

# Economic and Environmental Sustainability of Waste Plastics Chemical Recycling from the Consequential Perspective

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Consequential life cycle assessment (CLCA) enables evaluating the environmental consequences of market dynamics overlooked by attributional life cycle assessment (ALCA). A consequential life cycle optimization (CLCO) framework is developed in this work to determine the economically optimal and most environmentally sustainable chemical recycling pathway for high-density polyethylene waste. The framework includes CLCA and techno-economic analysis (TEA), and the CLCO problem is formulated as a multi-objective mixed-integer nonlinear fractional programming problem (MINFP) that is effectively solved by an optimization algorithm. This multi-objective optimization problem aims to minimize the unit life cycle environmental impacts with maximum unit net present value (NPV). Environmental assessment results show that the total greenhouse gas (GHG) emissions evaluated by the CLCA approach are 14.22 % lower than those assessed by the ALCA approach. By evaluating the ReCiPe end-point score, CLCA further reduces the total environmental impacts corresponding to particulate matter formation by 17.37 %.

## 1. Introduction

Global plastic production surged to 359 M t in 2018 (Evangelidou et al., 2020), and the annual waste production is estimated to increase to 3,400 Mt within 30 years (Bergmann et al., 2019). Mismanaged waste plastics released into the environment will triple from 2015 to 2060 (Lebreton and Andrady, 2019), and these waste plastic emissions are now damaging the bio-ecosystem and causing human health concerns (Sigler, 2014). Chemical recycling processes are employed to reduce those disruptive consequences and has advantages over incineration in reducing greenhouse gas (GHG) emissions and fossil fuel use (Meys et al., 2020). Life cycle assessment (LCA) approaches, including attributional life cycle assessment (ALCA) and consequential life cycle assessment (CLCA) (Falcone et al., 2017), are used for quantifying these environmental impacts. ALCA approach mainly focuses on target processes and overlooks environmental consequences corresponding to market dynamics (Dalgaard et al., 2008) corresponding to various downstream products manufactured from chemical recycling processes (Bora et al., 2020). CLCA approach, on the other hand, incorporates economic models, such as partial equilibrium (PE) (Patouillard et al., 2020) or computable general equilibrium (CGE) models (Garcia and You, 2018) to quantify market dynamics (Kretschmer and Peterson, 2020). The system boundary is therefore expanded by integrating many consumers' and marginal suppliers' processes and this system expansion enables evaluating the environmental consequences associated with market dynamics (Earles and Halog, 2011). Nevertheless, the CLCA on waste plastic chemical recycling remains a knowledge gap despite the applicability of CLCA in evaluating the environmental consequences of market dynamics.

High labour intensity poses difficulties in evaluating environmental consequences of highly coupled decisions, such as selecting technology alternatives from the waste plastic chemical recycling process, by the CLCA approach (Gong and You, 2017). The consequential life cycle optimization (CLCO) framework is therefore introduced to address these methodological difficulties by determining the optimal technology pathway and evaluating the system expansion's impact on the total environmental performance. However, there is no existing study that employs this CLCO framework associated with waste plastic chemical recycling. This work then develops a consequential life cycle optimization (CLCO) framework to determine the economically optimal and the most environmentally sustainable waste high-density polyethylene (HDPE) chemical recycling pathway. The framework includes the methodologies of CLCA and techno-economic analysis (TEA), and the CLCO problem

is formulated as a multi-objective nonlinear fractional programming (MINFP) problem that is effectively solved by an optimization algorithm. This multi-objective optimization problem aims to minimize the unit life cycle environmental impacts with maximum unit net present value. The differences between CLCA and ALCA results on assessing waste HDPE processing systems are demonstrated by evaluating system expansion's impact on the total environmental performances based on the GWP (Global Warming Potential) and ReCiPe end-point score (Goedkoop et al., 2009).

**2. LCA Methodologies and CLCO Model**

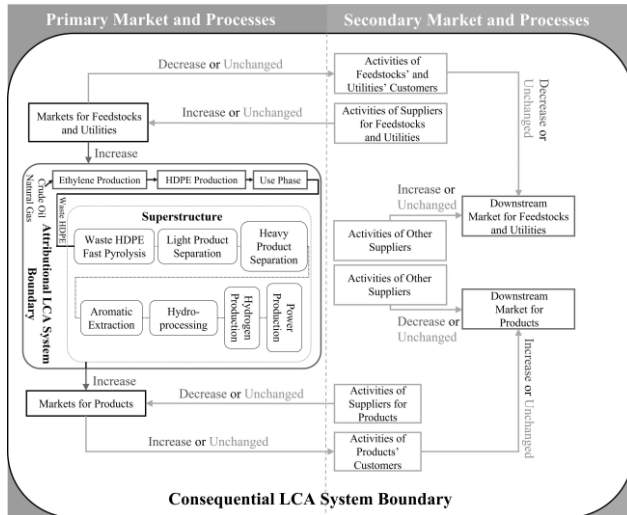


Figure 1: System boundary of the ALCA and CLCA on waste HDPE chemical recycling

This work develops a CLCO framework that accounts for the CLCA approach for determining the economically and environmentally optimal waste HDPE chemical recycling pathway. System expansion is typically used for treating co-product allocation within the CLCA system boundary given in Figure 1. The impacts of system expansion are deciphered by evaluating and quantifying the ALCA and CLCA results with the help of the following information.

