

Technical, Economic, and Environmental Comparison of Closed-Loop Recycling Technologies for Common Plastics

Taylor Uekert,* Avantika Singh, Jason S. DesVeaux, Tapajyoti Ghosh, Arpit Bhatt, Geetanjali Yadav, Shaik Afzal, Julien Walzberg, Katrina M. Knauer, Scott R. Nicholson, Gregg T. Beckham, and Alberta C. Carpenter



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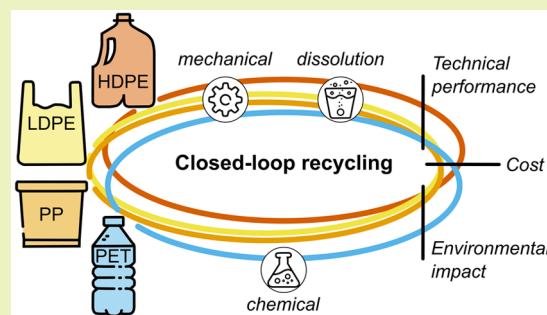
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ABSTRACT: Over 400 million metric tons of plastic waste are generated globally each year, resulting in pollution and lost resources. Recycling strategies can recapture this wasted material, but there is a lack of quantitative and transparent data on the capabilities and impacts of these processes. Here, we develop a data set of material quality, material retention, circularity, contamination tolerance, minimum selling price, greenhouse gas emissions, energy use, land use, toxicity, waste generation, and water use metrics for closed-loop polymer recycling technologies, including mechanical recycling and solvent-based dissolution of polyethylene, polyethylene terephthalate (PET), and polypropylene, as well as enzymatic hydrolysis, glycolysis, and vapor methanolysis of PET. Mechanical recycling and PET glycolysis display the best economic (9%–73% lower than competing technologies) and environmental (7%–88% lower) performances, while dissolution, enzymatic hydrolysis, and methanolysis provide the best recyclate material qualities (2%–27% higher). We identify electricity, steam, and organic solvents as top process contributors to these metrics and apply sensitivity and multicriteria decision analyses to highlight key future research areas. The estimates derived in this work provide a quantitative baseline for comparing and improving recycling technologies, can help reclaimers identify optimal end-of-life routes for given waste streams, and serve as a framework for assessing future innovations.

KEYWORDS: plastic, recycling, circular economy, life cycle assessment, techno-economic analysis



INTRODUCTION

With over 90% of all polymer products landfilled, incinerated, or leaked into the environment since 1950, plastic has become a symbol for the linear “take–make–waste” economy.¹ Innovation within the recycling space is essential to reach targets set by the U.S. Plastics Pact and European Union to achieve 50% plastic packaging recycling or composting by 2025.^{2,3} Several closed-loop (plastic-to-plastic) and open-loop (plastic-to-x) recycling techniques are entering this landscape and can be classified as physical or chemical. Physical recycling preserves the molecular structure of the polymer and includes mechanical processing and solvent-based dissolution. Chemical recycling deconstructs plastic into molecular intermediates. Examples include depolymerization of heteroatom-containing polymers into their constituent monomers or oligomers by glycolysis, methanolysis, or hydrolysis, as well as high-temperature and high-pressure techniques such as pyrolysis and gasification for converting plastic into fuels or chemical feedstocks.⁴

Given the breadth of plastic recycling research, it is challenging to determine how to implement these technologies in an economically and environmentally beneficial manner that improves polymer circularity. Analysis can help bridge this

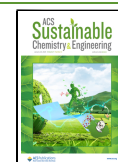
gap.^{5,6} Techno-economic analysis (TEA) and life cycle assessment (LCA)—which determine the costs and environmental impacts of a product—have been used to estimate the feasibilities of various plastic recycling techniques.^{7–17} However, the different assumptions and background data used in these studies prevent a fair assessment of plastic recycling technologies across multiple technical, economic, and environmental metrics.

The current study uses a rigorous modeling framework to quantitatively compare current and next-generation plastic recycling technologies. We focus on closed-loop recycling of the most widely consumed polymers, including mechanical recycling and solvent-based dissolution of high- and low-density polyethylene (HDPE and LDPE), polyethylene terephthalate (PET), and polypropylene (PP), as well as

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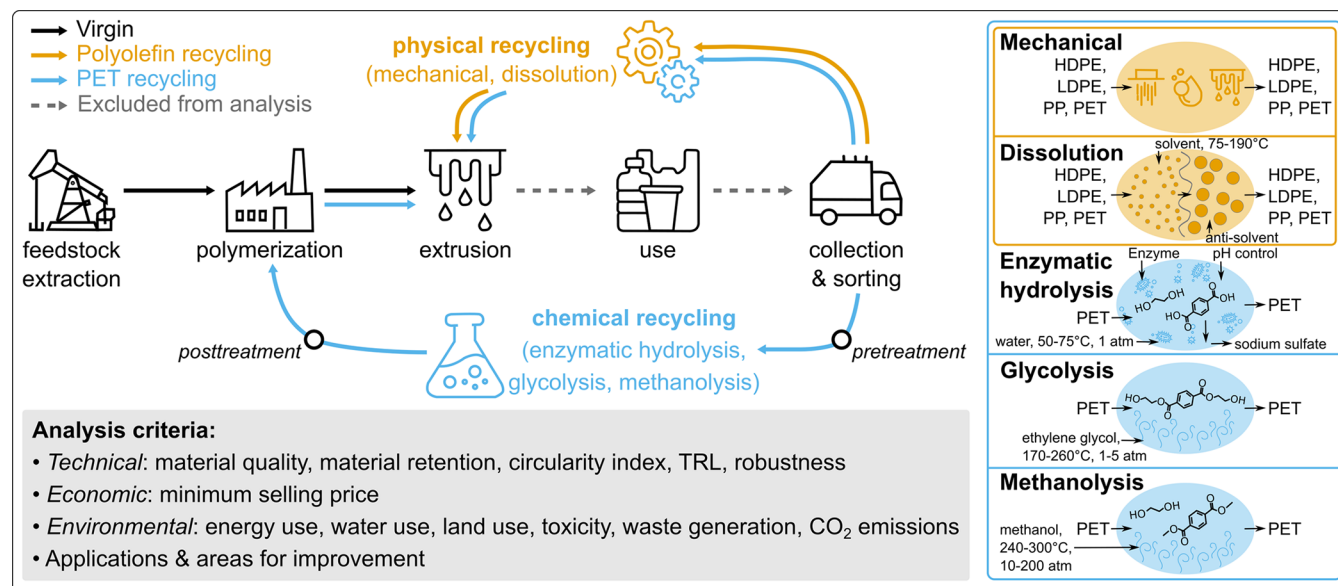


