**Techno-economic and environmental assessment of plastic waste pyrolysis: From a linear to a circular economy**

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Abstract

Single use plastics, although beneficial, pose environmental challenges when discarded. Chemical recycling offers a sustainable solution by breaking down plastic waste into monomers, allowing for continuous production of plastic without downgrading its properties. In this contribution, a lab-sized pyrolysis reaction of polypropylene waste was transformed into an industrial process using a simulation-optimization approach with Aspen HYSYS. This baseline simulation, which includes energy integration, was used as a basis to create nine alternative scenarios using different technologies. In addition, a circularity metric was used to assess the degree of material circularity obtained in the pyrolysis process. Heat integration allowed for a 61% reduction in energy costs of the process. In addition, our study proves that product recovery from waste polypropylene pyrolysis could be 94% circular, which can play a significant role in reduction of raw material extraction and carbon emissions. Pyrolysis has the potential of processing a wide range of plastic types, making it a promising technology to combat and add value to plastic waste.

**Keywords**: Plastic Waste, Pyrolysis, Circular Economy, Process Systems Engineering

* 1. Introduction

Since the mid-20th century, plastics have been known for significantly impacting our lives. However, they also pose a substantial environmental threat, with over 8300 million tons produced globally, 70% becoming waste, and 84% of that waste finding its way into the environment (Smet et al., 2019). In Europe, approximately 32% of plastic waste is recycled, 43% is incinerated, and the remaining 25% is sent to landfills (Mortensen & Tange, 2021). However, both landfills and incineration pose long-term threats to the environment, with incineration offering a minor detour with energy recovery (Payne et al., 2019). Recycling, mostly done mechanically today, cannot preserve plastic properties, resulting in a product that is no longer suitable for the original application. Hence, it can only reduce partially the accumulation of plastic waste and raw material extraction.

The need to incorporate the circular economy concept into advanced recycling technologies is a key driver for adding value to plastic waste, thereby lessening carbon emissions as well as the reliance on fossil-based feedstock to produce commodity plastics. In this context, chemical recycling emerges as a promising alternative. Chemical recycling, unlike mechanical, enables the return to the monomer, ensuring the production of high-quality plastic further down the value chain. This form of recycling can be considered as a closed-loop process, where the recycled plastics are used completely and entirely to produce again plastics (the same or others) with the original properties. Pyrolysis, a form of chemical recycling, operates at moderate to high temperatures in the absence of oxygen, breaking down the polymer chain into shorter carbon-based chains (Stallkamp et al., 2023). Prior research has examined the use of pyrolysis oil extracted from polypropylene waste as a promising diesel substitute, finding that it has the potential to contribute significantly to mitigating the increasing challenge of waste disposal (Pacheco-López et al., 2021). Somoza-Tornos et al. (2020) utilized techno-economic and life cycle assessment to compare different end-of-life alternatives for polyethylene waste, concluding that pyrolysis can be both economically and environmentally beneficial. However, these contributions did not assess the degree of circularity that could be achieved through the pyrolysis process.

In this contribution, we study the conversion of plastic waste into value-added chemicals from an economic and an environmental perspective, the latter addressed through a circularity metric. To this end, we created a preliminary design of an industrial process for the chemical recycling of waste polypropylene (PP) and used it as a case study to explore the degree of circularity that can be achieved under different scenarios, and at which cost.

* 1. Methodology

We departed from lab scale experimental data of a pyrolysis reaction, which we turned into the core of a scaled-up industrial process, that we designed using a simulation-optimization approach with Aspen HYSYS, with Peng-Robinson as fluid package. The real-life model generated potential products and was further optimized. Primarily, the pyrolysis of polypropylene (PP) waste takes place at 700°C, yielding a myriad of products (Honus et al., 2016). Major components were retained, representing 92.9% of the total mass, and we normalized their composition to add to 100%. Hence, the reaction products considered are, on a mass basis: 6.7% methane, 10.4% ethylene, 7.6% ethane, 47.1% propylene and 28.3% 1-butene. Finally, the stoichiometric coefficients for the adjusted pseudo-reaction were obtained considering a molecular weight of 12,000 g/mol for PP, and a composition of 85.6% carbon and 14.4% hydrogen (equation (1)).

