PROCEEDINGS OF THE 30th European Symposium on Computer Aided Process Engineering (ESCAPE30), May 24-27, 2020, Milano, Italy

Modeling Framework for Joint Product and Process Synthesis with Material Recovery Opportunities

Ana Somoza-Tornos[a], Qi Chen[b], Moisès Graells[a], Antonio Espuña[a], Ignacio E. Grossmann[b*]

[a]*Department of Chemical Engineering, Universitat Politècnica de Catalunya Escola d'Enginyeria de Barcelona Est, C/ Eduard Maristany 16, 08019 Barcelona, Spain*
[b]*Center for Advanced Process Decision-making, Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA*
grossmann@cmu.edu

Abstract

The circular economy paradigm requires process synthesis to be expanded beyond the consideration of production activities aimed at market needs and to integrate valorization processes upcycling waste from different sources (industrial and urban). With this aim, this contribution presents a modeling approach for the joint synthesis of production processes and products from a waste-to-resource perspective.
The system is modeled through a superstructure with features from state-task network (i.e. the activation/deactivation of units) and state-equipment network (i.e. multiple tasks in a unit) representations. The problem is formulated using a Generalized Disjunctive Programming approach (GDP).
The proposed approach is tested with a case study addressing the synthesis problem of polyethylene pyrolysis, as a central step required to address the need to close the associated material loops. Decisions are made on the separation and reuse of the pure or mixed light gases from the reactor outlet (material reuse vs. energy valorization).
Results demonstrate the ability of the proposed approach to represent alternatives that cannot be considered if only STN or SEN models were used.

Keywords: process modeling, synthesis, superstructure, optimization, generalized disjunctive programming, circular economy.

1. Introduction

State-task network (STN, (Kondili et al., 1993)) and state-equipment network (SEN, (Smith, 1996)) are two process representations commonly used as a base for the superstructure representation required to address the conventional problem of process synthesis. While the STN representation is easier to formulate, the SEN representation is more suitable for modeling equipment networks, as it reduces the number of process nodes and prevents zero-flow singularities (Chen and Grossmann, 2017).
However, both conceptual models generally rely on the premise that product specifications are narrowly bounded (i.e. final products are single-component with a defined purity), and fail to consider other decisions that would affect the final result (i.e. solutions in which intermediate products or mixtures may be sold or recycled into the

process). This problem becomes crucial in the synthesis of processes addressing the circular economy paradigm, where material recovery alternatives are numerous and diverse. Hence, this work presents a novel modeling approach for the optimal synthesis of processes with flexible product composition, including equipment activation/ deactivation, and the possibility of selling/recycling mixed streams.

This is particularly interesting for the application of circular economy principles to process design, which has been gaining importance in recent years (Avraamidou et al., 2020). Processes for the chemical upgrading and recycling of polymers, such as the pyrolysis of plastics, lead to hydrocarbon mixtures similar to those from crude oil cracking but with different compositions. The two main alternatives for these products include their use as fuels (i.e. waste-to-energy, Honus et al., 2016) and their separation to recover the monomers that can be used to produce new chemicals or polymers (Hong and Chen, 2017), which results in a more efficient use of valuable resources and may increase incentives for recycling and closing material loops.

2. Problem statement

The problem addressed in this paper can be stated as follows: given is a set of raw materials (usually subproducts/waste) and process alternatives (equipment and tasks), the objective is to find the path to convert these materials into the most valuable resources, taking into account current market requirements.

In order to achieve this objective, these elements have to be represented in a flexible superstructure that considers different alternatives for pure or mixed products (i.e. selling or recycling) and also different flowsheeting alternatives and equipment design.

3. General framework for joint process and product synthesis of

The proposed general framework for addressing the synthesis problem consists of a three-step approach based on the work by Yeomans and Grossmann (1999): superstructure representation, modeling (Generalized Disjunctive Programming - GDP), and model resolution.

3.1. Superstructure representation

Separation processes are generally modeled considering that the inlet is separated in all the products that integrate it. STN leads to easier problem formulations, whereas SEN is more easily solved since it prevents zero-flow singularities (Chen and Grossmann, 2017). However, the synthesis of waste-to-resource processes requires a more flexible superstructure representation of separation sequences, including the activation and deactivation of equipment (as in STN) and the flexible assignment of tasks to equipment (as in SEN). This is done through the implementation of the most general form of SEN network (Yeomans and Grossmann, 1999) which does not avoid zero-flow singularities.

