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Sustainable Design and Synthesis of Waste High-Density Polyethylene Recycling Process

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Global optimization in sustainable waste high-density polyethylene (HDPE) chemical recycling process is addressed under economic and environmental criteria. In this work, by far the most comprehensive superstructure of the HDPE recycling process with 867 processing routes is developed to produce valuable products from waste HDPE. Using the methodologies of life cycle assessment and techno-economic assessment, the superstructure optimization problem is then formulated as a multi-objective mixed-integer nonlinear fractional programming (MINFP) problem to address the sustainable waste HDPE recycling process with maximum unit net present value (NPV) and minimum levelized ReCiPe end-point score. A tailored parametric algorithm is utilized to efficiently convexify the fractional objective functions. With the help of the piecewise linearization method, nonlinear economic constraints can be linearized and effectively solved. This research proposes to use the 'ε-constraints' method to obtain the Pareto-optimal curve. Results show that the optimal unit NPV ranges from -65 USD/t HDPE to 170 USD/t HDPE, and the levelized ReCiPe point of the most environmentally friendly design is 0.6 times of that of the most economically competitive design.

1. Introduction

The growing amount of waste plastics generated in the U.S, which has reached more than 34×10^6 t in 2017, is a serious concern for plastic recycling. Only 8 % (wt.%) of the waste plastics have been effectively recycled (US EPA, 2017), while 76 % (wt.%) of the waste plastics have been treated as landfilled trash (Casazza et al., 2019). The landfilled plastics pose harmful impacts on biodiversity by entangling the marine species in the sea, or being watered into microplastics to be absorbed by plant roots (Jain, 2019). Hence, it is vital to apply the effective recycling process for waste plastics. Mechanical recycling and chemical recycling are the two main categories. However, the contaminants formed by plastic additives in mixed plastics (Horodytska et al., 2018) degrades the quality of recovered plastic in the mechanical process. Comparatively, the chemical recycling process can decompose plastic into monomeric molecules or petroleum products (Iribarren et al., 2012) to be used in downstream polymer plants or petroleum refineries. Fast pyrolysis has a high yield of monomers (Gartzen et al., 2017), which is favored by factories that produce recovered plastics. Moreover, compared to the landfill process, the pyrolysis process can enhance the material utilization and pose less environmental impacts on the surroundings (Pinto et al., 2015). Various studies have been performed to evaluate the environmental impacts and economic performance of (Antelava et al., 2019) a fixed pyrolysis method and technology for separating products from waste plastics. Although various studies have focused on environmental sustainability in the waste plastic recycling process, the sustainable process design of recycling waste plastics has never been systematically addressed. To systematically compare and identify the optimal processing route for plastics recycling with maximum economic competitiveness and environmental sustainability, a superstructure optimization problem (Yeomans and Grossmann, 1999) is formulated based on the life cycle assessment (LCA) and techno-economic assessment (TEA).

In this work, by far the most comprehensive superstructure of the waste plastic recycling process is constructed by integrating the plastic pyrolysis process and a series of technology options for separating and processing products. The superstructure further integrates the electricity generation section to reduce the cost of purchasing electricity, and the wastewater treatment section to purify the wastewater, as well as the carbon-dioxide

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sequestration section to decrease the environmental impacts of direct emissions. Therefore, the proposed superstructure involves 867 diverse processing routes with nine sections. Based on this novel superstructure, a “cradle to gate” LCA and TEA are performed to provide environmental and economic parameters to the superstructure optimization problem. One ton of waste HDPE treated in the recycling process is used as a functional unit in the LCA. The superstructure optimization problem is then formulated as a mixed-integer nonlinear fractional programming (MINFP) problem to simultaneously maximize net present value (NPV) per ton of HDPE treated (unit NPV) and minimize ReCiPe points per ton of HDPE treated (levelized ReCiPe points) (Gong et al. 2016). However, the combinatorial nature and the pseudo-convexity of fractional objective functions make this MINFP problem computationally challenging (Gao and You, 2015). To efficiently solve this model, the parametric algorithm is used to convexify reformulate fractional objectives and use piecewise linearization to convexify nonlinear constraints in capital cost calculation. The optimal design of HDPE recycling with maximum unit NPV and minimum levelized ReCiPe points is obtained based on the resulting Pareto-optimal curve.