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| $$0.0833 PP\rightarrow 4.176 CH\_{4}+3.697 C\_{2}H\_{4} +2.514 C\_{2}H\_{6}+11.188 C\_{3}H\_{6}+5.042 C\_{4}H\_{8}$$ | (1) |

We assumed that the plant operates 330 days a year and 24 hours a day, with a feed rate of 19,000 kg/h. The pyrolysis furnace modelled as a conversion reactor, uses diesel as a fuel and achieves a 100% conversion, as reported from lab experiments.

After the reactor, a multi-stage compressor was added to minimize the cryogenic operating temperatures in the condensers of the distillation columns downstream. The compressor operating conditions (inner temperatures and stage pressures) were optimized to minimize the annualized cost using Aspen HYSYS optimizer tool. For economical analysis, the following unit costs, retrieved from literature, were considered: plastic waste (175 €/ton), sorting (314.56 €/ton), methane (1.43 €/litre), ethylene (1.12 €/kg), ethane (0.26 €/kg), propylene (0.89 €/kg), 1-butene (1.13 €/kg), natural gas (0.07 €/kWh), gasoline (1.63 €/litre), ethanol (1.93 €/litre), diesel (1.59 €/litre), steam (2.29·10-4 €/kJ), electricity (0.12 €/kWh), cooling water (1.4 €/kJ).

After the compressor, a separation train, comprising four distillation columns, was incorporated to ensure 98% recovery for the five products. The optimal sequence for this train was obtained based on the minimum vapour flow method (Modi & Westerberg, 1992). In addition, the design of each column was optimized separately to minimize its total annualized cost (Towler & Sinnott, 2008), using the number of stages as the decision variable. With pyrolysis being an energy intensive process, energy integration was implemented to reduce energy consumption. Applying the pinch methodology is essential to optimize the recovery of process heat, enabling the ideal equilibrium between capital (size of the heat exchanger network) and operational expenditure (utility cost).

With the process simulation for the baseline case at hand, nine different scenarios were created by varying different technological decisions. On the one hand, four different fossil-based fuels were employed in the furnace: diesel (baseline), ethanol, gasoline and natural gas. Five additional scenarios were also explored where pyrolysis products were individually reused as fuel in the furnace, instead of sold as a products. No additional fuel was needed to fulfill the furnace requirements in all these cases, which hilighlights the energetic value of pyrolysis gases. In addition, the nine scenarios consider the implementation of carbon capture and sequestration (CCS) in the pyrolysis furnace, with a capture rate of 90% of the CO2 in the flue gas. The application of CCS aligns with environmental goals of mitigating carbon emissions and contributing to the overall reduction of greenhouse gases, thereby addressing climate change concerns.

Finally, we introduce the concept of the material circularity index (MCI), which serves the purpose of facilitating the assessment of material circularity. This index could help decision-makers to identify processes with a high degree of circularity, emphasizing on the extension of resource lifespan, and reducing the need for raw material extraction. The MCI (equation 3) employs a straightforward scale ranging from 0 to 1, where 0 represents a linear process, and 1 signifies a fully circular one, indicating the potential for both resource and product lifespan extension. In our case, the numerator represents all the products obtained from the pyrolysis, while the denominator denotes the amount of feed (i.e., sorted waste polypropylene) (Circle Economy & Deloitte, 2023). We considered all products recovered since they can be reused as virgin material in other processes, either to produce PP or other products.

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| $$Material Circularity Index (MCI)=\frac{Product mass recovered}{Total material consumption}$$ | (3) |

* 1. Results and discussion
		1. *Process simulation and economic performance*

The optimized process flowsheet is depicted in Figure 1, where we intentionally avoided most of the heat exchanger network to facilitate visualization.



*Figure 1: Flowsheet of the polypropylene pyrolysis process*

In this case, we found the direct separation sequence to be the optimal, where we first separate methane (stream A), then ethylene (B), then ethane (C) and finally propylene (D) and 1-butene (E). Purities of the major components in each product stream range 91%-99% on a molar basis. As aforementioned, we next use the results (mass and energy balance) from this baseline simulation to explore the performance of nine alternative scenarios in terms of cost and circularity.

We next turn our attention to a comprehensive economic assessment of the nine scenarios considered (Figure 2). Bars provide the breakdown of process expenditures, with product sales depicted as negative values. The final net cost is given by markers over the bars.

*Figure 2: Annual cost breakdown of the optimized pyrolysis process for the nine scenarios studied.*

Importantly, we found no scenario with the capacity to become economically appealing, since product sales are always lower than process expenditures. This situation can be explained by the large cost of sorting the plastic waste, which is the most expensive item in the nine scenarios. Sorting costs represent between 25% and 31% of the total expenditure of the process. We note that the sorting process is crucial to separate specific plastic fractions, which sometimes allow for more efficient chemical recycling processes, therefore aligning with the principles of circular economy.

