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Analysis of Infrastructures for Processing Plastic Waste using Pyrolysis-Based Chemical Upcycling Pathways

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ABSTRACT

Modern mechanical recycling infrastructure for plastic is capable of processing only a small subset of waste plastics, reinforcing the need for parallel disposal methods such as landfilling and incineration. Emerging pyrolysis-based chemical technologies can "upcycle" plastic waste into high-value polymer and chemical products and process a broader range of waste plastics. In this work, we study the economic and environmental benefits of deploying an upcycling infrastructure in the continental United States for producing low-density polyethylene (LDPE) and polypropylene (PP) from post-consumer mixed plastic waste. Our analysis aims to determine the market size that the infrastructure can create, the degree of circularity that it can achieve, the prices for waste and derived products it can propagate, and the environmental benefits of diverting plastic waste from landfill and incineration facilities it can produce. We apply a computational framework that integrates techno-economic analysis, life cycle assessment, and value chain optimization. Our results demonstrate that the infrastructure generates an economy of nearly 20 billion USD and positive prices for plastic waste, opening opportunities for compensation to residents who provide plastic waste. Our analysis also indicates that the infrastructure can achieve a plastic-to-plastic degree of circularity of 34% and remains viable under various external factors (including technology efficiencies, capital investment budgets, and polymer market values). Finally, we present significant environmental benefits of upcycling over alternative landfill and incineration waste disposal methods, and comment on ongoing work expanding our modeling methodology to other chemical upcycling pathway case studies, including hydroformylation of specific plastics to chemicals.

Keywords: Polymers, Optimization, Supply Chain, Modelling, Interdisciplinary

INTRODUCTION

With global rates of plastic production reaching upwards of 400 million tonnes per year and growing [1], nations require integrated, reliable plastic waste management systems [2]. Despite reductions in the environmental impacts of plastic production [3], global plastic recycling rates remain well below 10% [4], necessitating the continued use of parallel disposal methods (such as landfilling and incineration) and allowing significant plastics to accumulate in the environment [5].

Most current recycling occurs by well-established mechanical methods, which are designed to accept bottles of polyethylene terephthalate (PET) and high-density polyethylene (HDPE), together making up less than 12% of post-consumer plastic waste [6]. However, these methods also produce degradation and allow these plastics to only be recycled a finite number of times [7]. Mechanical recycling strategies cannot process most other resins or flexible, multilayer plastics (which account for 35% of plastics currently produced [8]). The parallel disposal methods required for these plastics cause significant environmental and social issues.

Chemical upcycling has been proposed as a complement to mechanical systems. These pathways apply a complex sequence of chemical processing steps to decompose plastics into constitutive chemical species that can be further refined into value-added products, such as fuels, chemicals, and plastic precursors. Due to their improved feedstock versatility, chemical upcycling pathways can be more scalable than mechanical recycling pathways. We analyze a pathway which requires pre-sorting and purification of mixed plastic waste from post-consumer recyclable streams, occurring at material recovery facilities (MRFs) and plastic reprocessing facilities (PRFs). Sorted plastic waste is baled and mechanically reduced to flakes before undergoing pyrolysis and steam cracking to produce plastic monomers, which can finally be repolymerized into resins (here, low-density polyethylene and polypropylene) with identical quality as the starting materials and the same original properties as virgin plastics [4]. Thus, chemical upcycling pathways can help to achieve a circular economy for plastic waste.

Many recent analyses have explored the potential contributions of upcycling to broader plastic waste management systems [4, 7, 9, 10, 11] and its economic and environmental performance [12, 13, 14]. However, a significant gap exists in our understanding of the deployment of upcycling technologies at scale. Previous works have developed models to analyze plastic waste supply chains [15, 16] and supply chain optimization frameworks for studying these problems at regional scales [6]. Large supply chain models can help to analyze complex economic behavior of networks of processing facilities and track the transportation of products in the value chain.

In this paper, we summarize the methods and results of our recent full paper on the national-scale modeling of an upcycling pathway for the continental US [17], and present updates in ongoing comparative analysis of other upcycling pathways with different process details and outcomes on the same national scale, including the recently explored hydroformylation process [18].

We overview a computational framework to examine the potential for deploying a national-scale upcycling infrastructure for post-consumer plastic waste in the continental US, and present an optimal upcycling value chain solution (see Figure 1) for techno-economic analysis and lifecycle assessment. The infrastructure is optimized to maximize total profit while minimizing investment, operating, and transportation costs. We show that this infrastructure can be interpreted as a value chain (an economy) under which stakeholders exchange plastic waste and derived products to maximize profit. In our model, upcycling creates high value from the production of virgin polymers from post-consumer waste, producing a total economy (market) size of 19.7 billion USD per year and remaining viable under various changes to market inputs.