Figure 1. Overview of the analysis scope. A cradle-to-gate approach is used to assess the recycling technologies from the materials recovery facility plastic bale through to recycling, repolymerization (if applicable), and extrusion to pellets; production, use, and collection of the original plastic are not included. Virgin polymers are assessed for fossil feedstock extraction, monomer synthesis, polymerization, and extrusion; use and disposal are not included.

Table 1. List of Assessed Technical Metrics and Their Descriptions

Metric	Definition	Unit
Material quality	Melt flow rate (MFR) of the recycled plastic divided by MFR of the corresponding virgin material ⁴	MFR/MFR
Material retention	Quantity of recycled material obtained per quantity of incoming postconsumer plastic bale	kg/kg
MCI	Circularity of recycled material, see SI for details ²³	unitless
Robustness A	Quantity of other plastic contaminants that the recycling process can tolerate per quantity of incoming postconsumer plastic bale	kg/kg
Robustness B	Quantity of other material contaminants (e.g., biomass, metals) that the recycling process can tolerate per quantity of incoming postconsumer plastic bale	kg/kg
TRL	Estimate of maturity of recycling technologies, on a 1–9 scale	unitless

⁴MFR is one of the most common criteria for characterizing polymer quality and stability and is standardized as the weight of polymer extruded in 10 min through a tubular die of a specified diameter at a constant temperature and applied pressure.²⁴

glycolysis, methanolysis, and enzymatic hydrolysis of PET (Figure 1). Using a combination of literature review, Aspen Plus modeling, the Materials Flows through Industry (MFI) tool,¹⁸ and process-based LCA,¹⁹ we assess the technologies across technical (material quality, material retention, circularity index, contamination tolerance, and technology readiness level [TRL]), environmental (greenhouse gas [GHG] emissions, energy use, land use, toxicity, waste generation, and water use), and economic (minimum selling price [MSP]) metrics for a functional unit of 1 kg of recyclate. Top process contributors to the metrics are identified and used to define sensitivity cases and multicriteria decision analysis (MCDA), as well as future research directions that can improve the viability of the technologies. This work provides a baseline for the current and upcoming plastic recycling landscape and establishes a characterization methodology for assessing emerging recycling technologies.

METHODS

Scope. Mechanical recycling and dissolution of polyolefins and polyesters, as well as enzymatic hydrolysis, glycolysis, and methanolysis of PET, are currently highly studied closed-loop plastic recycling technologies (Figure S1). Mechanical recycling involves sorting, shredding, washing, and extrusion of postconsumer plastic. Dissolution is a physical process in which a polymer is dissolved in a

solvent and precipitated and purified by addition of an antisolvent. Enzymatic hydrolysis uses hydrolase enzymes to cleave the ester bonds in PET and generate the monomers ethylene glycol (EG) and terephthalic acid (TPA). Glycolysis uses EG to depolymerize PET into the oligomer bis(2-hydroxyethyl) terephthalate (BHET), while methanolysis uses vaporized methanol to deconstruct PET into dimethyl terephthalate (DMT) and EG. Additional closed-loop PET recycling methods such as acidic or basic hydrolysis, saponification, acetolysis, and aminolysis were considered less viable and are not included.⁴ Open-loop polyolefin recycling by pyrolysis, gasification, or other chemo-catalytic techniques are also beyond the closed-loop study scope. We consider HDPE (C₂H₄)_n, LDPE (C₂H₄)_n, PP (C₃H₆)_n and PET (C₁₀H₈O₄)_n packaging materials, which are the most commonly used plastics and have existing postconsumer supply chains.^{1,20} The recycling rates of these polymers were 15%, 10%, 2%, and 3% for PET, HDPE, LDPE, and PP, respectively, in 2019 in the United States (Figure S2).²¹

Technology Models. Process models for each technology were constructed in Aspen Plus V10 on a 150 t per day (TPD) basis. The average materials recovery facility (MRF) in the United States processes ~220 TPD, facilitating consistent access to postconsumer plastic bales for recycling facilities of the modeled size.²² Mechanical recycling, dissolution, and methanolysis models were built *de novo*, and the glycolysis and enzymatic hydrolysis models were adapted from our previous work to maintain consistent system boundaries.^{9,14} All models include applicable pretreatment (e.g., shredding of the postconsumer plastic bale), recycling steps, and applicable posttreat-

Table 2. List of Assessed Environmental Metrics and Their Definitions

Metric	Definition	Method	Unit
E-factor	Waste generation, including solid waste, gaseous or liquid byproducts, and wastewater	Literature review	kg/kg
Energy use	Supply chain energy used	MFI ^a	MJ/kg
GHG emissions	Supply chain GHG emissions produced	MFI ^a	kg CO ₂ eq/kg
Land use	Relative species loss caused by land use	ReCiPe ^b	m ² a crop eq/kg ^c
Toxicity	Noncarcinogenic and carcinogenic toxicological effects on humans	TRACI 2.1 ^b	CTUh/kg ^d
Water use	Potential for water depletion over one year	AWARE ^b	m ³ /kg

^aMFI uses United States-specific background data that provides a more accurate representation of supply chain impacts within the current market.¹⁸ MFI yields slightly different GHG emission results in comparison to traditional LCA methods due to this alternative background data (Figure S10). ^bUsing SimaPro LCA software and ecoinvent v3.3 background data (allocation, cutoff by classification—unit, United States-specific inventories when available, global inventories otherwise). ^cm²a crop eq = square meters of annual crop equivalent. ^dCTUh = comparative toxic unit for humans.

ment (e.g., repolymerization). These pre- and postrecycling steps are crucial for implementation, but they are often neglected in analyses of chemical recycling. Further details, process flow diagrams, and process inventories for all models are available in the Supporting Information (SI) Text, Figures S3–S7, and Tables S1–S13. A visual representation of the data sources and analysis methods used in this work is available in Figure S8.