2. Superstructure Description

The proposed superstructure of the HDPE chemical recycling process is given in Figure 1.

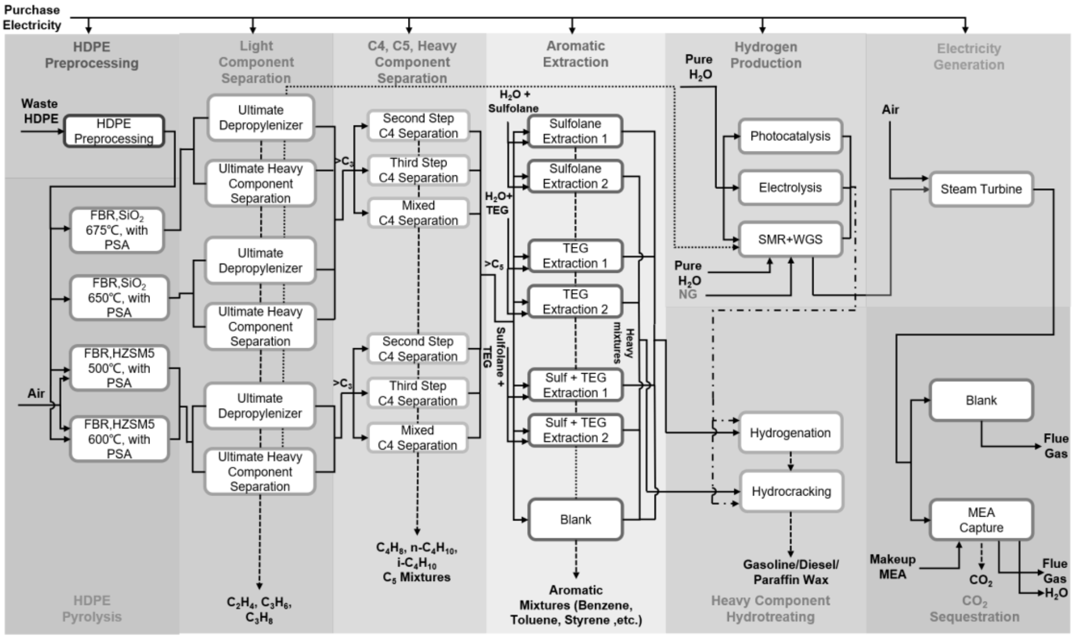

Figure 1: Detailed superstructure of waste HDPE recycling process

The waste HDPE needs to be collected and granulated into smaller particles in the HDPE preprocessing section. The HDPE particles are then transported into the fluidized pyrolysis reactor in the HDPE pyrolysis section, where the particles are decomposed into diverse hydrocarbons using different technology options with pressure swing-adsorption (PSA), namely “FBR, SiO_2, 675 °C, with PSA”, “FBR, SiO_2, 650 °C, with PSA”’, “FBR, HZSM-5, 500 °C, with PSA”, and “FBR, HZSM-5, 600 °C, with PSA”. The products from pyrolysis are firstly cooled down to separate heavy components and followed by the PSA unit to separate nitrogen, which is the fluidized gas. The treated stream of light component is then fractionated in the light component separation section, which resembles the procedure of the shale gas fractionation (Gong and You, 2018). The cryogenic conditions for separating methane and ethylene from the light component mixture is maintained by refrigeration cycles. The products, which involve ethylene, propylene, and propane are separated in high purity in this section. In the “C4, C5, heavy component separation” section, the stream of heavier components from the raffinate of the light component separation is split into n-butane, i-butane, butene as products. The stream of heavier components from the raffinate is directly fed into the heavy component hydrotreating section or aromatic extraction section. In the aromatic extraction section, the extraction process follows the method of the UOP extractive-distillation process (Asselin, 1977). The valuable aromatic mixture and petroleum products can be obtained from the aromatic extraction section and heavy component hydrotreating section (Swanson et al., 2010), respectively. Utilized in the hydrotreating process, the hydrogen is produced by the electrolysis, photocatalysis (Pinaud et al., 2013), or steam methane reforming process that uses methane produced from the overhead gas in the light component separation section. To reduce the electricity procurement cost, the steam turbine is used in the electricity generation section. All overhead gas streams from the upstream sections are mixed and combusted in the furnace. The energy from the flue gas stream outputted from the furnace is used to evaporate the precooled water, and the vapor pushes the steam turbine to do the work, which is then converted into electricity in the turbine generator of the steam turbine. The flue gas is fed into the CO_2 sequestration section, where the environmental impacts of the direct emissions from CO_2 are reduced. The reverse osmosis is used for purifying the wastewater produced from the electrolysis technology option in the hydrogen production section.

