produce base oils through the solvent extraction route for Group I base oil, as well as hydrotreating and hydrocracking routes for Group II and Group III base oils, respectively. ATM refers to atmospheric distillation.

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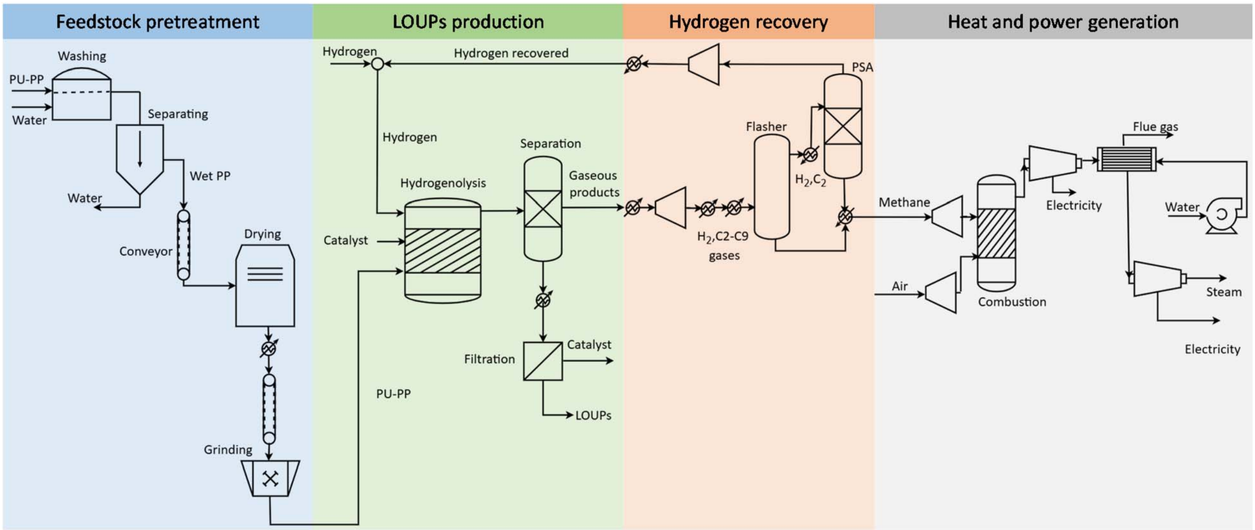

Fig. 2 Simplified PFD for the LOUP production from PU-PP. PSA: pressure swing adsorption.

have been analyzed in a previous publication.[46] This catalyst exhibits high dispersion, small particle sizes (<2 nm), and high stability of Pt supported on STO, outperforming other Pt-based catalysts in converting post-use polyolefins to narrowly distributed liquid alkane products for lubricants.[26,47] Catalyst characterization was conducted using techniques, such as scanning transmission electron microscopy (STEM), transmission electron microscopy (TEM), powder diffraction (PXRD), and inductively coupled plasma optical emission spectrometry (ICP-OES), to determine Pt content. The characterization analyses, along with the turnover frequency (TOF), have been published in prior studies.[26,48,49] This catalyst features distinct active sites free from uncontrolled deposition and molecular degradation, providing suitable conditions for spectroscopic and kinetic characterization. These characteristics support the optimization of both activity and selectivity.[48,50] Catalyst recyclability and performance testing (*i.e.*, activity and stability) confirmed its effectiveness, achieving both a high conversion rate (∼86%) and selectivity for base oil production.[26,47,48] Consequently, Pt/STO is highly favorable for the selective hydrogenolysis of polyolefins into hydrocarbon oils, offering a promising method for upcycling PUP.[26,48]

A preliminary design of an industrial process for the hydrogenolysis of PU-PP was designed to process 250 tonnes (t) per day (t/d), which is similar to other modeled advanced recycling facilities available in the literature[51] and is also a typical size of a material recovery facility (MRF) in the United States.[52] The processing volume of a facility of this extent signifies 0.21% of total plastic waste generated in 2019 in the United States.[51] The plant uses common equipment, and the model outcomes (mass and energy balances) were used as the basis for the inventory data used in the TEA and LCA. Fig. 2 presents the process flow diagram (PFD) for the modeled hydrogenolysis plant. As observed, products of PU-PP hydrogenolysis are recovered upon reaching the full conversion to LOUPs and gaseous products. The modeled plant uses PU-PP film shredded flakes collected from an MRF as feedstock, which are stored in the plant warehouse for future use. The plant is divided into four sections: (1) feedstock pretreatment, (2) LOUPs production, (3) H_2 recovery, and (4) heat and power generation. In the first section, PU-PP undergoes additional washing to eliminate residual impurities before being transported *via* conveyor belts to various processing operations within the facility. The PU-PP flakes are also pretreated through drying and size reduction processes and then subjected to catalytic hydrogenolysis to produce LOUPs in the second section. In the third section, gases (C1–C9) and H_2 are separated and recovered through downstream processes. The fourth section focuses on heat and power generation, where energy in the form of electricity and steam is efficiently recovered. Further details for each section of the plant are outlined below.

The pretreatment steps are designed to improve hydrogenolysis conversion. PU-PP is fed to a washing tank with hot water at a temperature of 80 °C to remove residual dirt from the MRF. After the separation of water and PU-PP, it is dried at a temperature of 66 °C using a hot drum to maintain a water content of less than 10%.[53] The dried flakes are cooled to 25 °C and reduced to a 2–3 mm particle size,[51] using a hammer mill grinder.

Following mechanical pretreatment, the granulated PU-PP flakes are then fed to continuous stirred-tank reactors (CSTRs) with a modeled Pt/STO catalyst loading of (1 : 100) in the base case, at 310 °C and 14.8 bar, with a 72 hour residence time, along with H_2 for producing LOUPs under catalytic hydrogenolysis. Pt/STO is produced directly in a catalyst plant, with an estimated production cost of $842.[46] The products of the hydrogenolysis after complete PU-PP conversion are liquids and gases. After the reactive separations are completed, the gas and liquid phases are collected and cooled to a temperature of 25 °C. The gas stream, which contains light alkanes (C1–C9) and H_2, has a weight fraction of ∼14 wt%, and the liquid stream contains predominantly LOUPs and Pt/STO. The liquid stream

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was passed through a filter to remove the catalyst Pt/STO from the solution, and LOUPs are subsequently recovered at a purity of more than 99 wt%. The overall yield of LOUPs in the process is modeled at approximately 86%. The collected catalyst is stored for further use. The gas stream is subsequently passed through a compressor and cooler to reach a temperature of −140 °C. Methane and H_2 gases are separated from the cooled gas stream using a flash separator. The top product from the flash, consisting primarily of H_2 and methane, is directed to a heater and then to a pressure swing adsorber (PSA) operating at 26 °C and 9.8 bar.[54] After passing through PSA, H_2 and methane are separated, with 85% of the H_2 recovered at a purity of 99%. The recovered H_2 is then compressed and heated to reach the reactor's operating conditions of 310 °C and 14.80 bar. The hydrogen is subsequently recycled back to the reactor for the next cycle. The purged methane from the PSA is combined with the bottom product from the flash separator, which is a liquid composed mostly of C2–C9 hydrocarbons. This combined stream passes through a heater and a compressor and is finally sent to a combustion reactor, modelled as a Gibbs reactor in Aspen Plus, for heat and power generation. In the combustion chamber, the gases are burned with excess air, and the resulting effluent gas is directed to a gas turbine to enable mechanical operation, thereby generating electricity.[55] The gas turbine generates electricity and produces high-temperature flue gas. This high-temperature flue gas was used to drive a steam turbine through a heat recovery steam generator, which is a type of heat exchanger that uses the high temperature of flue gas to heat the water to produce steam. The superheated steam from the heat recovery steam generator drives the steam turbine to generate electricity.[55]

The outcomes of the material and energy balances from the model were used to estimate the required equipment size and capital investment, whereas information on raw materials and utilities was used to estimate the variable operating expenditure (OpEx). The capital expenditure (CapEx) is the function of the equipment purchase cost, while fixed operating costs and variable operating costs were employed to estimate the OpEx. CapCost was also employed for the estimation of equipment costs.[56] The raw materials and the chemical costs were adjusted for the analysis year 2023 using the Producer Price Index for industrial chemicals[57] and, for equipment costs, the Chemical Engineering Plant Cost Index.[58] A discounted cash flow analysis for a certain financial parameter was applied to estimate MSP. It was assumed that the facility has a plant life of 20 years, operating for 8000 hours each year to consider maintenance and downtime. Process parameters and specific assumptions, financial assumptions, the baseline costs (*e.g.*, utilities and materials), and a summary of the results from the economic analysis of LOUPs production are shown in Tables S18 to S24 of Section S3 in the SI.