Technical Analysis. Technical metrics were curated from the literature, as defined in Table 1, and are reported as averages \pm standard deviation (σ) (see Tables S14–S19 for individual values).

Economic Analysis. Material and energy balances from the process models were used to estimate raw material consumption, utilities, other variable operating expenses, equipment sizing, and capital investment. Pricing of consumables and postconsumer plastic bales was obtained from industry databases (2016–2020 average).^{25,26} Postconsumer HDPE bale pricing is the average of natural, colored, and rigid bales, while that of postconsumer LDPE is the average of grades A and B bale qualities (Figure S9). Pretreatment shredding costs are directly incorporated (\$0.13/kg), reflecting a scenario in which bale processing to flake is performed in house or in close partnership with an external facility. This differs from our previous work,^{9,14} which added a literature-derived conversion factor of \$0.42/kg²⁷ to the postconsumer PET bale price. A discounted cash flow analysis approach with financial parameters listed in Table S20 and an assumed plant lifetime of 30 years was applied to estimate the MSPs of plastic obtained from each recycling process.

Environmental Analysis. The assessed environmental impact categories are listed in Table 2. These metrics were selected to address parameters expected to be crucial for mitigating climate change and its related water and land shortages, as well as characterizing emerging recycling methods that are reliant on organic solvents and energy-intensive purification steps.

For all recycling technologies, a cutoff approach was used in which the postconsumer plastic is assumed to be free of environmental burdens. Any generated waste was handled by 80% landfilling and 20% incineration, according to the ratio for mixed plastic waste disposal reported by the United States Environmental Protection Agency in 2018.²⁸ Impacts associated with infrastructure were estimated using ecoinvent entries for “Plastic processing factory” (for mechanical recycling and flake production) or “Chemical factory, organics” (for all other technologies).

Uncertainty Analysis. Uncertainty was estimated using a semiquantitative pedigree approach.²⁹ Each item in the life cycle inventory was given reliability, completeness, temporal correlation, geographical correlation, and further technological correlation scores (scale of 1–5) according to the rubric in Table S21. The sum of these scores was assigned a variability between $\pm 5\%$ and $\pm 25\%$ (Table S22), which was set to encompass known σ around process yields (Tables S14–S19). These variability ranges determined the low and high values of symmetric triangular distributions for each inventory item. Triangular distributions were selected because there was insufficient data to determine a more exact probability distribution function. For environmental metrics assessed in SimaPro, the uncertainties of background processes were provided by log-normal

distributions in ecoinvent. Pricing uncertainty was assigned a triangular distribution based on average, lowest, and highest annual costs between 2016 and 2020. With these distributions, a Monte Carlo analysis was performed with 1000 iterations, giving mean and σ values. Statistically significant results are highlighted in the text. Uncertainty analysis was not performed on energy use and GHG emissions as this functionality is under development in MFI.

Error propagation for summing or multiplying values was calculated with eq 1 or eq 2.

$$\delta Q = \sqrt{(\delta a)^2 + (\delta b)^2 + \dots + (\delta z)^2} \quad (1)$$

$$\frac{\delta Q}{|Q|} = \sqrt{\left(\frac{\delta a}{a}\right)^2 + \left(\frac{\delta b}{b}\right)^2 + \dots + \left(\frac{\delta z}{z}\right)^2} \quad (2)$$

where a , b , and z are the input values, Q is their sum or multiplied/divided value, and δa , δb , δz , and δQ are their corresponding standard deviations.

Sensitivity Analysis. Sensitivity analysis was conducted to determine how the alteration of key variables impacts the MSP, GHG emissions, energy use, land use, toxicity, and water use of the recycling technologies (see Table S23 for all variables, values, and their justifications).

MCDA. The analytical hierarchy process method³⁰ was used to rank recycling technologies across several criteria. Criteria and subcriteria included technical (material quality), economic (MSP), resource consumption (energy use, water use, land use, material retention, and MCI), and environmental impact (GHG emissions, toxicity, and E-factor). Robustness A and Robustness B were not included as the virgin materials cannot be assessed by these metrics. TRL was not considered because a low or high value does not necessarily correlate with better or worse performance.

Each subcriteria metric was normalized on a 0–1 scale (\bar{x}) by dividing the value for a given technology (x_n) by the maximum value (x_{\max} ; see Tables S24–S25), where 0 is defined as the “worst” and 1 as the “best”. For metrics where a higher value is preferential (e.g., high material quality), normalization was completed with eq 3. For metrics where a lower value is preferential (e.g., low GHG emissions), normalization was calculated with eq 4.

$$\bar{x} = \frac{x_n}{x_{\max}} \quad (3)$$

$$\bar{x} = 1 - \frac{x_n}{x_{\max}} \quad (4)$$

The subcriteria were summed into their respective criteria categories, each of which was assigned a weighting. Three scenarios with different weightings were evaluated (Table S26). For “equal” weighting, the technical, economic, and resource consumptions and environmental impact criteria scores were each set to one for a maximum (best) overall score of four. The “economic and technical” weighting set economics to 50% of the overall score of four, technical to 40%, and both environmental and resources to 5% each. For the