3. Life Cycle Optimization Approach

In this work, the LCA methodology is applied to provide environmental parameters to the superstructure optimization problem (Yue et al., 2016). As given in Figure 2, the system boundary is chosen from cradle to gate due to the absence of end-of-life phases of final products, such as n-butane. The system boundary confines four life cycle stages, involving the waste HDPE processing, wastewater treatment, electricity production, and utilities production. The functional unit in LCA is defined as processing 1 t of waste HDPE. The process simulation model in Aspen Plus and Ecoinvent V3.6 database are data sources for LCIs. The ReCiPe end-point score is addressed in LCA to fill in the knowledge gap of systematically addressing the sustainable design of the HDPE recycling process. Using the hierarchial ReCiPe end-point score, the life cycle environmental impacts per functional unit are transformed into levelized ReCiPe points.

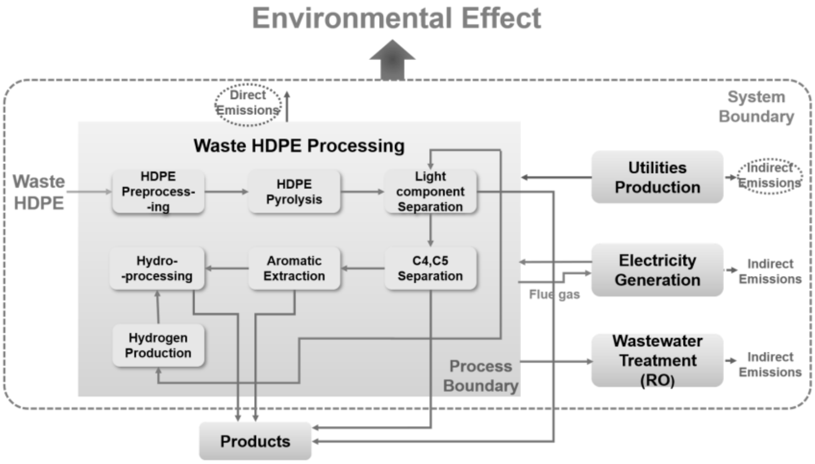

Figure 2: LCA system boundary depiction

TEA is used for passing economic parameters to the superstructure optimization problem. The TEA consists of the calculation of the capital expenditure (CAPEX) and operating expenditure (OPEX) for the waste HDPE recycling process. The CAPEX consists of the direct and working capital of all equipment units, as well as the land cost. The OPEX includes the feedstock cost, utility cost, operation and maintenance cost (O&M), property tax and insurance (PT&I), sale expense, and income tax (Gong and You, 2018). The data of capital costs for all units are taken from Aspen Economic Analyzer V10, as well as from the relevant literature.

4. Model Formulation

The superstructure optimization problem is formulated as a multi-objective model with economic and environmental objective functions. This model is subject to five types of constraints, namely logical constraints, mass balance constraints, energy balance constraints, techno-economic assessment constraints, and environmental impacts assessment constraints, which are shown as follows

$$\max \quad OBJ_{Eco} = \frac{NPV}{HDPE \times yr} \tag{1}$$

$$\min \quad OBJ_{Env} = \frac{RciPe}{HDPE} \tag{2}$$

s.t. Logical constraints

Mass balance constraints
Energy balance constraints
Techno-economic assessment constraints
Environmental impacts assessment constraints

In the mathematic model, the economic objective is to maximize the unit NPV, which is calculated by dividing the NPV by the mass of waste HDPE treated within the project lifespan. The environmental objective is to minimize levelized ReCiPe points, which are calculated by dividing the life cycle ReCiPe points by the mass of waste HDPE treated annually. Both objective functions are formulated as fractional forms to address functional-unit-based life cycle performances (Yue et al., 2013). Moreover, nonlinear terms are introduced in capital cost calculation, and all other constraints have linear relationships with continuous and binary variables (Gao and You, 2018). Thus, the superstructure optimization problem is formulated as a MINFP problem (Gao and You, 2017), which is solved using a tailored parametric algorithm, ε-constraint method, and piecewise linearization. The model is coded and solved in GAMS 24.8.3 with CPLEX solver.