Because the process uses PU-PP collected from a MRF as feedstock, there are differences in the waste composition, sorting schemes, and purity percentage of PP bales across MRFs, introducing variability in the quality and hence in the price of PU-PP.[59,60] This material usually undergoes partial pretreatment, such as segregation of mixed materials, removal of foreign items, compaction, and baling, before being transported from the MRF.[61] The bales of PU-PP are sent to nearby facilities to shred them into a size of 20 to 25 mm. The market baled PP price is $0.119 per kg and shredding the bales costs $0.264 per kg, and their sum reflects the total PU-PP price.[62,63] A baseline feedstock PU-PP cost of $0.383 per kg was used in this process design.

Life cycle analysis for conventional lube oil production and LOUPS

This analysis focuses on the evaluation of the greenhouse gas (GHG) emissions, fossil energy use, and water consumption for the refining of conventional base oils and the production of LOUPs through hydrogenolysis. Additionally, an evaluation for the conventional end-of-life (EOL) management of PU-PP is presented in Section S5 of the SI. The LCA was performed using the Research and Development Greenhouse gases, Regulated emissions and Energy use in Technologies (R&D GREET) model version 2023.[64] Depending on the system under study, the functional unit of the analysis was defined as the refining of one kg of base oil or the production of one kg of LOUPs with a cradle-to-gate scope. The analysis considered the United States as the geographical location; therefore, the U.S. average electricity grid generation was considered, and the associated impacts of natural gas, crude oil, and chemical production were considered in accordance with those available in R&D GREET.

Life cycle assessment of conventional base oil production. The LCA considered a cradle-to-gate scope, which excluded the use phase and disposal of the base oils, and used a functional unit of one kg of the three base oil production pathways shown in Fig. S5 to S7 in the SI. These pathways had a similar processing for the pre-flash, atmospheric, and vacuum distillation, and de-asphalting stages, and differed in the processing applied to the heavy vacuum gasoil (solvent extraction in Group I base oil production) and base oil (dewaxing, hydrotreating, and hydrocracking for Group I, II, and III production, respectively). Because these production pathways are sequential, where the output of one stage connects to the input of the next, we used a stage-level analysis approach. In this approach, the environmental impacts of the input stream in a subsequent stage are estimated based on the environmental burdens associated with the material and energy used in the preceding stage that generated this input. This approach was followed stage by stage until the environmental impacts of the end product (base oil) were estimated. Due to the multiple output streams obtained at each stage, allocation methods were utilized to distribute the environmental impacts of the stage among the different co-products (see Tables S32 to S34 in the SI). These methods distribute the burdens of the process among its co-products using allocation factors that reflect the share of each output stream based on properties such as energy content, mass, or market value. For the pre-flash, atmospheric, and vacuum distillation stages, an energy allocation method was used because these stages generate energy products. In contrast, the subsequent stages, which involve the treatment of vacuum residue, heavy vacuum gasoil, and base oil, used a mass-

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Table 1 Material and energy inputs (after allocation) of the production of one kg of conventional base oils[a]

Input category	Input type	Group I	Group II	Group III
Material inputs	Crude oil (MJ)	40.91	44.19	43.09
	Furfural (kg)	1.41×10^{-3}	—	—
	Propane (kg)	0.04	3.01×10^{-5}	1.16×10^{-5}
	Hydrogen (MJ)	—	3.77	4.66
Energy inputs	Natural gas (MJ)	2.14	6.14	10.42
	Electricity (MJ)	0.12	4.43	7.47

[a] Material and energy inputs after allocation for each stage are available in Section S4 of SI.

allocation approach since these stages generate chemicals. In the energy allocation method, certain output streams, such as water, were excluded from the allocation factor calculations because they were not considered energy products. After developing the process model of each of the three pathways, data on material and energy inputs were collected for each stage. The allocation factor of a desired output stream was then multiplied by the material and energy requirements of its generating stage to obtain the specific life cycle inventory data for that output stream. The application of this stage-level approach enabled the acquisition of the life cycle inventory data for each base oil. The summary of the inventory data is presented in Table 1. Individual inventory data for each stage of each base oil production pathway are shown in Tables S35 to S37 of the SI. The inventory data were subsequently entered into R&D GREET to estimate the GHG emissions, fossil energy use, and water consumption of the base oils.

Life cycle assessment of the production of LOUPs. As observed in Fig. 2, the output stream of PU-PP from the MRF is sent to the conversion facility, where it undergoes pretreatment before the hydrogenolysis stage. The products from the hydrogenolysis reactor are subsequently separated through a series of processes, including flash separation, gas separation, and pressure swing adsorption. From the separation processes, LOUPs, recycled hydrogen, methane, and gas are obtained as co-products. The recycled hydrogen is sent back to the hydrogenolysis reactor, while gas and methane are used for heat and power generation, which are utilized internally. Because the power generated is higher than the electricity requirements of the conversion facility, there is surplus electricity that is sold to the grid. The amount of heat generated fulfills 89% of the heat requirements of the facility. Therefore, electricity and LOUPs are the only co-products generated by the conversion facility. During the heat and power generation process, a residual stream of flue gas is released into the atmosphere. This flue gas contains 8.9% wt. of CO_2, which contributes to the total GHG emissions of the facility. Since the flue gas is not utilized at any capacity, it does not qualify as a co-product.

A system-level approach was employed to conduct the LCA considering a functional unit of one kg of LOUPs. This approach was selected because of the lower number of co-products per stage in comparison to conventional base oil production. Additionally, the separation stages after the hydrogenolysis reaction contribute to the generation of heat and power that reduces the overall requirements for the entire conversion facility. The system-level approach distributes the burdens of the material and energy requirements of all the stages among the co-products, which in this facility are LOUPs and electricity.

Market-based allocation, energy-based allocation, and displacement were the three co-product treatment methods utilized to distribute the environmental impacts for the LCA. Market-value allocation was chosen due to the heterogeneity between LOUPs and electricity, which are classified as a chemical and as an energy product, respectively. The price of LOUPS was obtained from the TEA results, while the price of electricity was estimated from the average price of commercial electricity in the U.S. from 2019 to 2023 (0.115 USD per kWh).[65] The displacement method attributed all the burdens of the conversion process to LOUPs and subtracted a credit from the electricity generated that is sold to the grid. The life cycle inventory data derived from the process modeling are presented in Table 2. These data are shown before the application of co-product treatment methods to show the quantities of LOUPs produced and electricity generated. These data were entered into the R&D GREET model for the estimation of GHG emissions, fossil energy use, and water consumption.