A generic example of superstructure representation of a process flowsheet including flexible product composition and material recovery is shown in Figure 1.

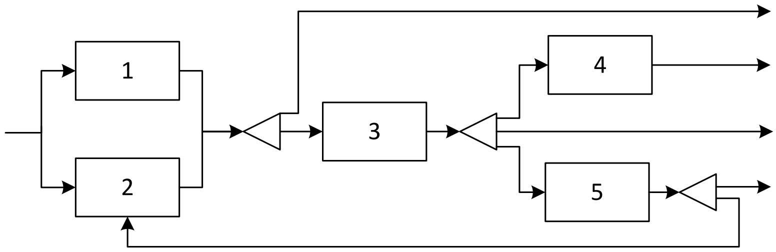

Figure 1. Example of superstructure for joint product and process synthesis.

3.2. GDP formulation

The superstructure defined in the previous step is now modeled and formulated using GDP (Raman and Grossmann, 1994). Let $j \in J$ define the set of equipment in the superstructure and $k \in I_j$ the set of tasks that can be performed in each equipment j. x_j and z_{jk} denote the continuous variables representing the operating conditions of the system, while the Boolean variables Y_j and W_{jk} represent whether equipment j is active and whether task k is assigned to it, respectively. The resulting formulation is as follows:

$$\min z = \sum_{j \in J} c_j + f(x_j, z_{jk}) \tag{1}$$

$$\text{s.t.} \quad f(x_j, z_{jk}) \leq 0 \tag{2}$$

$$\begin{bmatrix} Y_j \\ \bigvee_{k \in I_j} \begin{bmatrix} W_{jk} \\ f_{jk}(x_j, z_{jk}) \leq 0 \\ c_j = \gamma_{jk} \end{bmatrix} \end{bmatrix} \vee \begin{bmatrix} \neg Y_j \\ x_j = z_{jk} = 0 \\ c_j = 0 \end{bmatrix} \quad \forall j \in J \tag{3}$$

$$\Omega(W_{jk}) = True \tag{4}$$

$$Y_j \in \{True, False\} \quad \forall j \in J \tag{5}$$

$$W_{jk} \in \{True, False\} \quad \forall j \in J, k \in I_j \tag{6}$$

The objective function to be minimized (Eq. 1) includes the fixed cost associated to the active equipment units and a function of the continuous variables (i.e. variable costs and income from selling the products). Algebraic constraints in Eq. (2) are equalities and inequalities that must be satisfied for any realization of the discrete variables, typically including mass balances that define the connections among the nodes of the superstructure. On the other hand, constraints that are inherent to equipment activation and task assignments are modeled in nested disjunctions. The external ones are based on the existence of equipment j, while the internal ones define task selection. Thus, if equipment j is active ($Y_j = True$) and task k is selected ($W_{jk} = True$), constraints $f_{jk}(x_j, z_{jk}) \leq 0$ are applied and the related fix costs are considered in the objective function $c_j = \gamma_{jk}$. Conversely, if equipment j is not selected ($Y_j = False$) continuous variables and fix costs are set to 0. Finally, logical constraints among the nodes of the superstructure are given by $\Omega(W_{jk})$ (Eq. (4)). These include enforcements of consecutive tasks in order to meet recipe-based constraints.

3.3. Model resolution

The model is implemented in Pyomo and solved with DICOPT after its reformulation to a MINLP using the Big M method.

4. Case study

The proposed framework has been applied to the synthesis of the process of polyethylene pyrolysis for the recovery of hydrocarbons. Experimental data from the literature is used to model the outlet from the pyrolysis furnace. Kannan et al. (2014) reported high conversions (>99%) of the polymer to gas when operating at 1000ºC, leading to outlet compositions of: 5% methane, 46% ethylene, 18% propylene, 3% propyne, 2% 1-butene, 13% 1,3-butadiene and 13% benzene. The main objective is to identify to which extent the gas resulting from the pyrolysis of polyethylene at such conditions should be separated into its compounds, according to the cost of separation and the market price for pure or mixed compounds. The model should also identify if any of the streams could be used as fuel to satisfy the energy requirements of the furnace used to maintain the operating conditions.