**2.1 Superstructure Description**

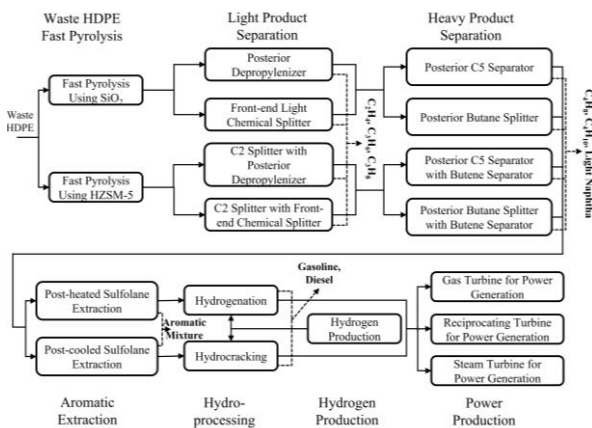


Figure 2: Superstructure of waste HDPE chemical recycling

The determination of optimal processing pathways requires a systematic evaluation and comparison of the environmental performance of each processing technology pathway with the help of the CLCA approach. This can be secured by developing a comprehensive superstructure of the waste HDPE chemical recycling incorporating multiple possible technology pathways shown in Figure 2. Seven processing sections are incorporated within this superstructure to produce various value-added basic chemicals from fast pyrolyzing

waste HDPE and downstream product separation. Heating utilities, such as high-pressure steam, are utilized within the heat integration for maintaining high-temperature operating conditions while cooling water is consumed onsite as cooling utilities. Specifically, the waste HDPE particles are fast pyrolyzed by using SiO<sub>2</sub> or HZSM-5 catalysts and producing various gaseous hydrocarbons (ethylene, propylene, propane, butene, butane) that are separated via fractionation (Yang et al., 2018) within technology alternatives in Light Product Separation and Heavy Product Separation processing sections. Aromatic mixture products are manufactured from two technology alternatives of the extractive-distillation process, and the remaining flow is treated by hydrogen to produce gasoline and diesel. The steam-methane reforming process is employed for manufacturing hydrogen onsite, while the remaining organic flow from the hydroprocessing processing section is sent to the power production section to generate electricity.

## 2.2 Goal and Scope Definition

The goal of this LCA work is to evaluate and compare the environmental impacts of waste HDPE chemical recycling through ALCA and CLCA approaches. Both LCAs are performed based on a “cradle-to-gate” system boundary due to the absence of the “end-of-life” phases for various hydrocarbons produced from the superstructure. For a specific processing technology pathway, the operational parameters like machinery efficiency are assumed not to vary with the treatment capacity, which results in a linear relationship with the material and energy input or output. Therefore, the functional unit is chosen as one ton of waste HDPE treated in this chemical recycling process. Notably, climate change and the air pollution caused are environmental hotspots assessed in studies on waste plastic treatment. The global warming potential indicator over the course of 100 y (GWP<sub>100</sub>) are employed to quantify the GHG emissions (Yue et al., 2014), while the ReCiPe hierarchical end-point score enables evaluating the environmental problems of air pollution and fossil fuel use.

## 2.3 Life Cycle Inventories

For ALCA, life cycle inventories (LCIs) are built based on the mass and energy relationships among all life cycle stages within the system boundary. Specifically, Ecoinvent V3.7 Database compiled with Aspen Plus-based process simulations is employed to extract the LCI data corresponding to mass and energy flows (Tian et al., 2020), while the electricity mix data is referred from the USA e-GRID Database. Compared with the ALCA approach, CLCA enables quantifying the environmental consequences of market dynamics with the help of economic models. The PE model is adopted in this work due to its applicability in specifying the environmental consequences corresponding to each downstream product (Weintraub, 1957) from waste HDPE chemical recycling. Market information, including the marginal suppliers of feedstocks and downstream products, as well as their corresponding consumers, are needed to be identified, while their environmental consequences are quantified with the help of the aggregate supply and demand functions that are built based on the collected price and elasticities data.