To provide a comprehensive analysis, the system expansion approach was also applied, as it is commonly used in LCA of upcycling technologies for plastics.[66] In this approach, the emissions avoided by diverting PU-PP from conventional EOL management are subtracted from the baseline GHG emissions. As detailed in Section S5 of the SI, conventional EOL management involves a combination of landfill storage and incineration, with the latter occurring either with or without energy recovery (*i.e.*, electricity generation from heat produced during combustion). The subtraction of avoided EOL management

Table 2 Life cycle inventory data (before allocation) to produce one kg of LOUPs

Input category	Input type	Amount	
Material inputs	PU-PP	1.16	kg
	Hydrogen	0.01	kg
	Catalyst	1.34×10^{-6}	kg
	Hexane	2.00×10^{-3}	kg
	Water	3.12	L
Energy inputs	Natural gas	0.35	MJ
Products	LOUPs	1.00	kg
	Electricity	1.04	MJ
Residues	Flue gas	5.72	kg

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emissions from both scenarios has been considered in the analysis. An alternative LCA was conducted to evaluate hydrogenolysis as a technology for PU-PP valorization. This analysis utilized a functional unit of one kilogram of PU-PP processed. The results from this approach were compared to those of conventional PU-PP EOL management. Further details about this analysis are provided in Section S6.3 of the SI.

Sensitivity analysis for life cycle assessment

This study evaluated the variations in different factors, including plant capacity, the CO_2 content in flue gas, and the use of natural gas, H_2, and PU-PP. Each factor was individually varied ±50% relative to the baseline value. The 50% variation was chosen to maintain consistency with the sensitivity analysis performed in the TEA. This analysis aimed to identify which of the evaluated factors has the most significant influence on the GHG emissions associated with LOUP production.

Results and discussion

TEA results for conventional lube oil and LOUPs

The estimated MSPs obtained for the conventional base oil production from crude oil for Group I, Group II, and Group III base oils are $2.59 per gal, $4.27 per gal, and $5.66 per gal, respectively. Fig. 3 displays the contribution of the different cost components to the MSPs. The estimated MSPs were compared with the market prices available in the literature;[67] however, the operating conditions in those sources are either unknown or may differ from those used in this analysis, as shown in Fig. 3 and Table S25 of the SI.[67] In every instance, the raw material costs are the largest and the significant driver of the MSP. The findings revealed that the estimated MSPs of Groups I and II are lower than the market price, with reductions of 18.6% and 13.4%, respectively. Conversely, the MSP of Group III exceeds the market price by 5.8% (see Fig. 3). The molecular weight and the density of the modeled conventional base oils were also compared with reference sources,[17,68,69] showing consistency, as indicated in Tables S26 and S27.

The cost breakdown of hydrogenolysis technologies is shown in Fig. 4a. As previously discussed, the base case scenario involves a 250 t/d sized plant that yields LOUPs while generating electricity as a co-product. By including revenue from the electricity sold at its market price, LOUPs achieves an MSP of $2.18 per gal, with the majority of the cost attributed to raw materials (~71%), primarily the cost of PU-PP, as highlighted in light green in the simplified cost breakdown chart (Fig. 4a). Furthermore, increasing the catalyst-to-feedstock (C/F) ratio from 1/100 (base case) to 1/10, while keeping the base catalyst cost at $842 per kg, increases the estimated MSP of LOUPs to $2.58 per gal (~18% increase over the base case MSP).

Compared with the Group I, II, and III base oils, the MSP of LOUPs is ~16%, 49% and 61% lower, respectively (see Fig. 4a), which indicates that the production of LOUPs *via* hydrogenolysis is competitive with conventional base oil production in terms of MSP. The estimated MSP was also compared with the lubricant base oil market price (1.5–1.8 $ per kg).23 This comparison indicates that the MSP of LOUPs ($2.18 per gal or $0.72 per kg) is lower than the market price of base oils, highlighting the significant potential of hydrogenolysis of PU-PP as a feasible industrial process. Furthermore, the production cost of the catalyst Pt/STO used for PU-PP upcycling was $842 per kg.[46] In the future, with the development of more efficient and low-priced catalysts, the LOUP production using the hydrogenolysis route could become more feasible and economical. The base case result for the MSP of LOUPs has been further outlined by the different process sections. A comprehensive cost breakdown of the MSP for LOUP production from PU-PP using hydrogenolysis technology, including the total installed capital

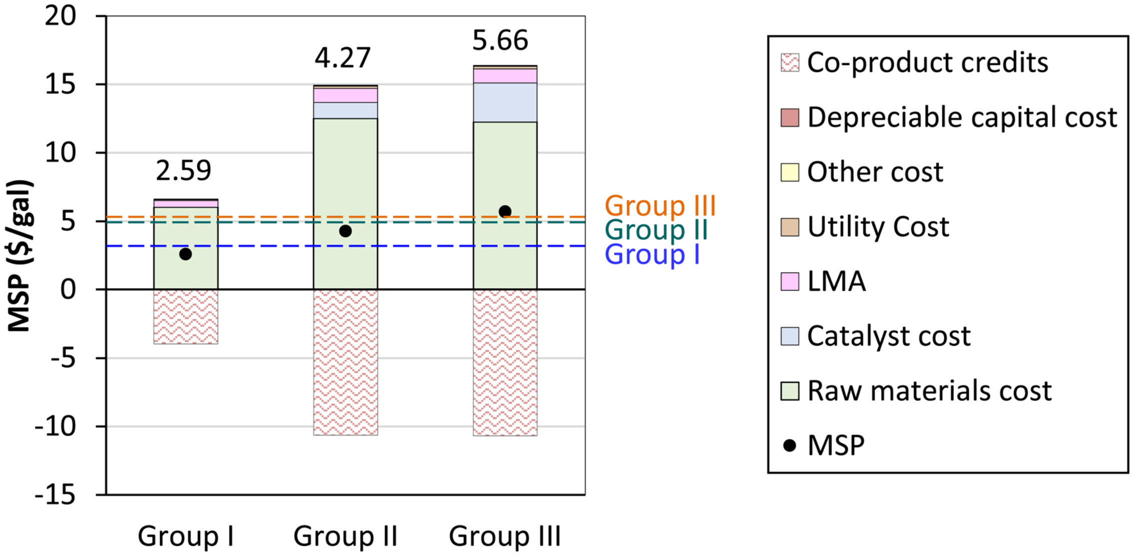

Fig. 3 MSP cost breakdown to produce Group I, II, and III base oils from crude oil as a function of process variables. The market price of Group I, II, and III base oils is 3.18, 4.93, and 5.33 $ per gal, respectively (see dashed lines). MSP: minimum selling price. LMA: labor, maintenance, and administrative costs.