5. Results and discussion

In this section, the results for the synthesis of the case study are presented following the methodology described in section 3.

5.1. Superstructure representation

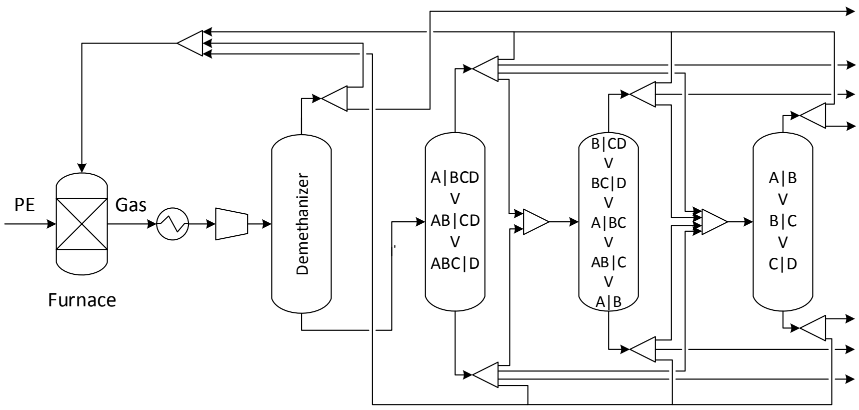

Figure 2. Superstructure representation of the process.

Figure 2 shows the superstructure for the proposed case study. The outlet of the pyrolysis reactor is cooled and compressed to enter the distillation sequence where the different hydrocarbons may be recovered. For the sake of simplicity and due to the different boiling point of methane compared to the rest, the stream is demethanized before entering the distillation sequence. After this step, a four component mixture distillation train is considered, in order to split the inlet into its fractions of ethylene (A), propylene (B), 1,3-butadiene (C) and benzene (D). Propyne and 1-butene are recovered with 1,3-butadiene since their low concentration would not justify two extra separation stages. The first column considers the three possible tasks for the first level separation of the four-component mixture. The second one includes the three-component separations of the streams resulting from the previous column, plus the separation A|B in case AB|CD is selected in column one. Finally, column 3 can perform the two-component separation of outlet streams from column two. All three distillation columns can be active or inactive, but the existence of one implies that the previous ones need to exist. All outlet streams can be introduced to the next separation level, sold as final product, or reused in the process as fuel for the furnace.

5.2. Model formulation

The model is formulated following the GDP described in section 3.2 with the following considerations:

- The objective function is the profit maximization taking into account: the income for product sales (proportional to its purity), fix and variable costs for the active distillation columns, and fresh fuel savings.
- $f(x_j, z_{jk}) \leq 0$ include the mass balances at the nodes of the superstructure (e.g. the distillate of column one can be sold as a product, used as fuel at the furnace or go to column two if AB or ABC mixes are produced).
- $f_{jk}(x_j, z_{jk}) \leq 0$ represent the equations that depend on the column activation and task selection (e.g. mass balance of the distillation columns or reflux ration calculation).
- $\Omega(W_{jk})$ denotes the logical constraints that should be enforced (e.g. column 3 can only be active if column 1 and 2 are also active).

5.3. Solution

Figure 3 depicts the optimal solution for the flowsheet design for the material recovery from polyethylene pyrolysis. In this particular case all units were selected, so zero-flow singularities are not present.

The methane from the gas demethanization is sold, and the bottoms are sent to column 1. Here, task A|BCD is active, leading to the production of ethylene. Likewise, propylene and 1,3-butadiene are recovered in the distillates of columns 2 and 3, respectively. Thus, direct distillation was found to be the optimal option. Ethylene, propylene and benzene are sold, while 1,3-butadiene is burned as fuel at the furnace due to its low purity.