Upcycling as a national plastic waste management strategy achieves a 34% plastic-to-plastic degree of circularity for converting residential plastic waste into high quality polymers. The system compares favorably to existing disposal methods for mixed plastic by environmental metrics, including 69-75% reductions in greenhouse gas (GHG) emissions relative to incineration and 86 avoided new landfills over the next 50 years).

In contrast to this form of pyrolysis-based upcycling (a plastic-to-plastic method for mixed plastics), hydroformylation has been envisioned as a plastic-to-chemicals pathway for more specific plastic feedstocks. To date, hydroformylation has only been explored for one post-consumer resin (HDPE) and three virgin resins (HDPE, LDPE, and PP) [18]. Thus, hydroformylation would compete with mechanical recycling for HDPE. We comment that current market prices for the products of hydroformylation may not support the pathway's use relative to prices for mechanically recycled HDPE.

METHODOLOGY

We model the US as a collection of county-level waste-producing centers of population. Our chemical upcycling pathway comprises a series of six processing technologies, including MRFs, PRFs, pyrolysis, steam cracking, LDPE polymerization, and PP polymerization. The value chain contains 16 different products, including inputs (post-consumer recyclables), intermediates (e.g., plastic waste bales, plastic flake, pyrolysis oil, ethylene), and outputs (e.g., LDPE, PP, fuels). Only feedstock recyclables, plastic bales, plastic flake, and pyrolysis oil are modeled as transportable to represent the infeasibility of transporting many intermediates (e.g., ethylene and propylene gas) and simplify analysis of market demand.

Each component of the value chain has techno-economic definitions from Ma et al. [6]. We applied a mixed-integer program (MIP) formulation to our supply chain optimization model, also proposed by Ma et al. [6] to study upcycling in the US Midwest region. The MIP designs a value chain by placing processing facilities in locations that best capture residential waste, maximize profit, and minimize investment, processing, and transportation costs. Furthermore, we apply LCA methodologies to compare GHG emissions of upcycling, landfilling, and incineration. Emission factors for each technology are adapted from Ma et al. [6].

To limit model size and computational complexity for a national case study, we allow transportation routes between adjacent county centers. Still, the national scale presents challenges for current solvers, so we report close-to optimal solutions with optimality gaps demonstrating their measured distance from the estimated upper bound on total profit.

The model comprises 3,108 county centers and 4,010 sites available for technology placement, with two sizes of technology possible at each potential site. In total, the MIP problem contains 796,756 continuous variables, 8,020 binary variables, and 587,405 constraints.

To implement and run the MIP formulation, we utilize the algebraic modeling package JuMP [17] for the Julia programming language and solve using Gurobi 9.1.0. [20]. We use the MIP to design the physical arrangement