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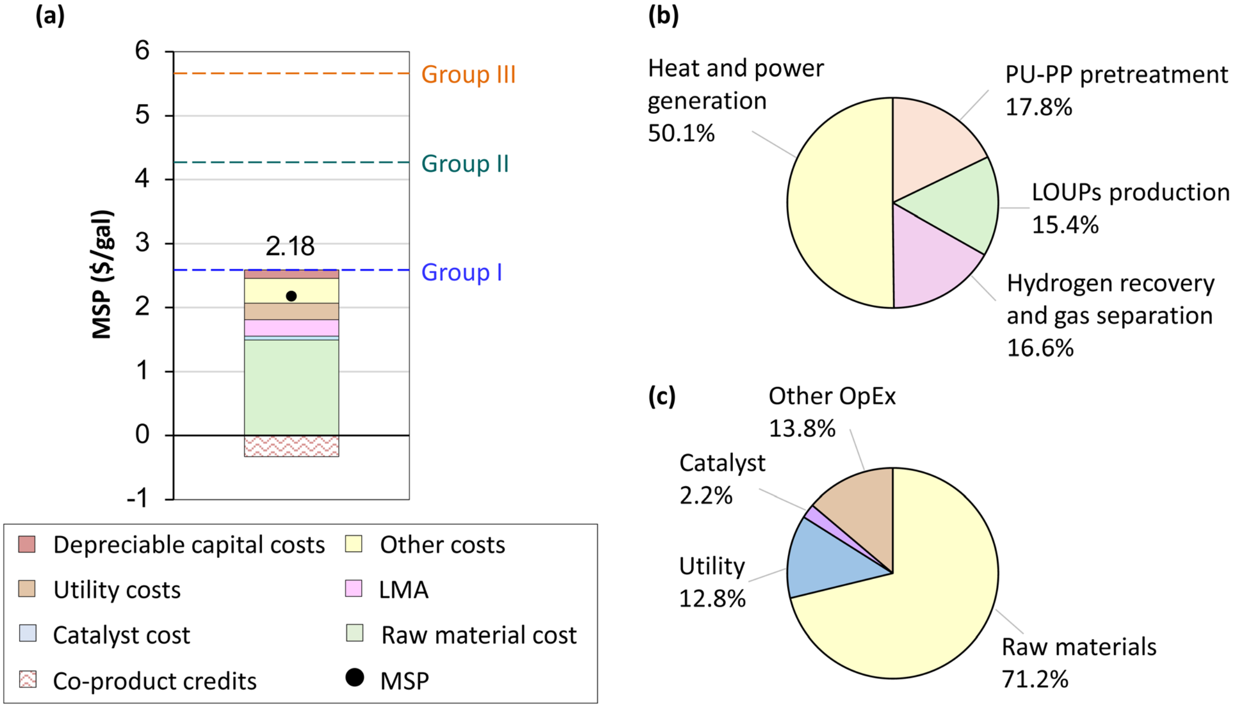

Fig. 4 TEA results for the production of LOUPs from PU-PP by the catalytic hydrogenolysis technology proposed in this study. (a) The cost breakdown of LOUPs as a function of process variables. The MSPs of Group I, II, and III base oils are 2.59, 4.27, and 5.66 $ per gal, respectively (see dashed lines). Comparison of the MSP of the base lube oil from Group (I–III) and the PP *via* the hydrogenolysis method. (b) The breakdown of capital costs for each process section, with a total installed capital (TIC) of $40.16 M and a total capital cost of $75.85 M. (c) Breakdown of the annual operating expense of $47.37 M for LOUP production.

cost (TIC) and annual operating contributions, is presented in Fig. 4b and c, respectively. Fig. 4b shows the installed capital costs breakdown in the different process sections (see Fig. 2). The total installed capital cost (TIC) for the plant is estimated at approximately $40.16 million (M), with the heat and power generation section being the largest contributor to installed costs. Specifically, for compressors and turbines, this section accounts for ∼50.14% of the total installed capital (TIC). The feedstock pretreatment is the second-highest contributor (17.83%), as this section uses energy-intensive equipment (*i.e.*, dryer and grinder). The hydrogen recovery and gas separations are the third key contributor (16.61%) of TIC, mainly due to the compressors, cooler, and PSA to separate H_2 due to expensive equipment costs. The contribution of the LOUP production section is 15.41% of TIC, with approximately 83% of the contribution from the reactor cost. Additionally, the plant entails $47.37 M per year in operating costs. Fig. 4c illustrates the breakdown of the annual operating costs (OpEx). Raw materials, including PU-PP and H_2, are the largest contributor to OpEx, accounting for ∼71% of the total. Other contributors (such as labor and maintenance) and utility costs each account for ∼13–14% of OpEx. Utility costs contribute 13%, primarily due to the cleaning, drying, and grinding of PU-PP after its transfer from the MRF, as well as the separation of gases and H_2 during LOUP production. Complete information on the TIC for each process section and annual OpEx requirements is provided in Tables S28 and S29 of the SI. The feedstock pretreatment section is the largest consumer of electricity, accounting for ∼90.4% of total electricity usage, primarily for washing, drying, and grinding. In other process areas, the cost of H_2 and the catalyst, particularly in reactors, and the use of refrigerant for the coolers are the primary drivers.

Due to the lack of publicly available data on LOUP production from PU-PP *via* hydrogenolysis, the estimated MSP is compared with the MSP of lubricating oil produced from polyethylene under catalytic hydrogenolysis.[23,70] However, it is important to note that the operating conditions, type of catalyst, C/F ratio, and conversion are not similar to those of the PU-PP hydrogenolysis analyzed here. Hernández *et al.*[23] estimated the MSP of lube oil from the conversion of LDPE using hydrogenolysis technology using a ruthenium catalyst supported on tungstated zirconia ($Ru/WO_3/ZrO_2$). The estimated MSP of this study is 1.67 per kg ($5.04 per gal). Note that the conversion to $ per gal is estimated by assuming that the density of the lube oil is the same as that of this study (3.02 kg per gal), as the density of the lube oil is not provided. Capello *et al.*[70] estimated the MSP of high-density liquid (a substitute for polyalphaolefin lubricants) from the conversion of HDPE using catalytic hydrogenolysis. The MSP was in the range of $0.6–$1.98 per kg of lubricant, depending on the operating conditions. The authors considered the density of the lube oil to be 3.0 kg per gal. Based on this density, the estimated MSP ranges from $1.80 per gal to $5.94 per gal, depending on the C/F ratio, which varies from 1/20 to 1/10. A comparison of the MSP of the base lube oil from

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PU-PP *via* the hydrogenolysis method in our study and that in the reference study is illustrated in the SI in Fig. S4.

Sensitivity analysis of the TEA results of LOUP production

A univariate sensitivity analysis was performed using the Aspen Plus model to assess the impact of varying process variables (*i.e.*, OpEx, PU-PP cost, CapEx, and plant size) on the MSP; the results are presented in Fig. 5a. Numerical data for this figure are reported in Table S30a of the SI. The minimum and maximum values used in this study are shown in the *y*-axis labels of the figure as white and black bars, respectively. The central line indicates the MSP of LOUPs in the base case.

