

Design of Plastic Waste Chemical Recycling Process Considering Uncertainty

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ABSTRACT

Chemical recycling of plastics is a promising technology to reduce carbon footprint and ease the pressure of waste treatment. Specifically, highly efficient conversion technologies for polyolefins will be the most effective solution to address the plastic waste crisis, given that polyolefins are the primary contributors to global plastic production. Significant challenges encountered by plastic waste valorization facilities include the uncertainty in the composition of the waste feedstock, process yield, and product price. These variabilities can lead to compromised performance or even render operations infeasible. To address these challenges, this work applied the robust optimization-based framework to design an integrated polyolefin chemical recycling plant. Data-driven surrogate model was built to capture the separation units' behavior and reduce the computational complexity of the optimization problem. It was found that when process yield and price uncertainties were considered, wax products became more favorable, and pyrolysis became the preferred reaction technology.

Keywords: Process Design, Design Under Uncertainty, Optimization, Polymers, Technoeconomic Analysis, Plastic Waste

INTRODUCTION

Global plastic waste has been on the rise, making efficient plastic recycling process design imperative [1]. Chemical recycling and upcycling strategies not only reduces the mismanaged plastic waste, but also has the potential to reduce the carbon footprint to meet sustainable goals [2].

One challenge in plastic recycling process design arises from uncertain feedstock compositions. The type and proportion of plastic waste can exhibit variations influenced by factors like geographic location, resulting in substantial differences in both economic and energy values [3]. In addition, feedstock compositions affect the strategies for plastic recycling. For instance, one advantage of the pyrolysis process is that it can easily handle a mixed plastic waste feedstock with different ratio, especially noncatalytic pyrolysis unit. However, most of other chemical recycling technologies, including hydrogenolysis, typically requires relatively pure feedstock after careful sorting or impurity removal to ensure good catalyst performance [4,5].

Polyolefins (PO), including polypropylene (PP) and polyethylene (PE), are main source of plastic waste. Their inert carbon-carbon backbones make it challenging to breakdown the long chains and produce valuable products [6]. Many reaction pathways have been developed recently to effectively deconstruct PO, among which pyrolysis and hydroconversion (i.e., hydrocracking and hydrogenolysis) have shown promising potentials. Thermal pyrolysis reactions operate at elevated temperature and shorter residence time, which generates products a wider distribution and more gas that are most useful as fuels [7]. Hydroconversion, on the other hand, operates at milder conditions and produces liquid hydrocarbon within the fuel or lubricant ranges [6,8].

Depending on the feedstock composition, different technologies operate at different conditions to produce different products [3,9]. As the product selectivity and the use of catalyst depend on the plastic waste type [3], feedstock variability could affect not only the process performance but also its feasibility.

Existing studies in process design of plastic recycling typically focus on a particular recycling strategy and

its operating conditions [9]. For instance, Hernandez *et al.* compared the costs and emissions for four waste LDPE treatment processes – gasification, pyrolysis, hydrogenolysis [4]. Bora *et al.* performed life cycle assessment and technoeconomic analysis on waste PP treatment processes and demonstrated that chemical recycling had low emissions but only profitable at large scales [10]. Zhao and You utilized the superstructure framework to optimize the net present value and greenhouse gas emissions of monomers, aromatic mixtures, and fuels production from waste HDPE [11].

An integrated plastic waste recycling technology selection and product separation provide opportunities for performance improvement. Moreover, it is important to consider the feedstock variability when designing such integrated chemical recycling facilities to ensure feasible operations among each connecting subunit. Robust optimization has been established as a computationally efficient framework to incorporate uncertainties and improves process performance [12]. Li *et al.* applied robust optimization to refinery production planning problem considering yield, cost, and price uncertainties [13].

This study proposes a methodology for the development of a chemical recycling facility for plastic waste. The design involves the selection of reaction technology and product separation guided by an optimization model. Since feedstock composition variability and product yield distribution are unavoidable in waste plastic treatment and affects the separation efficiencies, it is vital to guarantee feasible operation and good performance under these uncertainties. Rigorous process flowsheet simulations in Aspen Plus (Aspen Technology) [14] were carried out to obtain surrogate models of separation processes. Design decisions include chemical treatment technologies, distillation column design and unit connectivity. A robust optimization model is formulated to maximize profit under the worst case and ensure process feasibility (e.g., normal process operation) for all scenarios [6]. This robust optimization model will improve the chemical recycling process feasibility and performance under the worst uncertain case than the traditional deterministic optimization model [2,13]. Applying robust optimization instead of stochastic programming will also largely reduce the computational complexity, especially in the process design problem with high dimensionality arising from feedstock variability feedstock variability, yield uncertainty, and price fluctuation [15,16].