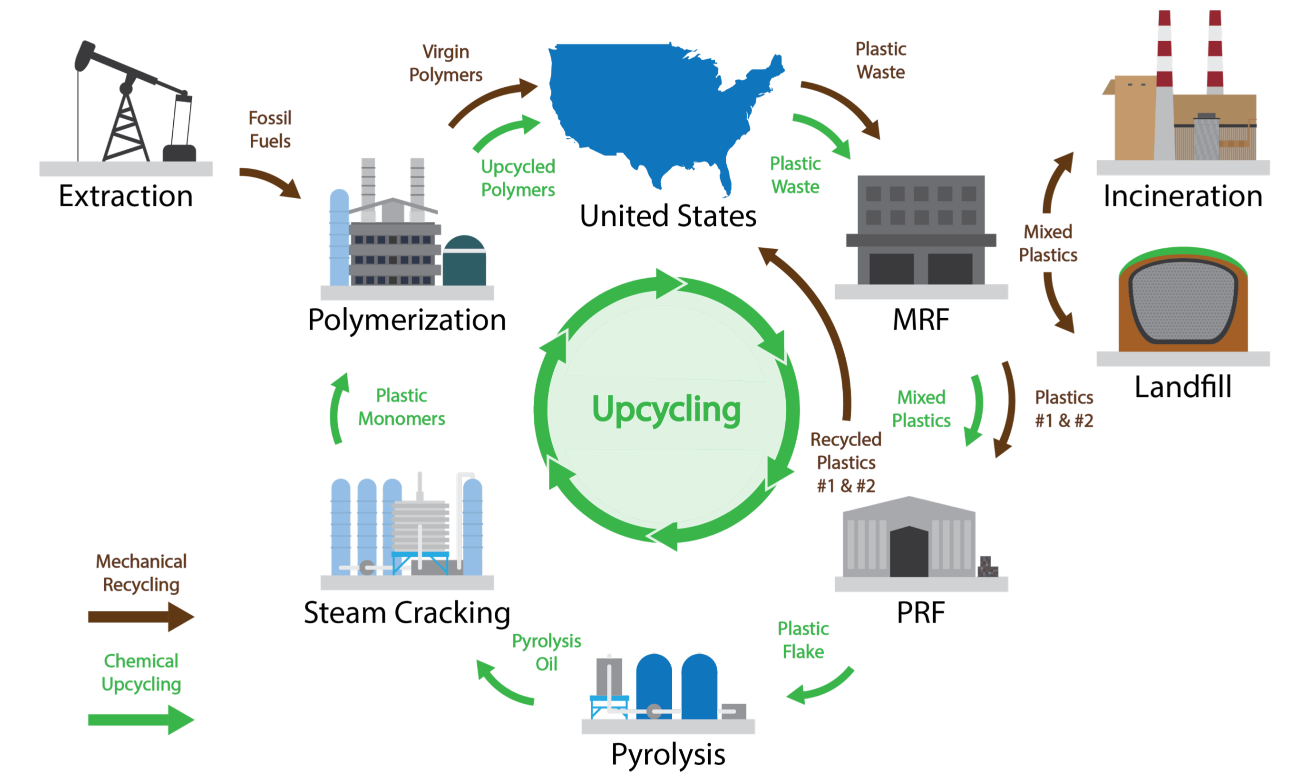

Figure 1: Overview of a pyrolysis-based chemical upcycling value chain. In chemical upcycling, waste plastics are recovered as fuels, chemicals, and virgin-quality polymers in a sequence of processing steps. Our case study examines a combination of mechanical recycling for PET (plastic type #1) and HDPE (plastic type #2), and chemical upcycling of mixed plastics (plastic types #3 - #7). This combined network creates an economy worth nearly 20 billion USD annually and processes 100% of US residential plastic waste, achieving a degree of circularity of 33.9% (more than one-third of input waste plastics are recovered as mechanically or chemically recycled plastics) while most other remaining inputs are converted into valuable byproduct fuels and chemicals.

of the upcycling value chain and then perform economic and environmental analysis using the **CoordinatedSupplyChains.jl** package (**CSC.jl**) developed by Tominac and Zavala [21] and modified for this work with visualization capabilities. After 180 hours of computational time, the MIP was solved to an optimality gap of 4.66%, meaning that the identified system design provides an economic result within 5% of the optimal solution. The optimal design reaches all 3,108 counties and captures 100% of residential plastic waste; in other words, the model indicates that there are sufficient economic incentives to harness all the post-consumer waste to obtain virgin plastics.

RESULTS

Circularity & Economic Evaluation

The final upcycling network captures 100% of residential waste, recovering 11.6% of input waste plastics (the fraction of PET and HDPE bottles) as mechanically-recycled HDPE and PET through MRFs and PRFs, and 22.3% as LDPE and PP through pyrolysis, steam cracking, and repolymerization. This leads to a total plastic-to-plastic degree of circularity of 33.9%, but overall, 94.6% of residential plastics are recovered as valuable products (including significant fuel and chemical byproducts of upcycling which enhance upcycling's economic viability).

Prices for plastic waste are positive for all county centers, demonstrating that the upcycling network is economically viable without added incentives through policy, largely due to the production of high-value LDPE and PP through chemical upcycling. Upcycling creates an economy worth 19.7 billion USD annually (a measure of total economic surplus, excluding capital costs) and nearly 10 billion in residential profit, which could be applied to compensate residents for recycling properly (or donating their waste plastics to upcycling systems) at a rate of 31 USD / year.

Sensitivity Analysis

External market factors and technical specifications impact the economic performance of the upcycling network. Total waste capture remains above 99% for resident offering costs up to 40 USD / tonne (rather than donating plastic waste for free). Improving the efficiencies of key technologies (such as pyrolysis and steam cracking) increases economic surplus but can exhaust the processing capacity of the network faster, excluding some residential waste providers from the market. Increasing transport costs for feedstocks and intermediates has a minimal impact on process economics (transport costs can be doubled and the system processes 99.47% of residential waste plastic). The network can withstand 20% reductions in LDPE and PP market value (major economic drivers of viability) without significant decreases in waste capture. Finally, limiting the overall capital cost investment available for constructing the network limits its extent of waste capture, prioritizing regions of higher waste production density excluding regions including the Great Plains and Northwest.