5. Results and discussion

The formulated superstructure optimization problem is solved by the aforementioned algorithm, and the results are directly given on a Pareto-optimal curve in Figure 3. Three technology integrations of the waste HDPE recycling process are linked with their corresponding optimal points. As given in Figure 3, the optimal process design of optimal solutions A and B have levelized ReCiPe points of 86 points/t HDPE and 97 points/t HDPE, respectively. These results are understandable due to the utilization of CO_2 sequestration in the waste HDPE recycling process, which can reduce the life cycle environmental impacts via removing CO_2. Moreover, the trade-offs between the economic and environmental performances are revealed when comparing the unit NPV and levelized ReCiPe points at point A (the most environmentally friendly solution) with those at point C (the most economically competitive solution). The CO_2 sequestration section is not chosen in the process design of solution C to enhance the unit NPV to 170 USD/t HDPE. However, the process design of solution B keeps high unit NPV and low unit ReCiPe score simultaneously. With the electricity purchased from the market, this process design decreases the life cycle environmental impacts of the direct emissions from the flue gas in the electricity generation section. Moreover, the capacity of CO_2 sequestration is reduced, which decreases the capital cost and maintains high unit NPV.

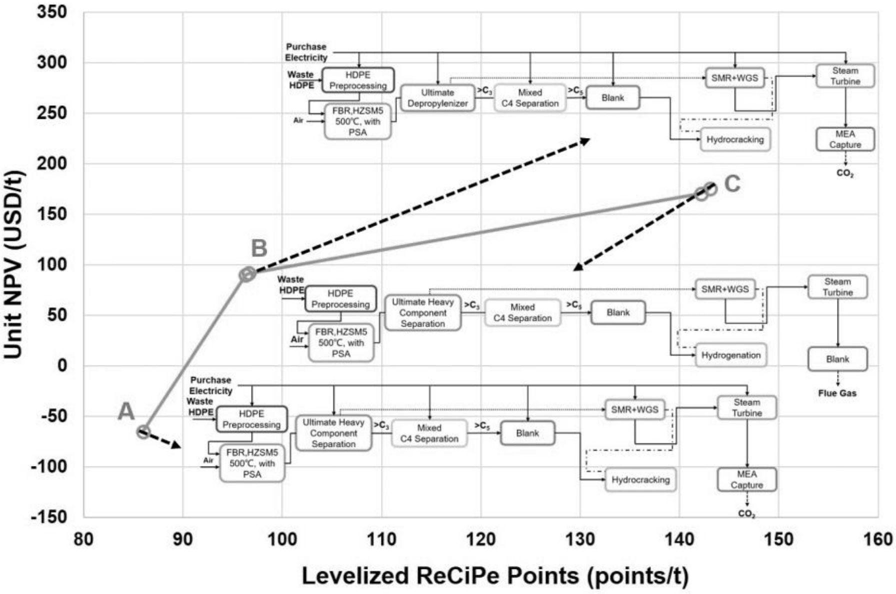

Figure 3: Pareto-optimal curve and technology integrations corresponding to optimal solutions

To systematically evaluate the economic performance of the optimal design of waste HDPE recycling process, the annualized CAPEX and OPEX breakdowns corresponding to optimal solutions A, B, and C are displayed in Figure 4. The lifespan of the waste HDPE recycling plant is 20 y, and the interest rate is chosen as 0.1. The breakdowns are based on seven categories, namely income tax, general expense, O&M, annualized total capital investment, utility cost, feedstock cost, and PT&I. All of the process designs corresponding to the optimal points have low ratios of utility cost to the total expenses (the summation of CAPEX and OPEX). The ratios are lower than 0.06, showing the optimality in energy savings. For the ratio of annualized total capital investment to the total expenses, the process design of the good-choice solution B remains the smallest, which is 0.09, illustrating the economic competitiveness revealed on the Pareto-optimal curve.