MODEL FORMULATION

Deterministic Superstructure Optimization

The superstructure elements for the chemical recycling facility includes plastic waste, other feedstocks, products, reaction technologies, and separation

alternatives (Figure 1). In this study, decisions are made on three levels. First, the combination of reaction or separation technologies is selected. Second, we determine the connectivity among feed, technologies, and products. Third, we decide the exact realization of a technology (e.g., distillation column operation conditions) by choosing an option. On this third level, incompatible connections, such as a liquid/solid stream entering a gas separator or an inappropriate reactant used by a particular reactor, could occur. Consequently, the connectivity of the superstructure elements is sorted *a priori* to eliminate those unproductive links. This step reduces the overall model size without cutting off potential candidate solutions [17].

As illustrated in Figure 2, the superstructure is connected with the inlet mixer and outlet splitters. The superstructure mass balance is established with the equations (1-4) for flow rate of each process stream going from i' to i for species k ($F_{i',i,k}^I$). Equation (1) specifies the mixer balance for $F_{i,k}^I$, the flow rate of an inlet stream into a superstructure element i for species k . The splitter balance is enforced in equation (2) for the outlet stream of a superstructure element $F_{i,k}^O$. While a splitter with split ratio to multiple outlet stream is possible, we choose to maintain the linearity of the problem by allowing exactly one destination i' for each superstructure element i with the binary variable $\eta_{i,i'}$. To ensure the feasibility of each unit, a capacity limit is imposed as shown in (5-7). The total flow rate F_i^T is decided from (5), and the technology capacity F_i^{CAP} is enforced with (6). A big M constraint (7) is used to ensure if a technology is not selected ($y_i = 0$), the capacity is 0.

$$F_{i,k}^I = \sum_{i' \in \text{inlets}(i)} F_{i',i,k}^I \quad \forall i \in I^{TECH} \forall k \quad (1)$$

$$F_{i,k}^O = \sum_{i' \in \text{outlets}(i)} F_{i,i',k}^O \quad \forall i \in I^{TECH} \forall k \quad (2)$$

$$\sum_k F_{i,i',k}^O \leq M \cdot \eta_{i,i'} \quad \forall i \in I^{TECH} \forall i' \in \text{outlets}(i) \quad (3)$$

$$\sum_{i' \in \text{outlets}(i)} \eta_{i,i'} = 1 \quad \forall i \in I^{TECH} \quad (4)$$

$$F_i^T = \sum_{k \in K} F_{i,k}^I \quad \forall i \in I^{TECH} \quad (5)$$

$$F_i^T \leq F_i^{CAP} \quad \forall i \in I^{TECH} \quad (6)$$

$$F_i^{CAP} \leq M \cdot y_i \quad \forall i \in I^{TECH} \quad (7)$$

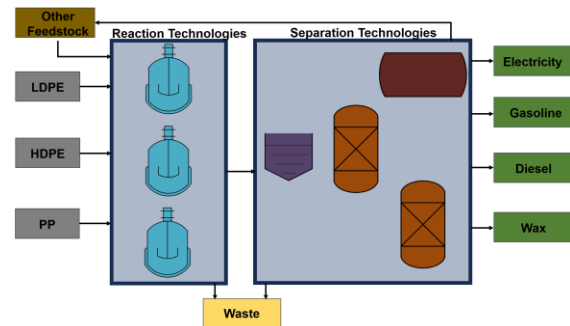


Figure 1. Elements of the chemical recycling plant superstructure.