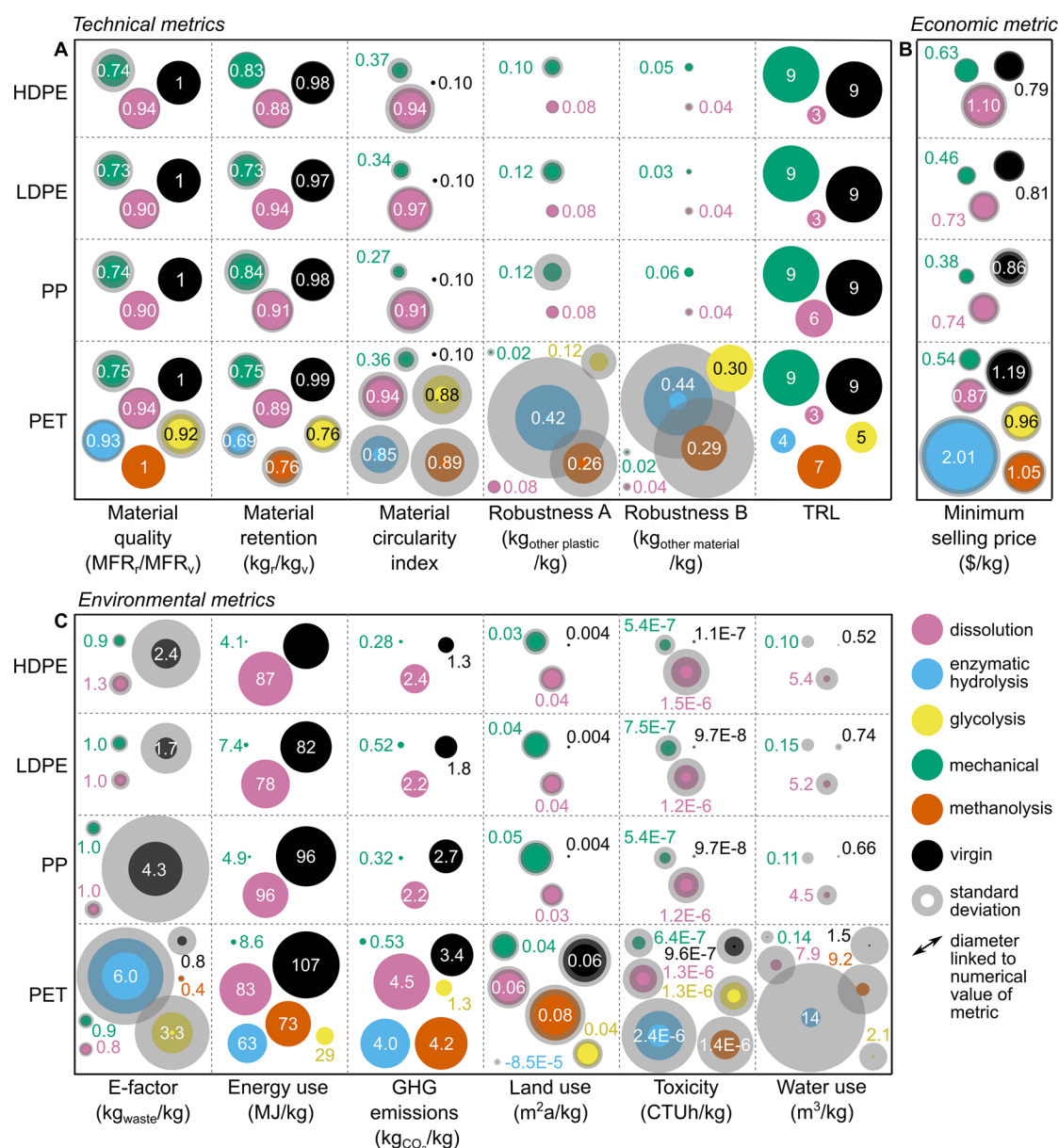


Figure 2. (A) Technical, (B) economic, and (C) environmental metric results for recycling of HDPE, LDPE, and PP by mechanical and dissolution technologies, as well as recycling of PET by mechanical, dissolution, enzymatic hydrolysis, glycolysis, and vapor methanolysis techniques. Metrics are compared to results for the corresponding virgin polymers when applicable. The sizes of the bubbles and gray circles are linked to the numerical values of the metric averages and standard deviations; there is no additional significance to overlapping bubbles. The MFI tool does not have uncertainty capabilities, and thus, standard deviations for energy use and GHG emissions are not shown. Raw data are available in Table S27.

“environment and resources” weighting, the environmental score was set to 50%, resources to 40%, and economic and technical to 5% each.

RESULTS AND DISCUSSION

Technical, Economic, and Environmental Results.

Figure 2 shows the technical capabilities, MSPs, and environmental impacts of HDPE, LDPE, PET, and PP recycling, as well as their manufacture from virgin feedstocks (Table S27).

When considering technical metrics, material quality is lowest for mechanical recycling from 0.73 ± 0.19 to 0.75 ± 0.20 (Figure 2A, Tables S14–S19, S27). This alteration in MFR is linked to the extrusion process, where the combination of heat and shearing cause thermo-oxidative chain scission, chain branching, or cross-linking.³¹ Chain extenders or other

additives are needed to maintain material qualities suitable for plastic reuse, which would add costs and environmental impacts to our model.³¹ Material quality for polymers recycled by dissolution varies from 0.90 ± 0.09 to 0.94 ± 0.06 due to thermal-induced chain scission, which is less extensive than in mechanical recycling because of lower temperatures and lack of physical shearing.³² Chemical recycling has material qualities of 0.92 ± 0.11 for glycolysis, 0.93 ± 0.05 for enzymatic hydrolysis, and 1.0 (σ unavailable) for methanolysis. Based on MFR guidelines for plastic products,²⁴ we estimate that a recycled polymer must have a material quality of at least 0.75 to be processed back into the same type of bottle, injection-molded item, or fiber, or at least 0.8 for film. Thus, mechanically recycled plastic will likely be reused only once before being downgraded to lower quality products (without

considering blending with virgin resin), whereas dissolution or chemical processes should enable at least three recovery cycles (Figure S11). Chemical processes are often cited as enabling infinite plastic recyclability,^{8,11} but this analysis shows that there is uncertainty around these claims and highlights the need to consistently characterize material quality. Nevertheless, chemical recycling can generate food-grade polymers, which must contain total residual contaminants (volatile or non-volatile polar or nonpolar molecules, or heavy metals) of less than 220 $\mu\text{g}/\text{kg}$ for PET or 300 $\mu\text{g}/\text{kg}$ for polyolefins.³³ Mechanical recycling and dissolution require additional processing steps such as advanced sorting techniques, incorporation of antioxidants or processing aids, or complete solvent removal to reach this requirement.^{31,33}