Within the chosen parameter ranges, OpEx has a significant impact on the MSP (see Fig. 5a). For instance, a ±50% variation in OpEx is found to change the MSP of LOUPs by about ±45.46% from the base case. As the total feedstock cost is influenced by the expense of converting bales into flakes, which varies over time, this factor will have a substantial effect on the economic viability of the process. Consequently, PU-PP cost is the second contributor driving the variability in MSP. A 50% increase in the feedstock PU-PP cost from the baseline ($0.383 per kg to $0.57 per kg) results in a 33.1% rise in MSP. Conversely, a 50% reduction in PU-PP cost ($0.19 per kg) lowers the MSP by 33.1% compared to the base case. This implies that there is a linear relationship between PU-PP price and the MSP of LOUPs. Capital estimates may vary significantly from the actual total CapEx. To account for this, a contingency of ±50% is applied to CapEx. A ±50% variation in CapEx modifies the MSP of LOUPs by ±12.4%. Additional sensitivity cases explored in this case study include varying the plant size and plant life. With respect to plant size, reducing the facility capacity by 50% (from 250 t/d to 125 t/d) increases the MSP by 9.9% compared to the base case. Conversely, expanding the plant size by 50% (to 375 t/d) lowers the MSP by 13.7%. This highlights the importance of developing a robust supply chain for large-scale plastic-to-base oil facilities to capitalize on economies of scale and achieve a reduced MSP. Additionally, extending plant life from 20 to 40 years resulted in a 1% reduction in the MSP. The effect of varying other key parameters (*i.e.*, cost of H_2 and catalyst cost) in the hydrogenolysis section on the MSP was also evaluated. The cost of H_2 can fluctuate based on the production technology and source. For the base case, it is set at $2.25 per kg and adjusted during the sensitivity analysis (Fig. 5a). Considering a contingency of ±50% in the hydrogen cost, the MSP varies by ±1.8% from the base case. An analysis of the variations in the catalyst cost and catalyst life (see Fig. 5a) showed that these factors have a minimal effect on MSP, with variations of only 1–2% compared to the baseline. Even under the highest variability (+50%) in key process factors, including OpEx and PU-PP cost, the MSP of LOUPs remains lower compared to conventional Group II and III base oils. Furthermore, sensitivity analyses were conducted assuming a percentage change in the conversion of PU-PP, co-product credits (*i.e.*, electricity credits), the utility cost reduction for steam production, and some equipment costs, such as the catalytic reactor, PSA, filter, combustion reactor, and dryer (see Fig. 5b). The numerical data of this analysis are reported in Table S30b of the SI. The maximum and minimum values used in this study are shown in the *y*-axis labels as black and white bars, respectively. The results indicate that increasing the LOUP yield to 90% lowered the MSP by 4%, whereas reducing the yield to 60% resulted in an increase in the MSP by 44% from the base case. Modifications on the desired rate of return (DROR) were also considered, demonstrating that a DROR of 20% resulted in a 29% increase in MSP compared to the base case with a DROR of 10%. On the other hand, a DROR of 7% decreased the MSP by 6% from the base case.

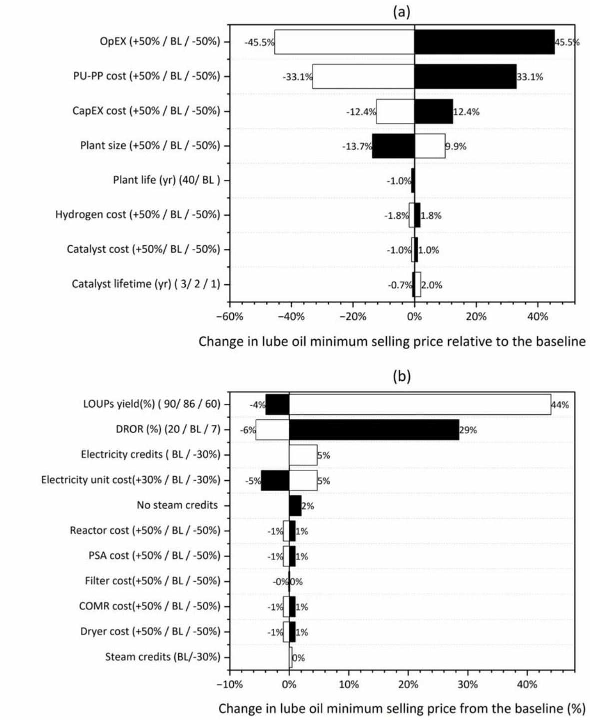

Fig. 5 Sensitivity analysis of the MSP of LOUPs. (a) The single-point sensitivity analyses showing the relative effect of process variables on the MSP of LOUPs ($2.18 per gal). (b) Sensitivity analysis assuming a % change in the PU-PP to LOUP conversion (*i.e.*, LOUP yield), co-product credits (*i.e.*, electricity credits), the utility cost reduction for steam production, some expensive equipment costs, and a financial variable. COMR: combustion reactor.

Since some of the major equipment costs were estimated using the software tool CapCost[56] and vendor quotes, a sensitivity analysis to illustrate the effects of variations in equipment cost on MSP was included. When a variation of ±50% is applied individually to the reactor cost, PSA cost, combustion reactor (COMR) cost, and dryer cost, the MSP only changes by ±1% from the base case. The cost contributions to OpEx are counterbalanced by the recovery of energy (*i.e.*, electricity and steam production) from the heat and power generation section. The base case estimates the MSP, including both co-product credit (*e.g.*, electricity) and credits for steam production. Therefore, to assess the effect of these credits on the MSP, sensitivity cases were also conducted for the three scenarios: (1) only changes in co-product credits were considered, (2) only changes in utility credits were contemplated, and (3) without credits for steam

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production (Fig. 5b). Reducing the co-product credits (*e.g.*, electricity) by approximately 30% decreases the MSP by 5% from the base case. However, a 30% decrease in steam production has no impact on MSP (decrease ~ 0.5%). By excluding steam credits, the MSP increases by 2% from the base case. In the end, the market unit cost of the co-product (*i.e.*, electricity), varying over a ±30% range, has a relatively minor impact on the overall MSP of LOUPs (±5% change in MSP). Details on the conversion and co-product credits can be found in Table S30b of the SI.

A multivariate sensitivity analysis was conducted in this study to understand the interaction of process variables, as a single-point sensitivity analysis may not be sufficient to reveal all process modeling insights.[51] When increasing the C/F from 1/100 (base case) to 1/10 with the same base catalyst cost ($842 per kg), the estimated MSP of LOUPs is $2.58 per gal (~18% increase from the base case of $2.18 per gal). However, the analysis was extended to evaluate the simultaneous change in both variables (C/F and catalyst cost) to identify non-linear relations that may affect the overall MSP (see Fig. 6). By raising the C/F and the catalyst cost to 1/10 and $1473 per kg (75% increase from the base catalyst cost), respectively, the estimated MSP increased by ~33.5% ($2.91 per gal) compared to the base case. Conversely, decreasing the catalyst cost to $210 per kg at C/F = 1/10 results in an MSP of $2.25 per gal (~3% increase in MSP from the base case). Numerical data of this analysis are provided in Table S31 of the SI. The potential challenge is that the Pt/STO catalyst is not yet commercially available. However, initial evaluations indicate that this type of catalyst meets some proof-of-concept criteria and shows potential for commercial viability as an upcycling catalyst.[48] It is noted that in the TEA study, the effects of energy required (*i.e.*, electricity) for PU-PP bales to shred into flakes at the plastic recycling facility, as well as the impact on the price associated with transportation for gathering and moving the PU-PP feedstock between the MRF and plastic recycling facility, and from the MRF to the plant, have not been considered. Also, the economic analysis does not include the maintenance cost of the filter used to separate LOUPs from the catalyst. However, the analysis was explored further by adding an extra price for hexane utilized to wash the catalyst after each cycle, which was not accounted for in the base case.[70] The amount of hexane required for the washing was estimated based on Capello *et al.*[70] With the addition of the cost of hexane, the MSP increases to $2.19 per gal (a change of only 0.5% from the base case).

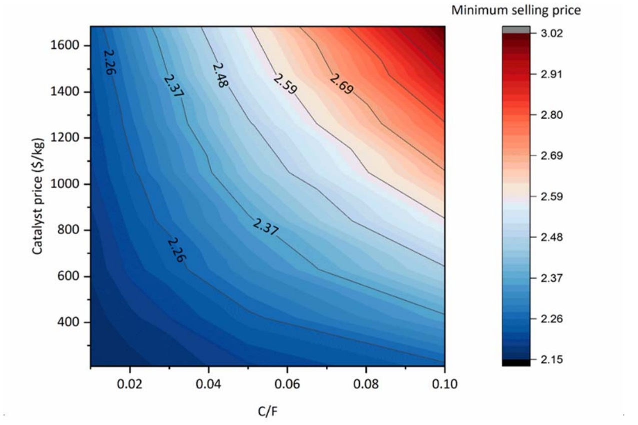

Fig. 6 A multivariate sensitivity analysis for the effect of the variation of catalyst cost and catalyst to feedstock ratio (C/F) on the MSP of LOUPs. The base case values for the C/F ratio and catalyst cost were 1/100 and $842 per kg, respectively.