## 2.4 Interpretation

The system boundary of CLCA is expanded through integrating the various marginal suppliers' and consumers' processes collected from the market information. The impacts of this system expansion require to be evaluated through quantifying and evaluating the environmental impacts of waste HDPE chemical recycling by ALCA and CLCA approaches. CLCA results are the summation of the GWP<sub>100</sub>-based or ReCiPe-based environmental impacts from the production of raw material, HDPE, and utilities, as well as various environmental consequences corresponding to suppliers' and consumers' processes.

## 2.5 CLCO Model and Solution Methodology

The CLCO framework incorporating the superstructure, as well as CLCA and TEA methodologies, is developed and formulated to determine the environmentally and economically optimal processing pathway of waste HDPE chemical recycling. This model is subjected to constraints corresponding to mass and energy balance, market equilibrium, CLCA and TEA methodologies, as well as superstructure network configuration. This optimization model is then reformulated as a multi-objective MINFP problem to minimize the unit life cycle environmental impacts per functional unit (Yue et al., 2013) while maximizing the unit NPV (Net Present Value) calculated by the total NPV within the project lifespan divided by the total waste plastic treatment amount in tons. The model formulation and acronyms for model formulation (Table 1) is shown below, and this nonlinear optimization problem is effectively solved by a tailored optimization algorithm within finite iterations (Zhao and You, 2021).

$$\max \text{OBJ}_{\text{Eco}} = \frac{\text{NPV}}{\text{sp} \cdot t \cdot \text{CAP}} \quad (1)$$

$$\min \begin{cases} GW = \frac{(EIP)_{GWP}}{CAP} \\ RECIPI = \frac{(EIP)_{RECIPIE}}{CAP} \end{cases} \quad (2)$$

s.t. Mass and energy balance constraints  
Market equilibrium constraints  
CLCA constraints  
TEA constraints  
Superstructure network configuration constraints

Table 1: Acronyms used in the model formulation

Acronyms	Meaning	Acronyms	Meaning
$OBJ_{Eco}$	Unit NPV	$CAP$	Hourly waste plastic treatment capacity
$NPV$	Net present Value	$RECIPI$	Unit ReCiPe end-point score
$GW$	Unit GWP	$sp$	Project lifespan
$(EIP)_{GWP}$	Hourly GWP	$t$	Conversion factor from one year to an hour
$(EIP)_{RECIPIE}$	Hourly ReCiPe end-point score		

### 3. Results and Discussion

The CLCO optimization problem is solved by the optimization algorithm coded by GAMS 24.8.3. Solved by the CPLEX 12.7 as the optimizer, the optimal technology processing pathway shown in Figure 3 pyrolyzes the waste HDPE by using HZSM-5 catalyst, and the hydrocarbon products are separated from “C2 Splitter with Front-end Chemical Splitter”, “Posterior Butane Splitter with Butene Separator”, and “Post-heated Sulfolane Extraction”. The remaining heavy hydrocarbon stream is hydrogenated by the hydrogen manufactured from the steam methane reforming process, while the electricity is generated from the steam turbine. CLCA results are compared with the ALCA results on GWP and ReCiPe bases to evaluate the impact of system expansion on the total environmental impacts.

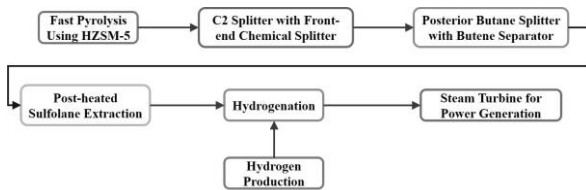


Figure 3: Optimal technology processing pathway of waste HDPE chemical recycling

System expansion is applied in the CLCA approach to treat co-product allocation by integrating various marginal suppliers' and consumers' processes within the CLCA system boundary. Figure 4 shows the impact of system expansion on reducing 14.22 % of total GHG emissions. Specifically, the total environmental consequence of suppliers' processes can alleviate 0.86 t CO<sub>2</sub>-eq/t HDPE treated, while those of consumers' processes can enhance the GHG emissions by 0.26 t CO<sub>2</sub>-eq/t HDPE treated.