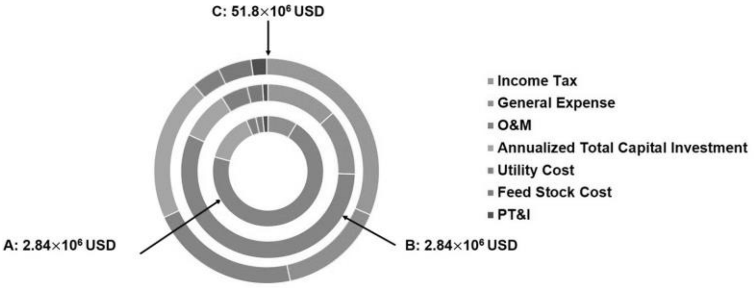

Figure 4: Annualized CAPEX and OPEX breakdowns of process designs of the optimal solutions

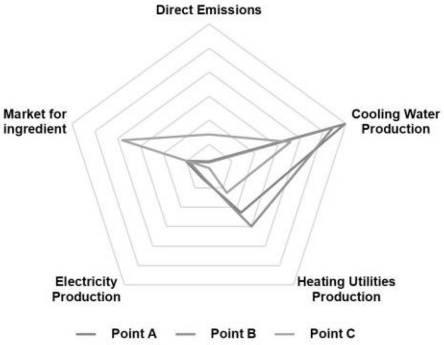

Figure 5: ReCiPe points breakdowns of process designs of the optimal points

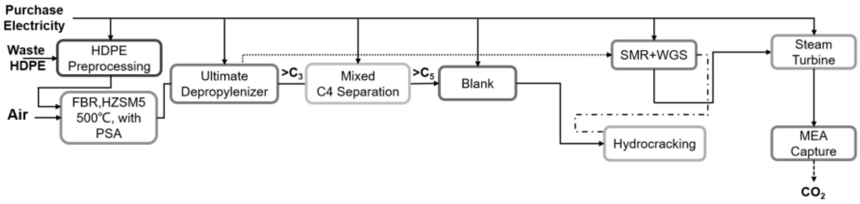

Figure 6: Process design of waste HDPE recycling process corresponding to good-choice solution B

The ReCiPe points breakdowns given in Figure 5 shows the life cycle environmental impacts caused by each life cycle stage within the system boundary. The values in this radar graph are in percentage. The results of the optimal solution C from the radar graph and the Pareto-optimal curve show that the capital cost is reduced via avoiding using the MEA capture to remove CO_2, which results in the highest percentage of the direct emission in the radar graph. Moreover, the electricity is self-produced to reduce the utility cost. This process leads to the high direct emissions of the greenhouse gas from the steam turbine. As shown in Figure 6, the optimal process design of the good-choice solution B integrates HDPE preprocessing, HDPE pyrolysis using FBR and HZSM-5 catalyst in 500 °C, ultimate depropylenizer, mixed C4 separation, steam methane reforming, hydrocracking, steam turbine, and MEA capture. The monomeric products, namely ethylene, propane, propylene, butane, and butene are produced in ultimate depropylenizer and mixed C4 separation. The remaining heavier components are directly hydrocracked to obtain gasoline, diesel, and paraffin wax as petroleum products.

6. Conclusion

In this study, by far the most comprehensive superstructure of the waste HDPE recycling process was proposed, which had 867 processing routes. Based on this superstructure, an LCO approach was applied to optimize the unit NPV and life cycle environmental impacts. A “cradle to gate” LCA and TEA were used to provide environmental and economic parameters to the superstructure optimization problem. A multi-objective MINFP problem was then formulated to systematically address the sustainable waste HDPE recycling process. A tailored parametric algorithm with the piecewise linearization method was used for effectively solving this nonconvex MINFP problem. The resulting Pareto-optimal curve gave a good-choice HDPE recycling process, which integrated HDPE preprocessing, HDPE pyrolysis using FBR and HZSM-5 catalyst in 500 °C, ultimate depropylenizer, mixed C4 separation, steam methane reforming, hydrocracking, steam turbine, and MEA capture. This sustainable process design had a unit NPV of 105 USD/t HDPE. Moreover, the levelized ReCiPe score of the most environmentally friendly design was 60 % of that of the most economically competitive design.

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