LCA results for conventional lube oils and LOUPs

GHG emissions, fossil energy use, and water consumption of conventional base oil production. The results of the LCA for the production of conventional base oils are presented in Fig. 7. The highest GHG emissions (2.3 kg CO_2e per kg) are observed for Group III base oils, primarily driven by the use of H_2 and electricity. This is attributed to the higher energy requirements for the hydrocracking process (see Tables S6 to S8 in the SI), which consumes nearly twice as much energy as the hydrotreatment process in Group II and approximately three times more than the solvent extraction and dewaxing processes in Group I. These differences arise from the adjustments in process conditions necessary to meet the specifications for each type of base oil. The GHG emissions associated with Group II and Group I base oils were estimated at 1.6 kg CO_2e per kg and 0.5 kg CO_2e per kg, respectively. Notably, Group I base oils exhibit the lowest energy requirements for processing, as they do not require hydrogen and involve minimal use of chemicals. This is further supported by the high recovery rates of solvents (such as furfural and propane) observed during the process. Despite differences in processing, the use of similar quantities of crude oil as feedstock resulted in comparable GHG emissions (~0.3 kg CO_2e per kg) associated with feedstock utilization across all three base oil groups. When comparing the results with other studies,[23,70,71] it was observed that previous research reported GHG emissions for conventional Group III base oils ranging from 1.0 to 1.2 kg CO_2e per kg, which are lower compared to the value presented in Fig. 7. However, in some studies,[61] the production of Group III base oil shows an inclusion of process stages, such as aromatic extraction and dewaxing, which, based on the process modeling conducted in this study, are exclusive to Group I base oil production. Additionally, some values, such as those reported by Hernandez *et al.*,[23] are presented as general base oil production rather than being specific to a particular type. The results presented in the current study, although derived from process simulations, only include the impacts of those stages involved in the production of each distinct type of base oil, as reported in the literature.[36]

The fossil energy use of conventional base oils ranged from 48 to 75 MJ per kg, with the lowest value observed for Group I base oil and the highest for Group III base oil. As expected, the higher energy consumption in Group III, compared to Groups I and II, is primarily driven by increased electricity use, with a smaller contribution from natural gas. Interestingly, when considering fossil energy use, the crude oil used as feedstock emerges as the largest contributor, accounting for up to 90% of the total fossil energy in the case of Group I base oil. This is because the crude oil fossil energy indicator includes both the

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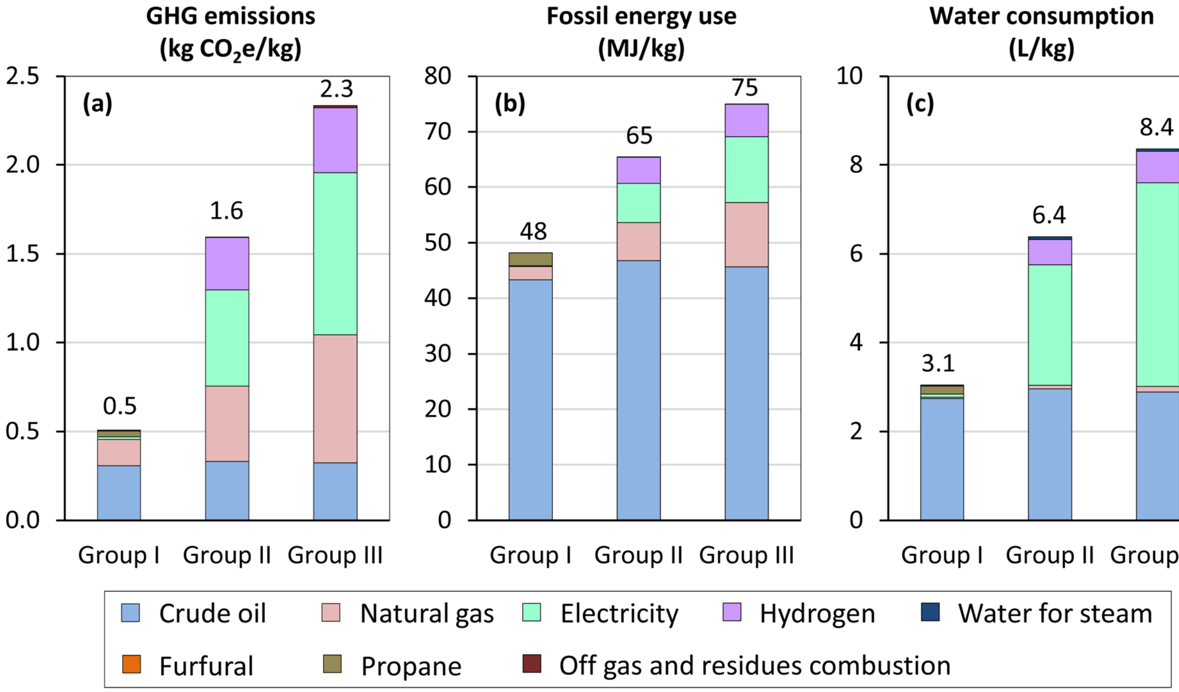

Fig. 7 Environmental impacts of conventional base oils: (a) GHG emissions, (b) fossil energy use, and (c) water consumption.

energy associated with extraction and the internal energy of the crude oil itself. In fact, approximately 94% of the fossil energy of crude oil is attributed to its internal energy. The water consumption of conventional base oils ranged from 3.1 L per kg for Group I to 8.4 L per kg for Group III. As shown in Fig. 7C, the higher electricity consumption associated with Group II and Group III base oils is the primary factor driving the increased water consumption observed in these two groups. Additionally, crude oil serves as a significant feedstock contributor to water consumption, accounting for 90% of the total water consumption in the case of Group I base oil. The percentage differences among the three base oils varied across the three impacts evaluated. For instance, GHG emissions exhibited the largest variation, with Group III base oils producing 78% higher emissions compared to Group I. The percentage differences between Group I and Group III for fossil energy use and water consumption were estimated at 36% and 64%, respectively. In the case of fossil energy use, a smaller difference was observed compared to the other impacts. This is primarily due to the high energy contribution from crude oil, which is similar across all three base oils. This similarity is attributed to the similar yield of base oil from crude oil, while the high fossil energy use results from the inclusion of crude oil's internal energy in the estimations.

GHG emissions from the production of LOUPs. The baseline GHG emissions to produce one kilogram of LOUPs are presented in Fig. 8. Depending on the co-product treatment method employed, these emissions ranged from 0.6 kg CO_2e per kg LOUPs (displacement method) to 0.71 kg CO_2e per kg LOUPs (energy-based allocation). The higher emissions observed in the market- and energy-based allocation methods, compared to the displacement method, are attributed to the high allocation factors (>0.95) for LOUPs, driven by their yield (∼0.9 kg per kg PU-PP) and price (2.1 USD per gal). Approximately 70% of the process-related GHG emissions are associated with the residual flue gas generated as a byproduct of the heat and power generation process, while the contributions from PU-PP and hydrogen requirements are 12% and 14%, respectively. Given the significant influence of flue gas emissions, a potential strategy to reduce GHG emissions could involve the incorporation of carbon capture and utilization technologies, such as chemical adsorption or pressure swing adsorption. However, implementing these strategies would likely increase production costs and, consequently, the price of LOUPs.