Comparison to Existing Processing Methods

The upcycling network emits 1.387 tonne CO2 eq / tonne of plastic, including processing and transportation contributions. Landfills produce limited direct GHG emissions (0.0236 tonne $CO_{2\ eq}$ / tonne of plastic), but impose other categories of environmental impacts including land use, leakage and microplastic pollution. Incineration or waste-to-energy (WtE) systems produce 2.3-3.1 tonnes CO2 eq of direct emissions per tonne of plastic combusted depending on resin type.

However, upcycling and WtE fulfill additional functions not achieved by landfills: upcycling produces virgin-quality polymers and WtE can produce electricity. Applying LCA methodology to credit upcycling for its avoided fossil fuel use and WtE for its avoided grid electricity consumption, we determine revised emissions factors of 0.508 tonne CO2 eq / tonne plastic (including transport) for upcycling and 1.6-2.0 tonnes CO2 eq per tonne of plastic combusted for WtE. With credits, upcycling emissions compare more favorably to WtE, whose credits based on grid electricity will gradually disappear as the grid becomes decarbonized.

Comparison to Hydroformylation

While we model a plastic-to-plastic upcycling pathway with fuel and chemical byproducts, other pathways with other aims also attract the interest of researchers. In particular, Li et al. [18] studied hydroformylation (another pyrolysis-based pathway) to produce aromatics, paraffin, alcohols, and dialcohols from post-consumer HDPE or virgin HDPE, LDPE, and PP. The inputs of this pathway also determine its applications: as developed in this publication for a HDPE waste feedstock, hydroformylation would not be an alternative to pyrolysis-based upcycling in this work and Ma et al. [6] but to mechanical recycling. However, since the pathway is still based upon pyrolysis, it could be applied to mixed plastic wastes in the future.

Li et al. [18] report yields and market values of the four chemical categories produced by hydroformylation of waste HDPE, which we apply as preliminary target yields for the hydroformylation of mixed plastic wastes. We determine that hydroformylation provides a high combined revenue for pyrolysis oil (2154 USD / tonne) from which additional operating costs (644 USD / tonne) detract significantly, leading to an adjusted pyrolysis oil value of 1509 USD / tonne and a corresponding post-MRF-and-PRF plastic waste value of 1035 USD / tonne. This is slightly above the market value prices for recycled HDPE produced at MRFs and PRF used in Ma et al. [6] and this work (991 USD / tonne), but is significantly above the market value produced for post-MRF-and-PRF mixed plastic wastes from plastic-to-plastic upcycling (711 USD / tonne).

We extend our model to consider a hydroformylation pathway and map its optimal infrastructure arrangement and product transportation flows in Figure 2. The comparatively higher value for mixed plastic waste supported by hydroformylation allows plastic waste to sustain greater transportation distances and costs, and thus, the resulting arrangement possesses fewer decentralized components. For example, the plastic-to-plastic upcycling solution placed 336 mostly small-scale MRFs (the first step in each pathway) whereas the hydroformylation solution placed only 38 mostly large-scale MRFs. The total MRF capacity of the hydroformylation solution was nearly 18% lower than that of plastic-to-plastic upcycling, but the utilization of available MRF capacity was much greater (98.8% compared to 81.2%). In contrast, the number of hydroformylation hubs in this solution is roughly comparable to the number of steam cracking and polymerization combined hubs in the plastic-to-plastic upcycling solution.

A more centralized network entails trade-offs with supply, as in the hydroformylation solution, only 99.36% of suppliers (3088 counties out of 3108) participate in the network, compare to 100% in the plastic-to-plastic upcycling solution. Future work will aim to refine this solution to achieve 100% waste capture. For the residents that participate in the hydroformylation network, a potential compensation rate of 30 USD / year is generated through the value chain, which is just below that of plastic-to-plastic upcycling.

Hydroformylation achieves different production objectives than plastic-to-plastic upcycling, and with different environmental performance. The uncredited emissions of the hydroformylation network reach 1.473 tonne $CO_{2\ eq}$ / tonne of plastic processed, which is greater than that of plastic-to-plastic upcycling. We note that this analysis does not consider that hydroformylation products would otherwise be produced by expensive fossil-based pathways. Furthermore, hydroformylation produces only chemicals, so its plastic-to-plastic degree of circularity is 0%.