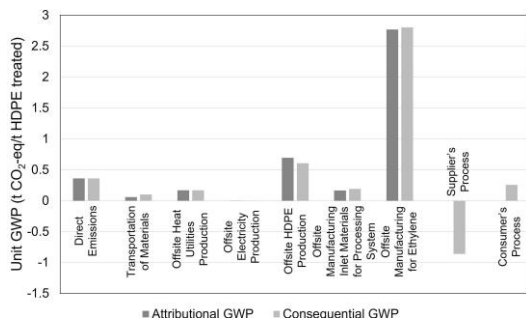


Figure 4: GHG emissions breakdowns of waste HDPE chemical recycling

Detailed environmental consequences for each supplier's or consumer's process are given in Figure 5, reflecting that the onsite production of 1-butene and propylene can reduce their manufacturing from offsite naphtha cracking (marginal suppliers' processes) and cut down their GHG emissions by 0.40 and 0.33 t CO<sub>2</sub>-eq/t HDPE treated. Since the natural gas is consumed to provide high-temperature heating energy used in the waste HDPE chemical recycling, this process reduces the natural gas for energy generation offsite. Thus, the production of alternative energy generation sources, such as heating oil, is enhanced to compensate for the natural gas consumption, leading to an increment of 0.16 t CO<sub>2</sub>-eq/t HDPE treated.

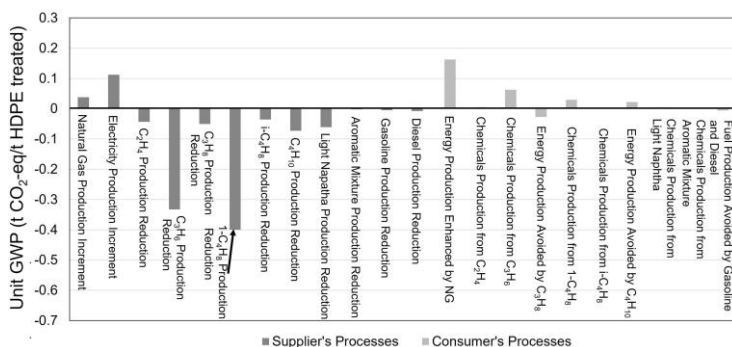


Figure 5: GHG emissions breakdowns for suppliers' and consumers' processes corresponding to waste HDPE chemical recycling

Figure 6 shows the impact of system expansion on the environmental performances of waste HDPE chemical recycling in terms of 17 impact categories. Specifically, the total environmental consequence of suppliers' processes and consumers' processes can reduce fossil fuel use by 20.14 % through the heat integration and substitution of fossil fuel by the hydrocarbons produced from the waste HDPE chemical recycling process. System expansion of CLCA can also mitigate the environmental problems corresponding to the particulate matter formation, which is another environmental hotspot, by 17.37 %. Other environmental impacts, including natural land transformation, terrestrial acidification, and terrestrial ecotoxicity, can also be reduced in the CLCA results. System expansion increases the ionizing radiation and urban land occupation by 43.32 % and 4.71 %.

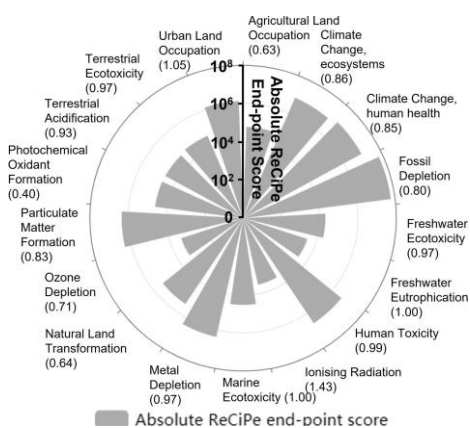


Figure 6: ReCiPe-based CLCA results and comparative results of CLCA and ALCA on waste HDPE chemical recycling, and the values in the brackets denote the ratio of CLCA and ALCA results corresponding to each impact category

#### 4. Conclusion

This work developed a CLCO framework to determine the most economically viable and environmentally sustainable waste HDPE chemical recycling processing pathway. The CLCO model was subjected to the constraints of CLCA and TEA approaches was formulated as a MINFP problem that was effectively solved by a tailored optimization algorithm within finite iterations. The environmental objective was to minimize the unit life cycle environmental impacts, while the economic objective was to maximize the unit NPV. The optimal technology processing pathway pyrolyzed the waste HDPE by using HSZM-5 catalyst, and the hydrocarbon

products are separated from “C2 Splitter with Front-end Chemical Splitter”, “Posterior Butane Splitter with Butene Separator”, and “Post-heated Sulfolane Extraction”. The remaining heavy hydrocarbon stream was hydrogenated by the hydrogen manufactured from the steam methane reforming process, while the electricity was generated from the steam turbine. System expansion used in CLCA reduced the total GHG emissions by 14.22 % compared to the ALCA results, while the particulate matter formation was decreased by 17.37 %. Specifically, the onsite production of 1-butene and propylene replaced their manufacturing from the offsite naphtha cracking (marginal suppliers’ processes) and reduced their GHG emissions by 0.40 and 0.33 t CO<sub>2</sub>-eq/t HDPE treated, respectively. The onsite consumption of natural gas led to the increment of total GHG emissions by 0.16 t CO<sub>2</sub>-eq/t HDPE treated.

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