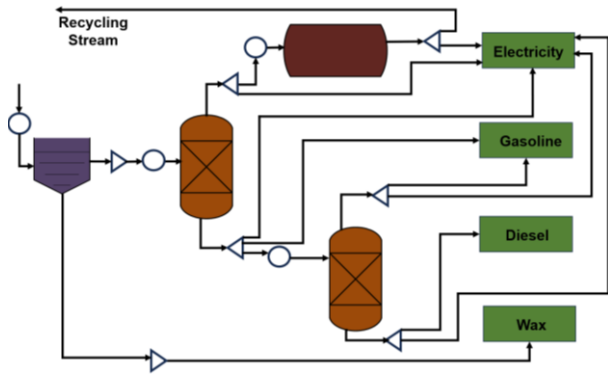


Figure 2. Possible connectivity of chemical recycling plant's separation units

The reaction conversion and selectivity of the chemical recycling technologies are taken from the literature to model stoichiometric reactor units in the superstructure. A basis in the reaction feed stream F_i^{Ref} and a conversion coefficient $\epsilon_{i,k}$ is used to determine the composition of the product streams as shown in (8). For some reactions, especially the hydroconversions [6,8], some undesired solid (e.g., coke) and gas formation are not well-characterized, leading to inaccurate estimation product yields thus violations of mass balance. To close the mass balance gap, we make a conservative assumption by including a waste stream that is not usable in the downstream operations (9). Admittedly, this assumption may not reflect the actual reaction. Thus, the effects of wax yield uncertainty are addressed in the robust optimization.

$$F_{i,k}^I + \epsilon_{i,k} \cdot F_i^{Ref} = F_{i,k}^P \quad \forall i \in I^{RXN} \quad \forall k \in Products(i) \quad (8)$$

$$\sum_k F_{i,k}^I - \sum_{k \neq WASTE} F_{i,k}^P = F_{i,WASTE}^P, \quad \forall i \in I^{RXN} \quad (9)$$

All separation units in this study are designed to achieve sharp separation (i.e., separators always isolate nearly all the light species to the light outlet stream $F_{i,k}^{OL}$ and the heavy species to the heavy outlet stream $F_{i,k}^{OH}$). The gas and fuel range products are separated in distillation columns with a sequence based on the target boiling points. To ensure that sharp separation is attainable with our design, surrogate models (12) are built to estimate the reflux ratio needed for 99% purity and the associated utilities from rigorous Aspen Plus process simulation. In practice, the distillation columns cannot deviate too much from the nominal reflux ratio once designed and installed. To ensure the operating feasibility, different design options are provided for the same distillation technology but at most one will be active as shown in (13). A 30% flexibility around the reflux ratio at nominal conditions is allowed for each design option p as presented in (13)-(15). When an option is selected ($w_{i,p} = 1$), two big M

constraints are used for to ensure the reflux ratio does not deviate from the design value for more than 30% (14,15).

$$F_{i,k}^I = F_{i,k}^{OL} \quad \forall i \in I^{SEP} \quad \forall k \in light(i) \quad (10)$$

$$F_{i,k}^I = F_{i,k}^{OH} \quad \forall i \in I^{SEP} \quad \forall k \in heavy(i) \quad (11)$$

$$f_i(Light, Heavy) = (RR_i, Q_i^{cool}, Q_i^{heat}) \quad \forall i \in I^{SEP} \quad (12)$$

$$y_i = \sum_p w_{i,p} \quad \forall i \in I^{SEP} \quad (13)$$

$$-M(1 - w_{i,p}) + 0.7RR_p^{nom} \leq RR_i \quad \forall i \in I^{SEP} \quad (14)$$

$$RR_i \leq M(1 - w_{i,p}) + 1.3RR_p^{nom} \quad \forall i \in I^{SEP} \quad (15)$$

A supply constraint is imposed to the plastic waste feedstock in (16).

$$\sum_{i' \in outlets(i)} F_{i,i',k} \leq F_{j,k}^{SUP} \quad \forall i \in I^{Plastic} \quad \forall k \in plastic \quad (16)$$

For fuel range products, we impose a maximum for the olefin content [17] as a product requirement (17). In addition, we specify the limit of product quantities below or above the typical boiling point ranges for fuel product such as gasoline and diesel (18,19). Equations (18,19) represent a linear simplification of the blending rule by only considering boiling point and olefin content, although more rigorous blending rule can be incorporated.

$$\sum_{k \in K^{olefin}} F_{i,k}^{prod} \leq \phi^{olefin} \sum_{k' \in K} F_{i,k'}^{prod} \quad \forall i \in I^{Fuel} \quad (17)$$

$$\sum_{k \in K^{BPLO}} F_{i,k}^{prod} \leq \phi_k^{klo} \quad i \in I^{Fuel} \quad (18)$$

$$\sum_{k \in K^{BPU}} F_{i,k}^{prod} \geq \phi_k^{kup} \quad i \in I^{Fuel} \quad (19)$$