Material retention considers plastic losses from feedstock sorting and pretreatment, as well as process yields. Most mechanical recycling losses (retention of 0.73 \pm 0.18 to 0.84 \pm 0.10) occur in the sorting steps, as reclaimers tend to oversort to obtain the pure polymer streams required for maximum material quality.³¹ Dissolution has a high material retention (between 0.88 \pm 0.07 and 0.94 \pm 0.01) due to less-extensive sorting requirements and efficient solvent–polymer separation. In contrast, over 50% of losses from chemical recycling are attributed to the depolymerization and monomer recovery phases, resulting in retentions of 0.69 \pm 0.15, 0.76 \pm 0.13, and 0.76 \pm 0.19 for enzymatic hydrolysis, glycolysis, and methanolysis, respectively. Material retention and quality are linked to circularity, as defined by the Ellen MacArthur Foundation's MCI.²³ Virgin polymers, which are assumed to flow linearly, have MCIs of 0.1. Mechanical recycling has the statistically lowest MCI (0.27 \pm 0.07 to 0.37 \pm 0.10) because the resulting material is unlikely to undergo another recovery cycle due to its low quality. Dissolution and chemical recycling offer MCIs > 0.8.

Robustness A and Robustness B increase alongside polymer selectivity. The robustness of mechanical and dissolution recycling is low (0.02 \pm 0.03 to 0.12 \pm 0.12), but these technologies are applicable to a wide range of polymers. While chemical recycling allows for higher contamination levels (low of 0.12 \pm 0.11 for glycolysis [Robustness A] and high of 0.44 \pm 0.31 for enzymatic hydrolysis [Robustness B]), it is selective to PET, polylactic acid, and in some cases polyurethane. Dissolution and chemical recycling can remove colorants and odors from the incoming material,^{11,34,35} although glycolysis generates a discolored recycle due to the utilized catalysts.³⁶ Many of these technologies are still at laboratory- or pilot-scale TRLs and will likely exhibit improved technical performance over time. Data on contamination tolerance and material quality are limited (1–7 sources were found per technology, Tables S14–S19), and consistent characterization of these properties by experimental researchers would be beneficial.

From an economic perspective, our process models for mechanical recycling, dissolution (except HDPE), PET glycolysis, and PET vapor methanolysis exhibit MSPs statistically lower than virgin manufacturing (Figure 2B, Table S27). Similarly, mechanically recycled plastics in the United States have actual selling prices equivalent to or lower than those of their virgin equivalents (2016–2020, Figure S12). Both the actual selling prices and estimated MSPs will depend on market dynamics, including supply and demand of recycle and changes in virgin polymer pricing, as well as legislation around recycled content requirements. The MSPs of mechanical recycling (0.38 \pm 0.04 to 0.63 \pm 0.03 $\$/\text{kg}$),

dissolution (0.73 \pm 0.07 to 1.10 \pm 0.09 $\$/\text{kg}$), and methanolysis (1.05 \pm 0.07 $\$/\text{kg}$) also fall within a similar range to previous reports (\pm 5%–30%, depending on feedstock costs).^{10,15,37,38} Mechanical recycling, with its lower operational and capital costs (Table S28), economically outcompetes all other options on a statistically significant basis.

When assessed for environmental impact, current processes outperform next-generation technologies (Figure 2C, Table S27). Mechanical recycling offers energy use (4.1–8.6 MJ/kg) and GHG emissions (0.28–0.53 kg CO₂/kg) an order of magnitude lower than the other recycling technologies for all plastics, as well as low E-factors (0.85 \pm 0.30 to 1.0 \pm 0.38 kg/kg), land use (0.032 \pm 0.012 to 0.049 \pm 0.010 m²a/kg), toxicity (5.4 \times 10⁻⁷ \pm 5.5 \times 10⁻⁷ to 7.5 \times 10⁻⁷ \pm 5.0 \times 10⁻⁷ CTUh/kg), and water use (0.099 \pm 9.59 to 0.153 \pm 18.3 m³/kg). Reports have similarly shown that mechanical recycling has lower GHG emissions than chemical recycling, landfilling, or incineration.^{15,39} Glycolysis of PET, which has fewer depolymerization and product recovery steps than enzymatic hydrolysis or vapor methanolysis, outperforms its chemical recycling competitors by 1.1–3.2 times for GHG emissions, energy use, and toxicity. The GHG emissions reported here for enzymatic hydrolysis differ slightly from our previous work, which used a system expansion approach to normalize impacts across the multiple potential lifetimes of the recycle.¹⁴ Most recycling strategies consume less energy than their virgin equivalents, which is due to the reduction of fossil feedstock requirements. The E-factor is statistically equivalent for virgin polymers and most recycling processes (except enzymatic hydrolysis, which generates 5.5 \pm 2.0 kg of wastewater). Toxicity is statistically higher for recycling than virgin polyolefin production; this metric is linked to disposal of plastic losses from the recycling process and thus is more comparable to values for plastic waste landfilling (7.37 \times 10⁻⁷ CTUh/kg). Water use is high for recycling processes that require extensive heating or cooling (dissolution, glycolysis, methanolysis, and enzymatic hydrolysis), although the standard deviations are high due to water availability uncertainty.⁴⁰

For comparison, we assessed gasification and pyrolysis on a closed-loop basis. These chemical recycling technologies—which use high temperatures and pressures to convert plastics into synthesis gas or synthetic crude oil—are typically considered open loop, but they can become closed loop if the produced chemicals are upgraded into plastic monomers. The economic and environmental metrics of pyrolysis and gasification are currently 10–100 times higher than virgin polymers due to low yields of monomers suitable for repolymerization (material retentions of 0.010 \pm 0.003 to 0.14 \pm 0.07) and high energy requirements for the conversion and subsequent upgrading processes (Figure S13, Table S29). Previous reports have shown that pyrolysis and gasification can be economically and environmentally competitive for the production of feedstock fuels and chemicals (e.g., benzene–toluene–xylene mixture, naphtha, or methanol) rather than plastics,^{16,17,41} although this may change with technology and policy advancements.