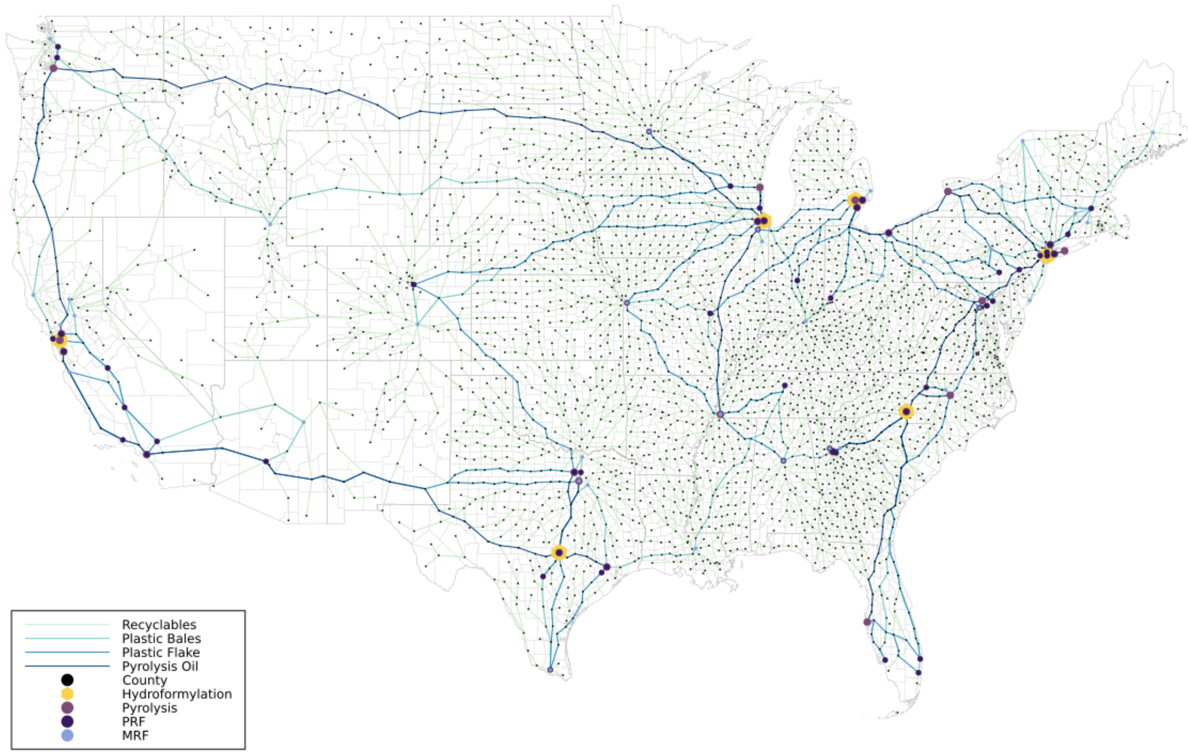

Figure 2: Optimal infrastructure placement and product transportation patterns for a hydroformylation alternative to plastic-to-plastic upcycling.

DISCUSSION & IMPLICATIONS

Previous research has defined the individual steps of upcycling and modeled their implementation at smaller scales. Our analysis reveals that upcycling remains viable when expanded to the continental US, contributing to a circular economy for plastics driven by the high value of upcycling's end products. Upcycling also presents environmental impact reductions in land use relative to landfills and GHG emissions relative to WtE plants.

As demonstrated in this work, system-level modeling projects have the potential to illuminate not only economic and environmental behaviors of potential value chains, but also comparative results between processing options. Our future work will continue expanding our case studies in hydroformylation and other proposed chemical upcycling pathways, while also incorporating ways to relax some of the major simplifying assumptions in our model structure. We are also interested in more directly modeling policy interventions (including plastic waste source reduction or incentives for upcycling technologies) to better represent the role of policy in national scale systems and the considerations of current US waste management, recycling, and plastic pollution prevention strategies.

DIGITAL SUPPLEMENTARY MATERIAL

The CSC.jl analysis package is publicly available at https://juliahub.com/ui/Packages/CoordinatedSupply-Chains. All scripts and data needed to reproduce the results are available at https://github.com/zavalab/JuliaBox/tree/master/National_Plastic.

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