The general deterministic formulation of this chemical recycling process design is presented as a mixed-integer linear programming (MILP) problem in equation (20):

$$\begin{aligned} \max \quad & (R^{PROD} + R^{ELEC} - C^{CAPEX} - C^{OPEX}) \\ \text{s. t.} \quad & \text{Superstructure Mass Balance} \\ & \text{Technology Constraints} \\ & \text{Product Rules} \\ & \text{Plastic Waste Feed Constraints} \\ & \text{Process Economics} \end{aligned} \quad (20)$$

Surrogate Model for Distillation Columns

The distillation column surrogate model is an essential part of the superstructure optimization to create a mapping between the inlet flow rates for both the light and heavy components and the actual reflux ratio, condenser duty, and reboiler duty. Artificial neural network (ANN) is selected as surrogate models given its excellent fitting performance and its ability to maintain the linearity of the problem. We refer to our previous work and use Rectified Linear Unit (ReLU) as the activation function with a feed-forward ANN [18]. The predicted value a_m^k of layer k and node m is calculated using a linear combination of the values from the previous layer as shown in (21).

The activation function ReLU, $z_n^k = \max(0, a_n^k)$, is modelled using the big M constraints as shown in (22a-22d)

$$a_m^k = W_{n,m}^{k-1} z_n^{k-1} + b_m^{k-1} \quad (21)$$

$$-M \cdot (1 - \delta_m^k) \leq a_m^k \quad (22a)$$

$$a_m^k \leq M \cdot \delta_m^k \quad (22b)$$

$$0 \leq z_m^k \leq M \cdot \delta_m^k \quad (22c)$$

$$a_m^k \cdot (1 - \delta_m^k) \leq z_m^k \leq a_m^k + M \cdot (1 - \delta_m^k) \quad (22d)$$

Robust Optimization

The uncertainty in this study arises from three sources. The first source is the dynamic supply of sorted bale plastic waste, which leads to uncertain feedstock flowrates. The second source arises from the product yield. For instance, not all solid products of hydroconversion of recycled plastic could be treated as waxes [6]. Instead, depending on the reaction condition, some solid plastic waste could remain unreacted or form coke. To reflect this, we incorporate a process yield uncertainty for waxes in hydrocracking and hydrogenolysis. Furthermore, the product selling price uncertainty is also included to reflect the price volatility of the petroleum refinery products. The fuel range product tends to be more volatile than waxes [19]. Therefore, a larger deviation should be considered.

To formulate the robust counterpart of the problem, we refer to the work of Li. *et al.* [13], in which the robust counterpart formulations have been derived for linear and mixed-integer linear programming with different uncertainty set. In this study, we consider box uncertainty set for all uncertain parameters. The robust counterpart for an inequality constraint (23) when considering left-hand-side (L.H.S.) uncertainty is presented in (24), and right-hand-side (R.H.S.) uncertainty in (25).

$$\sum_j a_{i,j} x_j \leq b_i \quad (23)$$

$$\sum_j a_{i,j} x_j + \sum_j \widehat{a}_{i,j} |x_j| \leq b_i \quad (24)$$

$$\sum_j a_{i,j} x_j + \widehat{b}_i \leq b_i \quad (25)$$

In this study, uncertainty parameters for process yield and product sale price occurs in equations instead of inequalities, which drastically restricts the feasible space of mathematical model and often causes infeasibility [16]. Therefore, we use the inequality constraints instead for process yield and sale price as presented in previous work in petroleum refinery by Leiras *et al.* [15] as shown in (26,27). Using this upper bound on wax produced and product sales, we now have L.H.S. uncertainty instead. While we acknowledge this reformulation creates a relaxation of the original problem mathematically, the profit objective typically encourages producing more products when possible. Therefore, these reformulated inequalities (26,27) are likely to be active at optimal solution. The robust formulation is obtained by applying the

properties of box uncertainty set as presented in (24,25).

$$F_{i,WAX}^{FEED} + \epsilon_{i,WAX} \cdot F_i^{Ref} \geq F_{i,WAX}^{PROD} \quad \forall i \in I^{RXN} \quad (26)$$

$$Revenue^{PROD} \leq Price \cdot F^{PROD} \quad (27)$$

ILLUSTRATIVE EXAMPLE

In this example, we considered three categories of reaction technologies (i.e., hydrocracking [6], hydrogenolysis [8], and pyrolysis [7]), three types of plastic waste streams (i.e., PP, LDPE, and HDPE), one solid separation, two distillation (each with five options), and a pressure-swing absorption unit for hydrogen recovery. The details for the technologies are summarized in Table 1-3. For reaction technologies that allow mixture plastic waste, we enforced the feed composition to be the same as reported from experiments in the literature. Gasoline, diesel, and waxes are selected as the product. Additionally, another stream for electricity generation is available for mixture outlet streams that do not meet the product requirement for fuels.