This analysis did not include postconsumer plastic collection and sorting at a MRF, which were assumed to be identical for all recycling technologies. Adding these processes would increase energy use, GHG emissions, land use, toxicity, and water use per kilogram of incoming postconsumer plastic by 2.98 MJ, 0.179 kg CO₂ eq, 0.0024 m²a, 1.79 \times 10⁻⁷ CTUh, and 0.0237 m³ respectively (Table S30), as calculated from

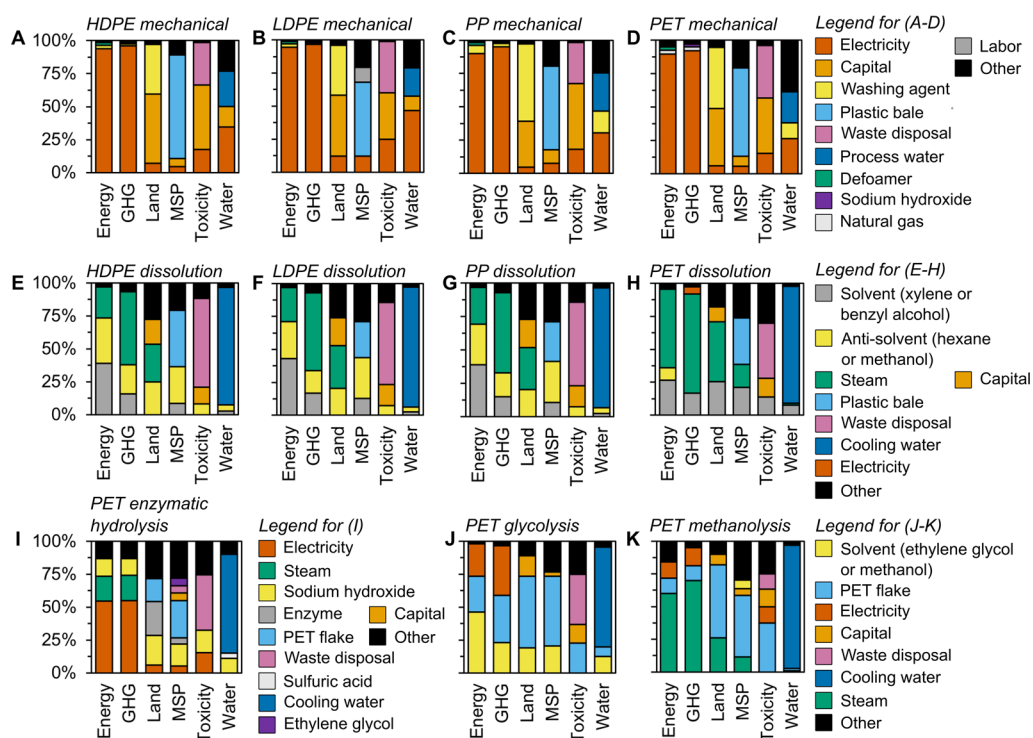


Figure 3. Top process contributors to energy use, GHG emissions, land use, MSP, toxicity, and water use for mechanical recycling of (A) HDPE, (B) LDPE, (C) PP, and (D) PET; dissolution of (E) HDPE, (F) LDPE, (G) PP, and (H) PET; (I) enzymatic hydrolysis of PET; (J) glycolysis of PET; and (K) methanolysis of PET. Raw data are available in [Tables S32–S34](#).

inventories in industrial reports ([Table S31](#)).^{42,43} Collection and sorting will impact material retention as well given that a mere 30% of PET is collected in the United States²⁷ and at least 10% of incoming plastic is estimated to be lost at MRFs.⁴² While these systems are beyond our current scope, postconsumer plastic collection will be explored in future work.⁴⁴ Due to data limitations, this analysis also did not consider social metrics such as employment.

Process Contributors. To understand the origins of the energy use, GHG emissions, land use, MSP, toxicity, and water use results, we identified the process components contributing to each metric ([Figure 3](#), [Tables S32–S34](#)). Across all technologies, water use is dominated by process and cooling water. Waste disposal, which includes plastic losses and process waste, is a contributor to toxicity; its role can vary by $\pm 10\%$ depending on the assumed diversion to landfill versus incineration (80:20 ratio used here). The MSPs of all technologies are linked to plastic bale prices, especially for the historically more expensive postconsumer HDPE and PET.

For mechanical recycling, electricity consumption accounts for 90%–97% of energy use and GHG emissions and is a top contributor to all other metrics ([Figure 3A–D](#)). Most of these impacts originate from coal- and natural gas-based electricity generation (63% of the assumed 2016 U.S. grid mix).⁴⁵ Here, 90% of electricity is used for drying, melt extrusion and filtration, pelletization, and solid-state polymerization, which enable the recovery of high quality recycle.³¹

The economic and environmental impacts of dissolution are linked to the solvent for polyolefins, benzyl alcohol for PET), antisolvent (hexane for polyolefins, methanol for PET), and steam for solvent recovery ([Figure 3E–H](#)). The high impacts of the organic solvents and antisolvents are associated

with fossil feedstock use and GHGs emitted during their production.

PET enzymatic hydrolysis has a diverse set of contributors ([Figure 3I](#)).^{14,46} Energy and GHG emissions are dominated by electricity—of which over 90% is used for the extrusion and cryo-milling of PET flake into amorphized powders, as well as steam for EG recovery and sodium hydroxide (NaOH) for pH control of the depolymerization reactor. NaOH is also present in the remaining metrics due to the high electricity use and chlorine emissions associated with its synthesis, and enzymes account for 26% of land use because of the glucose required for their production. For glycolysis, top contributors for most metrics include EG (the depolymerization solvent), pretreatment of the PET feedstock to flake (due to electricity and natural gas consumption for shredding and drying), and electricity (for refrigeration of the BHET crystallizer and extrusion of the produced PET) ([Figure 3J](#)). Vapor methanolysis is similarly affected by processing of PET to flake, as well as refrigeration and steam for the reaction and monomer recovery stages ([Figure 3K](#)). Methanol (the depolymerization solvent) is less influential than the organic solvents used in glycolysis or dissolution because its production has lower environmental impacts (0.43 kg CO₂/kg in comparison to 1.5 kg CO₂/kg for EG or 3.4 kg CO₂/kg for xylene).