These results fall within the range reported by Capello *et al.*,[70] who estimated GHG emissions for base oil derived from polyethylene at 0.5–1.2 kg CO_2e per kg, depending on production process yields. Consistent with the present study, their analysis identified combustion emissions from heat and power generation as the dominant source of GHG emissions. However, the present study shows lower hydrogen consumption due to a higher recycling rate compared with theirs. In contrast, the technology reported by Hernandez *et al.*[23] showed slightly higher GHG emissions, at 1.8 kg CO_2e per kg. Their process utilized a $Ru/WO_3/ZrO_2$ catalyst and achieved a lower lubricating oil yield of 15–20%, compared to the yields reported by Capello *et al.*[70] (60–90%) and this study (86%), which likely contributed to the higher GHG emissions in their process.

When the system expansion approach is included, the counterfactual scenario with energy recovery resulted in GHG emissions ranging between 0.14 and 0.26 kg CO_2e per kg, representing a reduction of 0.4–0.5 kg CO_2e per kg LOUPs compared to the baseline analysis. In the scenario without

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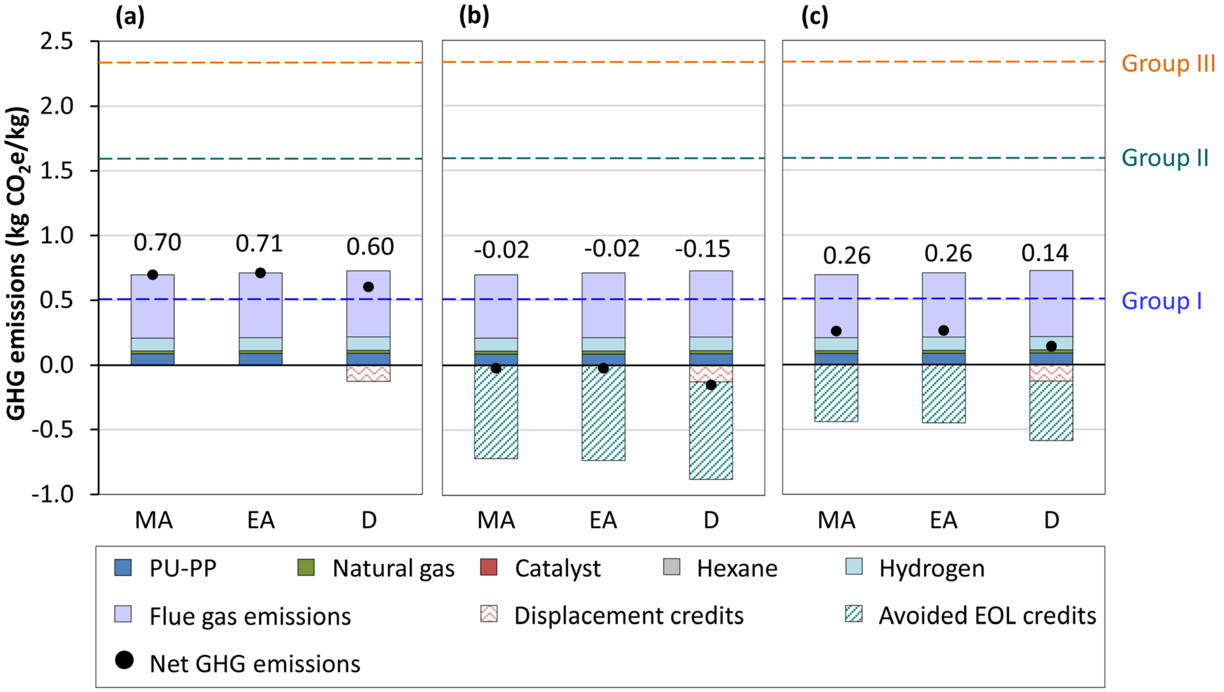

Fig. 8 GHG emissions of the production of LOUPs: (a) baseline analysis, (b) system expansion with a counterfactual scenario of conventional EOL management without energy recovery, and (c) system expansion with a counterfactual scenario of conventional EOL management with energy recovery. Dashed lines indicate the GHG emissions of conventional base oil production from crude oil (see Fig. 8a). The black dots represent net GHG emissions. MA: market-based allocation, EA: energy-based allocation, D: displacement, and EOL: end-of-life.

energy recovery, GHG emissions were estimated between −0.02 and −0.15 kg CO_2e per kg LOUPs, indicating a similar reduction of 0.7–0.8 kg CO_2e per kg LOUPs with respect to the baseline. Greater reductions are observed in the scenario without energy recovery, as more emissions from incineration are avoided. Fig. 8 also shows the GHG emissions for the three types of conventional base oils analyzed in this study. Comparisons of the baseline analysis indicate that LOUPs exhibit 18% to 40% higher GHG emissions than Group I base oil. However, for all co-product treatment methods, the GHG emissions of LOUPs are lower than those of Group II and Group III base oils. These reductions are as high as 62% (displacement method) for Group II and 74% (displacement method) for Group III. When system expansion is considered, the production of LOUPs demonstrates lower GHG emissions compared to all three conventional base oils. For instance, LOUPs show up to 2.5 kg CO_2e per kg lower GHG emissions than Group III base oils when emissions from conventional EOL management without energy recovery are included in the analysis. These findings highlight the importance of the methodology employed, as different approaches provide distinct perspectives—whether focusing solely on process-related GHG emissions or considering broader system-level impacts. Regardless of the methodology, the results indicate that LOUPs can achieve lower GHG emissions compared to Group II and Group III conventional base oils.

Fossil energy use and water consumption in the production of LOUPs. Fig. S11a of the SI shows the fossil energy use for LOUPs, which ranges from 7 to 8 MJ per kg LOUPs. The majority of the fossil energy use is attributed to the PU-PP feedstock, accounting for 73% of the estimated value. A comparison of the fossil energy use of LOUPs and conventional base oils indicates that LOUPs have significantly lower fossil energy use, achieving reductions of up to 85% compared to Group I base oil. The comparative reduction in fossil energy use is even greater for Group II and Group III base oils. These substantial reductions are primarily due to the process-related fossil energy use per kilogram of LOUPs being less than half of that observed for conventional base oils. Additionally, the processing and internal energy associated with PU-PP is lower than that of crude oil used as the feedstock for conventional base oils. Water consumption results are shown in Fig. S11b and range between 2.9 L per kg LOUPs (displacement method) and 3.4 L per kg LOUPs (mass-based and energy-based allocation methods). Approximately 89% of this water consumption is attributed to process water used for washing the PU-PP entering the conversion facility, steam production in the heat and power generation plant, and cooling water losses (see Section S6.2). When compared to conventional base oils, the water consumption of LOUPs is up to 55% and 66% lower than that of Group II and Group III base oils, respectively. However, the comparison with Group I base oil indicates that, only under the displacement method, the water consumption of LOUPs is lower than that of Group I. The process-related water consumption of LOUPs is approximately 15% higher than that of Group I base oil. However, further analysis suggests that this water consumption of LOUPs could be reduced by 36% if

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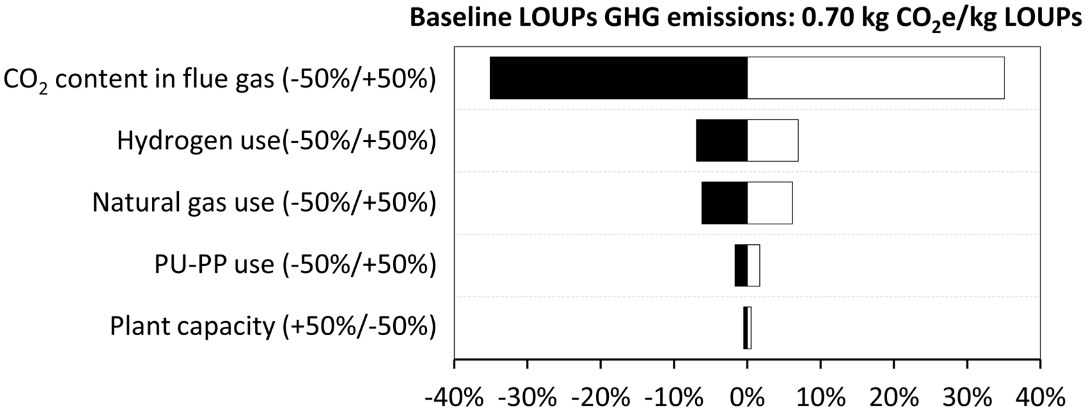

Fig. 9 Variations in GHG emissions of the production of LOUPs under variations for different process factors.

strategies such as treating and recycling the water used for washing are implemented.