Table1: Reactions considered

Code	Reaction Type	Feed
R1A	hydrocracking	LDPE
R2A	hydrocracking	HDPE
R3A	hydrocracking	PP
R1B	hydrogenolysis	PP
R2B	hydrogenolysis	PP/PE(1:1)
R1C	pyrolysis	PE
R2C	pyrolysis	PE/PP(3:2)
R3C	pyrolysis	PE/PP(2:3)
R4C	pyrolysis	PP

Table 2: Separations considered

Code	Separator Type	Separation Task
FL1A	solid/fluid	fluid/wax
DT1A	distillation	C4/C5
DT2A	distillation	C12/C13
PSA1A	pressure swing absorption	hydrogen/alkanes

Table 3: Reflux ratios at nominal operating conditions for different options

Option	1	2	3	4	5
Reflux Ratio(molar)	1	1.83	3.43	6.37	11.83

The data for the surrogate model training for distillation columns were obtained by running sampled simulations in Aspen Plus [14] with Python interface. The model was trained in PyTorch [20]. The ANN models for both distillation columns had 4 layers with 15, 18, 12 neurons in the hidden layers and 3 neurons at the output

layer. The R^2 of the ANN for DT1A was 0.9586 and for DT2A, 0.9675.

The operational basis of this example was based on the previous techno-economic analysis [4,11] with a supply of 12 tonne/hr PP, 10 tonne/hr HDPE, and 2.5 tonne/hr LDPE. The cost for the plastic collection and sorting depends on the population density and geographic variation [9]. The plastic waste collection and sorting cost was estimated in the work of Hernandez *et al.* [4] to be in the range of 250 to 700 \$/tonne. In this example, we assumed an overall cost for the sorted plastic to be \$500/tonne. The price for waxes was estimated to be \$1000/tonne [21]. Moreover, the fuel price at nominal condition was estimated to be \$960/tonne for gasoline [19] and \$920/tonne for diesel [19]. A wax yield of 0.5 was assumed for hydroconversions. The associated uncertainty type and deviations from the nominal value considered in this example is shown in Table 4.

Table 4: Uncertain parameters for illustrative example

Uncertain Parameter	Type	Deviation (%)
Plastic supply	RHS	10
Wax yield	LHS	40
Fuel price	LHS	30
Wax price	LHS	10

All optimization models were implemented in Pyomo [22] and solved with Cplex 22.1 solver [23] on a computer with Intel Xeon E-2274G CPU @ 4.00GHz 32 GB RAM. The deterministic model was solved under nominal condition, and the robust formulation was applied to include the uncertainty set in Table 4.

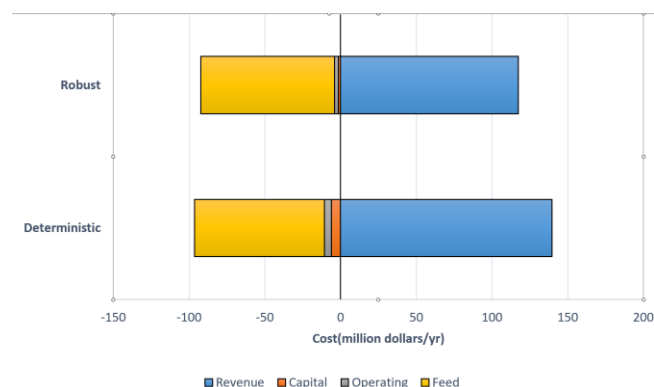


Figure 3. Cost breakdown of the plastic chemical recycling system.

As shown in Figure 3, under nominal condition, the optimal integrated chemical recycling process has a revenue of 139.58 million dollars/year and a total cost of 96.40 million dollars/year. The feedstock cost comprises of 89% of the total cost, which accounts for all upstream

cost for plastic waste collection and sorting. The profit of the process is 43.18 million dollars/year. However, when the uncertainties are considered in the robust formulation, the total revenue of the best design dropped to 117.45 million dollars/year with a total cost of 92.5 million dollars/year. This leads to a profit of 25.95 million dollars/year.