Sensitivity Analysis. Having identified the top cost and environmental impact drivers of the plastic recycling technologies, we explored their potential for improvement through univariate sensitivity analysis ([Figure 4](#), [Figures S14–S21](#), [Tables S35–S37](#)).

For mechanical recycling of HDPE, MSP is impacted by polymer yield (+5% for pessimistic case or –11% for optimistic case), bale quality (+14%/–10%), bale price (+8%/–6%), and

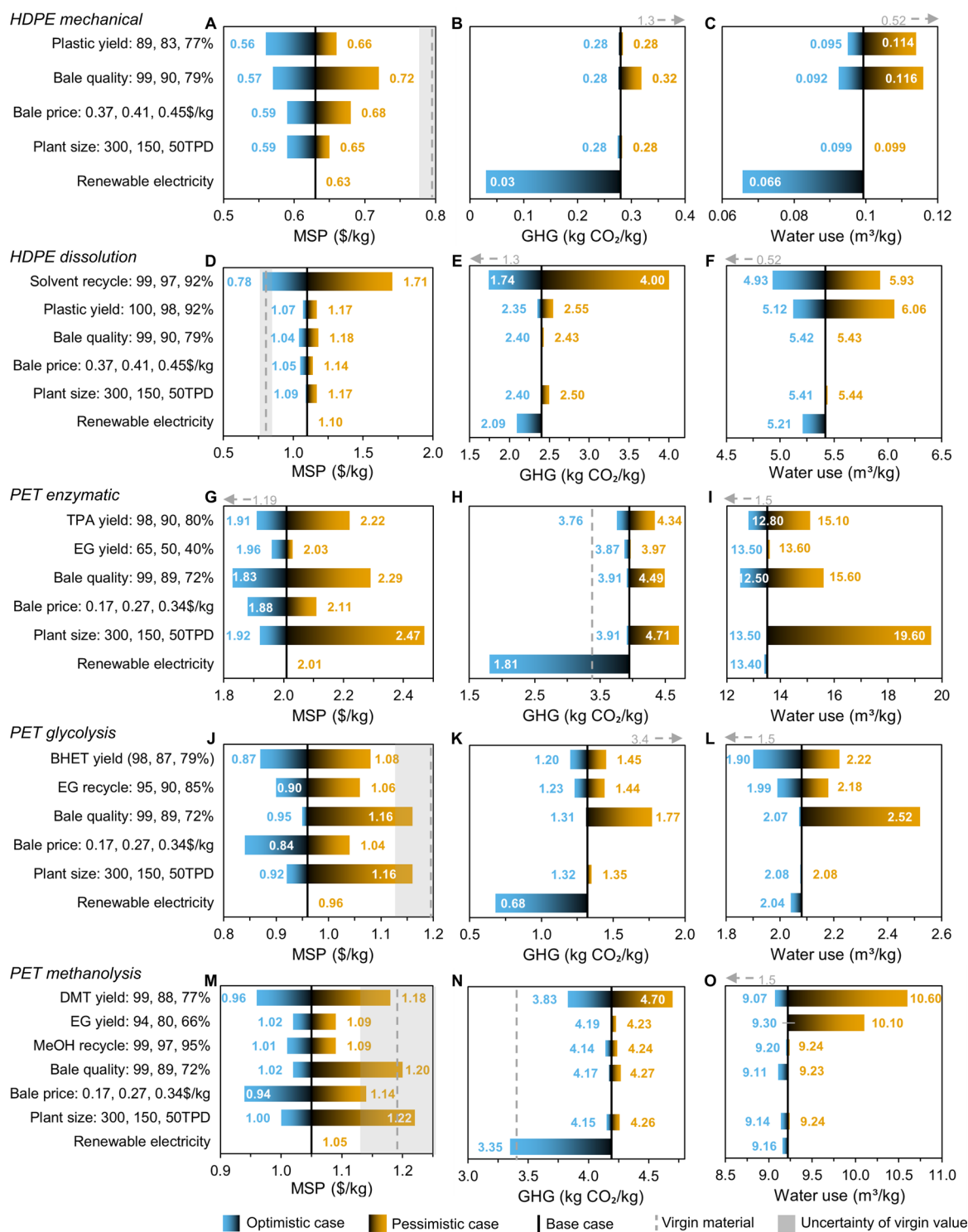


Figure 4. Sensitivity analysis of (A–C) HDPE mechanical recycling, (D–F) HDPE dissolution, (G–I) PET enzymatic hydrolysis, (J–L) PET glycolysis, and (M–O) PET vapor methanolysis. Pessimistic (orange) and optimistic (blue) cases are shown respective to the base case (black lines, same values as Figure 2) and virgin polymers (gray dashed lines, uncertainty shown in semitransparent gray when available). If the virgin value is beyond the graph range, the gray arrows indicate whether it is higher or lower than the assessed recycling metric. Energy use, land use, and toxicity, as well as sensitivity analysis of LDPE and PP, are in Figures S14–S21. Raw data are available in Tables S35–S37.

plant size (+3%/–6%) (Figure 4A–C, Figure S14, Table S35). Recovery rate and bale quality also affect water use but have minimal impacts on GHG emissions. Higher process yields slightly reduce the consumables (washing agents, defoamers, water) required per unit of recyclate. Electricity was shown to dominate environmental impacts; switching to a fully renew-

able grid therefore decreases GHG emissions, water use, energy use, and toxicity by 91%, 34%, 85%, and 15%, respectively (land use increases by 8%). In all scenarios, mechanical recycling maintains MSP, GHG emissions, energy use, and water use lower than virgin polymer. Toxicity and land use remain higher than virgin manufacture as these