The comparison of hydrogenolysis as a PU-PP valorization technology with conventional end-of-life (EOL) management of PU-PP revealed reductions in GHG emissions ranging from 0.3 to 2.2 kg CO_2e per kg PU-PP. Additionally, hydrogenolysis demonstrated significant reductions in fossil energy use compared to conventional EOL management. While water consumption for hydrogenolysis was generally lower than conventional EOL management without energy recovery, it was higher when Group I base oil was displaced. Furthermore, hydrogenolysis offers additional benefits, including reduced solid waste generation and improved recyclability compared to conventional EOL management of PU-PP. A detailed discussion of this analysis is provided in Section S6.3 of the SI.

Sensitivity analysis of the GHG emissions of LOUP production

As shown in Fig. 9, among the four factors evaluated, the CO_2 concentration in flue gas had the greatest influence on the GHG emissions of LOUPs, causing variations of 35% compared to the baseline GHG emissions, with a 50% change. This was followed by more moderate variations in GHG emissions from changes in H_2 use, natural gas use, and PU-PP use, which compared to the baseline resulted in variations of 7%, 6%, and 2%, respectively. Finally, changes in plant capacity did not have a significant effect on GHG emissions, as the material and energy requirements varied proportionally with the amount of PU-PP processed. These results highlight that the implementation of carbon capture and sequestration processes would have a more profound impact on reducing the GHG emissions of LOUPs compared to, for instance, using alternative sources of H_2 in the process.

Conclusions

An integrated economic and environmental assessment offers a more comprehensive understanding of the hydrogenolysis process, enabling a more informed evaluation and comparison of technologies. The modeled base case for hydrogenolysis projects the MSP of LOUPs at \$2.18 per gal, which is ∼1.2, 2.0, and 2.6 times lower than that of conventional Group I, II, and III base oils, respectively. In addition to evaluating the MSP of LOUPs, variations of parameters such as OpEx, PU-PP cost, co-product credits, CapEx, plant size, LOUP yield, desired rate of return, and equipment cost are also investigated, using sensitivity analysis. OpEx was found to be the most sensitive factor in the process, followed by plant size and capital costs. Raw materials, mainly the cost of PU-PP, are the largest contributor to OpEx. Increasing the plant size reduced the MSP, which demands the development of a strong supply chain for a large-scale waste-to-base oil facility. The heat and power generation section was the highest influential factor on installed capital costs, followed by the feedstock treatment section. The percentage of PU-PP to LOUP conversion plays a significant role in the MSP. The study indicates that increasing both the catalyst to feedstock ratio and catalyst cost leads to a higher estimated MSP compared to the base case. This study shows that significant opportunities exist to reduce the MSPs of LOUPs through the hydrogenolysis pathway. Furthermore, the conversion of PU-PP to LOUPs using the proposed hydrogenolysis process could benefit from the use of low-cost PU-PP, with reduced power consumption in the pretreatment section, and by conversion to high-value-added co-products to further improve the economics of the integrated plastic recycling facility. The LCA findings revealed that LOUPs production achieves lower GHG emissions, fossil energy use, and water consumption compared to Group II and III base oils. However, when compared to Group I base oils, LOUPs showed higher GHG emissions and water consumption. The analysis identified flue gas flaring emissions as the primary contributor to GHG emissions. Implementing strategies such as carbon capture to reduce CO_2 content in flue gas was shown to lower GHG emissions, although with the trade-off of increasing the MSP. The comparison of hydrogenolysis with conventional EOL management of PU-PP demonstrates significant reductions in GHG emissions and fossil energy consumption, primarily driven by the avoided emissions from displacing conventional base oil production. Additionally, hydrogenolysis reduces solid waste generation and enhances the recyclability of PU-PP when integrated into the supply chain. The sensitivity analysis indicated that moderate variations in GHG emissions can be achieved by

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adjusting hydrogen and natural gas usage within the process. The results provided in this study highlight the most important process variables to improve hydrogenolysis recycling technologies for PU-PP, serving as a foundation for future assessments.

Conflicts of interest

There are no conflicts to declare.

Abbreviations

ANL	Argonne National Laboratory
BAT	Best available techniques
C/F	Catalyst-to-feedstock
CapEx	Total capital costs
CO_2	Carbon dioxide
COMR	Combustion reactor
t/d	Tonnes per day
D	Displacement
DROR	Desired rate of return
EA	Energy-based allocation
EOL	End-of-life
GHG	Greenhouse gas
HDPE	High-density polyethylene
kg	Kilogram
L	Liter
LCA	Life cycle analysis
LDPE	Low-density polyethylene
LOUPs	Lubricating oils from upcycled plastics
MA	Mass-based allocation
MJ	Megajoule
MRF	Material recovery facility
MSP	Minimum selling price
OpEx	The total operational costs
PP	Polypropylene
Pt/$SrTiO_3$ or PT/STO	Platinum on strontium titanate
PUP	Post-use plastic
PU-POs	Post-use polyolefins
PU-PP	Post used polypropylene
R&D GREET	Research and Development Greenhouse gases, Regulated emissions, and Energy use in Technologies
SI	Supplementary information
TEA	Techno-economic analysis

Data availability

The life-cycle analysis conducted in this study was performed using the Research and Development Greenhouse gases, Regulated Emissions, and Energy use in Technologies Model (R&D GREET)® available at https://doi.org/10.11578/GREET-Excel-2023/dc.20230907.1.

Additional data supporting this article are provided as part of the supplementary information (SI). Supplementary information: (1) process specifications and material balances and energy use for the production of lubricant base oils from crude oil and LOUPs, (2) equipment cost, material costs, and assumptions utilized for the TEA of lubricant base oils from crude oil and LOUPs, (3) detailed methodology and data for the LCA of lubricant base oils from crude oil, (4) description of counterfactual scenarios for PU-PP EOL management, (5) LCA of conventional EOL management of PU-PP, and (6) discussion on the fossil energy use and water consumption of LOUPs. See DOI: https://doi.org/10.1039/d5su00910c.

Acknowledgements

We would like to thank Kathryn Peretti, Gibson Asuquo, and Eric Peterson from the U.S. Department of Energy (DOE) for their support. This work has been funded by the DOE Advanced Manufacturing Office and Bioenergy Technologies Office under the FOA project DE-EE0009300 Modular Catalytic Reactors for Single-Use Polyolefin Conversion to Lubricating Oils from Upcycled Plastics (LOUPs). We also want to thank Aaron Sadow, Massimiliano Delferro, Ali Erdemir, Wenyu Huang, Yi-Yu Wang, Ranjan Behera, Chao Meng, and Ken Hope for their valuable feedback, guidance, and support. We also thank Guiyan Zang from Argonne National Laboratory for reviewing the process design.

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