Under the nominal condition, the distillation column DT1A operates at a 6.64 molar reflux ratio, and operating condition option 4 is selected. For distillation column DT2A, the actual molar reflux ratio is 2.24, and option 2 is selected. When robust optimization is performed, the actual reflux ratio for DT1A becomes 0.07, and option 1 is selected. The technology DT2A is not selected at all as no gasoline product is pursued as shown in Figure 5.

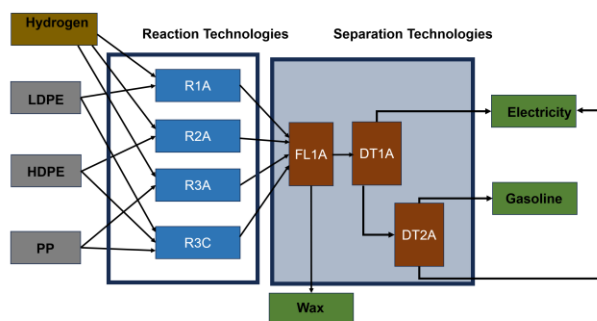


Figure 4. Superstructure under the nominal condition

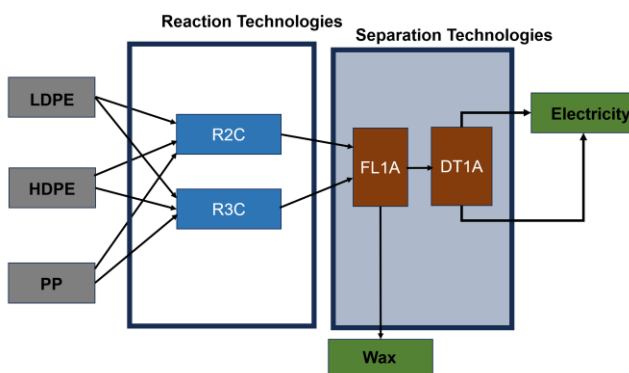


Figure 5. Superstructure under the robust formulation

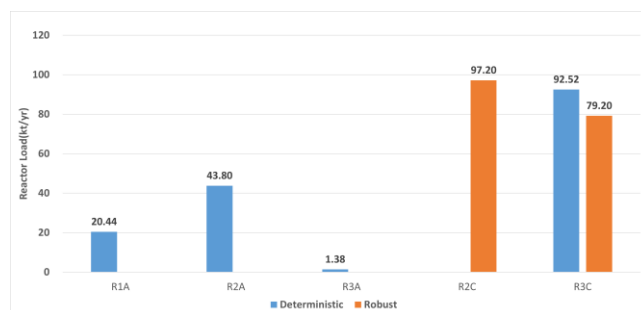


Figure 6. Reactor loads for process designs from both deterministic and robust optimization

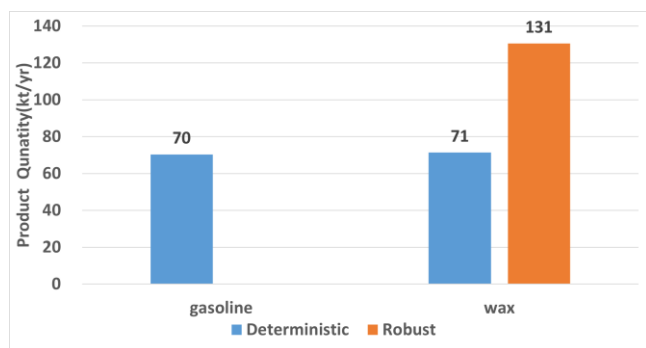


Figure 7. Product quantities for process designs from both deterministic and robust optimization.

As illustrated in Figure 4, the reaction of a combination of hydrocracking (R1A, R2A, R3A) and mixed olefin pyrolysis(R3C) is selected in the nominal case, and the reactor load is presented in Figure 6. When uncertainties are considered, only mixed olefin pyrolysis is chosen (R2C, R3C). The product produced changes from a combination of gasoline and wax to only wax. This change occurs primarily as a result of the price volatility of fuel products, which makes the wax product relatively more profitable than the nominal condition. In addition, the hydrocracking reactions are less economically favorable after the uncertainty in the wax yield of hydroconversions is included.

CONCLUSIONS

In this work, we have proposed the robust optimization framework for designing plastic waste valorization system. We used an illustrative example to demonstrate the how the feedstock supply availability, process yield, and product price uncertainties affect the optimal process design and operation. In the future, this model could be extended to include more considerations including more types of products, upgrading technologies, upstream sorting process, reaction technologies, and more kinds of plastic waste.

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