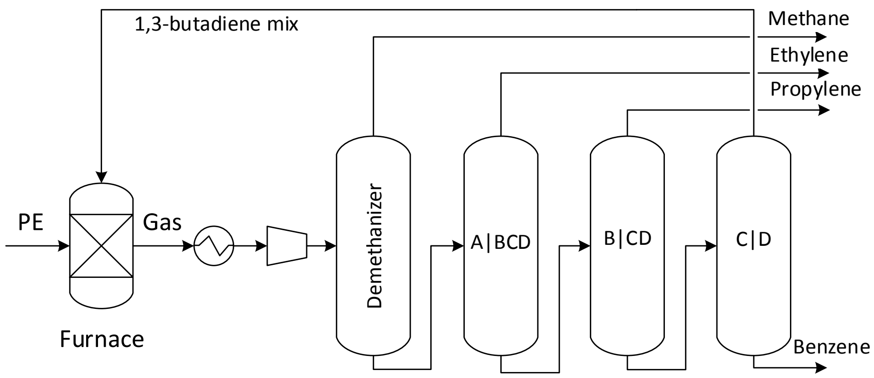

Figure 3. Optimal flowsheet design for the material recover from polyethylene pyrolysis.

6. Conclusions

This paper has introduced a general framework to represent, model and solve the joint product and process synthesis problems resulting from the consideration of waste-to-

resource transformations. To achieve this objective, the work has followed the three-step method proposed by Yeomans and Grossmann (1999). First, the model is represented through the generalized version of a SEN, including task selection and equipment activation and deactivation to address the singularities of processes for material recovery. Second, we formulate the model as a GDP. Finally, the model is transformed into a MINLP through the Big M method and solved in Pyomo/DICOPT. The capabilities of the model have been tested through its application to the synthesis of a flowsheet for the recovery of hydrocarbons from the pyrolysis of polyethylene. The proposed methodology has been proven useful to identify the optimal extent of separation and the most economically profitable products in a systematic way.
Future work will include the implementation of decomposition techniques to address the cases which present zero-flow singularities.

Acknowledgements

Financial support received from the Spanish "Ministerio de Economía, Industria y Competitividad" and the European Regional Development Fund, both funding the research Project AIMS (DPI2017-87435-R) is fully acknowledged.

Ana Somoza-Tornos thankfully acknowledges financial support received from the Spanish Ministry of Education, Culture and Sport (Ayuda para la Formación de Profesorado Universitario - FPU15/02932).

References

Avraamidou, S., Baratsas, S.G., Tian, Y., Pistikopoulos, E.N., 2020. Circular Economy - A challenge and an opportunity for Process Systems Engineering. Comput. Chem. Eng. 133, 106629. https://doi.org/10.1016/j.compchemeng.2019.106629

Chen, Q., Grossmann, I.E., 2017. Recent Developments and Challenges in Optimization-Based Process Synthesis. Annu. Rev. Chem. Biomol. Eng. 8, 249–283. https://doi.org/10.1146/annurev-chembioeng-080615-033546

Hong, M., Chen, E.Y.X., 2017. Chemically recyclable polymers: A circular economy approach to sustainability. Green Chem. https://doi.org/10.1039/c7gc01496a

Honus, S., Kumagai, S., Němček, O., Yoshioka, T., 2016. Replacing conventional fuels in USA, Europe, and UK with plastic pyrolysis gases – Part I: Experiments and graphical interchangeability methods. Energy Convers. Manag. 126, 1118–1127. https://doi.org/10.1016/j.enconman.2016.08.055

Kannan, P., Al Shoaibi, A., Srinivasakannan, C., 2014. Temperature effects on the yield of gaseous olefins from waste polyethylene via flash pyrolysis. Energy and Fuels. https://doi.org/10.1021/ef500516n

Kondili, E., Pantelides, C.C., Sargent, R.W.H., 1993. A general algorithm for short-term scheduling of batch operations—I. MILP formulation. Comput. Chem. Eng. 17, 211–227. https://doi.org/10.1016/0098-1354(93)80015-F

Raman, R., Grossmann, I.E., 1994. Modelling and computational techniques for logic based integer programming. Comput. Chem. Eng. 18, 563–578. https://doi.org/10.1016/0098-1354(93)E0010-7

Smith, E.M., 1996. On the optimal design of continuous processes, Ph.D. Dissertation, under supervision of C. Pantelides. Imperial College of Science, Technology and Medicine, London, UK.

Yeomans, H., Grossmann, I.E., 1999. A systematic modeling framework of superstructure optimization in process synthesis. Comput. Chem. Eng. 23, 709–731. https://doi.org/10.1016/S0098-1354(99)00003-4