In this case, heat integration resulted in 23 heat exchangers, which allowed us to bring down the annual costs of energy from 188 million euros to 74 million euros (61% reduction). Note that these results include the condensers and the reboilers of the distillation columns. Considering a lifespan of 30 years for the heat exchangers, this one-time purchase accounts for less than 1% of the total energy costs of the process.

In the first four scenarios, where the products obtained are sold to other industries, these contribute with sales worth 135 million euros annually, thus reducing the (gross) process expenditures. Stream D, mostly propylene, adds large benefit to each of these scenarios, covering about 46% of the total sales. Stream E, mainly 1-butene, is responsible for 34% of total sales. This is because both streams are produced in large quantities, in addition to their high value, with a propylene price of 0.89 €/kg and 1-butene of 1.13 €/kg. On the other hand, these scenarios are also characterized by a high process cost, ranging 159-187 million euros annually, since outsourced fuels are used to operate the furnace. In this regard, gasoline emerges as the costliest option (0.08 €/MJ), while natural gas is the cheapest alternative (0.02 €/MJ).

In the other five scenarios, pyrolysis products are reused as fuel in the furnace, reducing sales as only the unused portion of the streams is marketed. In turn, this leads to significant savings, as there is no requirement for outsourced fuels in the furnace. The magnitude of these savings depends on the reference fuel used in the furnace and on the process stream reused as fuel. According to our estimates, reusing methane (stream A) and ethane (C) are the best options among all the scenarios considered. This is not surprising given that methane and ethane have the largest MJ/€, standing at 101 and 186 MJ/€, respectively.

* + 1. *Environmental Assessment: material circularity*

In the initial four scenarios, where all the product streams are sold to other industrial processes and fossil-fuels are used in the furnace, a 100% MCI for the pyrolysis process is achieved. This value can be deemed as an upper bound on the overall material circularity that could not be achieved in practice, since losses will likely occur in other parts of the supply chain (e.g., transformation of monomers back into polymers).

In the other five scenarios, where some product streams are reused as fuel in the pyrolysis furnace, not all the material is recovered, thus decreasing the MCI to approximately 94%. Recall that these were the scenarios achieving the best economic performance, which shows that a trade-off exists between these two metrics. Hence, to substantiate the decision-making process, we plot these metrics in Figure 3.

*Figure 3: Scatter plot depicting the material circularity vs the process expenses for the nine scenarios studied.*

Indeed, we found that using outsourced fuels in the furnace leads to higher circularity, despite not being as cost effective as when product streams replace them. Analysing these results through the lens of multi-objective optimization, only two strong Pareto optimal solutions are found: selling all products and outsourcing natural gas for the furnace (Natural Gas scenario), and reusing methane as fuel in the furnace (Stream A). Hence, methane (the main component of natural gas) at 99% purity, proves to be an excellent substitute for natural gas. It displays 21% lower expenses compared to the other strong Pareto scenario, while maintaining a commendable MCI 94%. The remaining scenarios explored can all be regarded as weak Pareto points (within a certain numerical tolerance), and therefore inferior to the two aforementioned cases.

* 1. Conclusions

This study capitalized on laboratory-scale data of a pyrolysis reaction of waste PP to design an industrial process for its chemical recycling. To this end, we used a simulation-optimization approach, complemented with a heat-exchanger network that allowed to reduce the process costs by 61%. Results from this baseline simulation were used to examine nine scenarios involving different options for the fuel used in the furnace: four scenarios with different outsourced fuels, and five scenarios utilizing the products streams as fuels. We found scenarios to achieve high values for material circularity in the pyrolysis process, ranging from 93%-100%. These values should be understood as upper bounds on the real material circularity that can be achieved, since material losses are expected to occur in other transformation processes necessary to close the circle. Unfortunately, these promising results are not paired by an equally appealing economic performance, since none of the scenarios managed to achieve a positive profit (net expenses ranging from 159 to 187 million euros annually). This demonstrates the need to devote further research efforts to improve this process. Meanwhile, subsidies could help penetrate chemical recycling in the market, as these processes generate valuable chemicals from plastic waste, while achieving significant degrees of material circularity. Our contribution can aid with generating a solution to the accumulation of plastic waste.

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