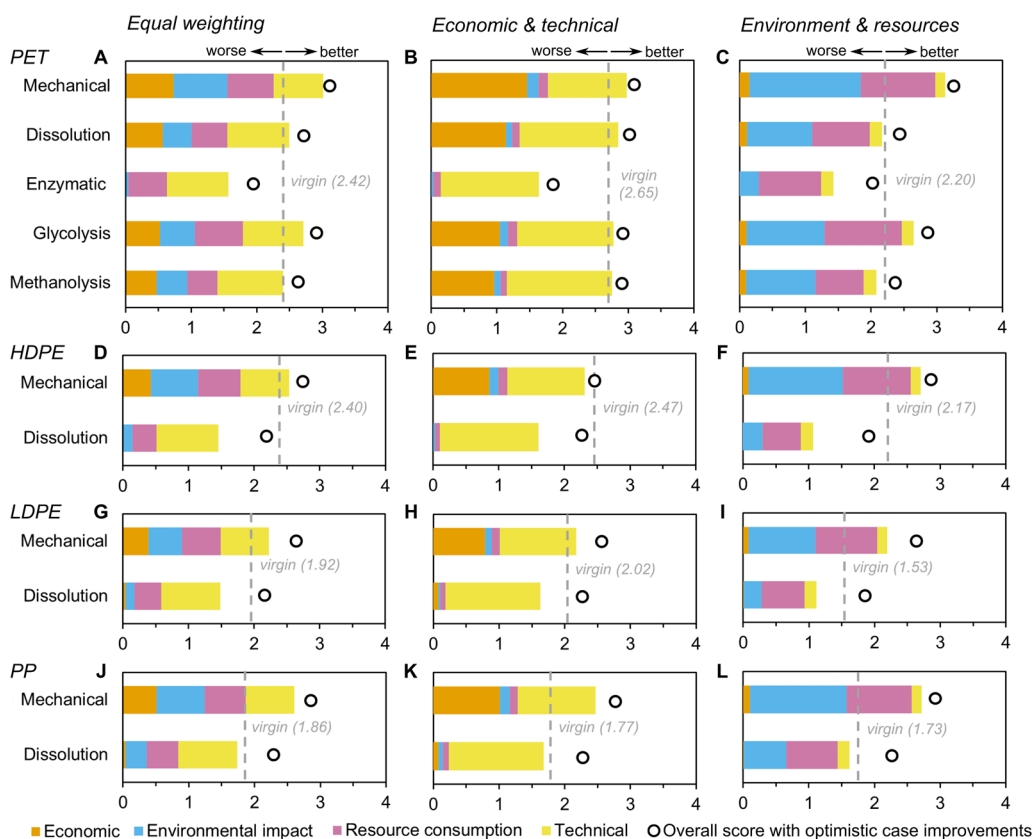


Figure 5. MCDA for recycling of (A–C) PET, (D–F) HDPE, (G–I) LDPE, and (J–L) PP, as well as of virgin PET, HDPE, LDPE, and PP manufacture for comparison. Higher scores are preferential. The left-most plots use equal weightings for economic (orange), environmental impact (blue), resource consumption (pink), and technical (yellow) scores. The middle graphs scale preferentially toward the economic (50% weighting) and technical (40% weighting) scores. The right-most plots scale preferentially toward the environmental (50% weighting) and resource consumption (40% weighting) scores. The black circles indicate an idealized future scenario where all the optimistic cases from the sensitivity analyses are combined for a given recycling technology. Raw data are available in Table S38.

metrics are linked to washing agent use and mechanical recycling infrastructure, neither of which is significantly altered in the sensitivity cases. Mechanical recycling of LDPE, PET, and PP show comparable trends (Figures S15–S17).

The feasibility of dissolution is determined by solvent recycling rate (Figure 4D–F, Figure S14, Table S36). Given that xylene and hexane drive all metrics, solvent recycling rate adjusts MSP, GHG emissions, and water use by +55%, +67%, and +9%, respectively, in the pessimistic case, or by –29%, –28%, and –9%, respectively, in the optimistic case. Although steam consumption increases with higher solvent recycling, this is counteracted by the reduction of environmentally impactful and costly solvents. Polymer yield, bale quality, plant size, and a renewable electricity grid have moderate effects. Sensitivity analyses of LDPE, PET, and PP dissolution yield similar results (Figures S18–S20). While HDPE dissolution remains more costly and environmentally impactful than virgin production in most scenarios, dissolution of LDPE, PP, or PET offers MSP, energy use, and GHG emissions competitive with or better than their virgin equivalents.

Sensitivity cases for chemical recycling of PET show that process yields are crucial variables (Figure 4G–O, Figure S21, Table S37). Improving process yields results in more product, fewer incoming utilities per unit product, and less outgoing waste and is influenced by monomer yields, extraction of PET from the incoming feedstock, and solvent recycling, among others. Pessimistic and optimistic cases for monomer recovery

can alter MSP by as much as +13%/–9% (BHET yield for glycolysis and DMT yield for methanolysis), GHG emissions by +12%/–9% (glycolysis and methanolysis), water use by +15%/–2% (methanolysis), energy use by +11%/–11% (glycolysis), land use by +12%/–8% (methanolysis), and toxicity by +16%/–14% (glycolysis). Material retention is also dictated by the quantity of PET recoverable from the incoming feedstock; pessimistic and optimistic bale qualities therefore impact all metrics. Solvent recycling affects overall process efficiency as well, and higher recovery of EG (glycolysis) or methanol (methanolysis) can reduce MSP by 6% or 4%, respectively, GHG emissions by 7% or 1%, and water use by 9% or 1%. Enzymatic recycling is an electricity-intensive process rather than an organic solvent-intensive process, and thus, utilizing renewable energy sources decreases GHG emissions by 54% to a level lower than virgin PET production and approaching glycolysis. Glycolysis and methanolysis, which are less reliant on electricity, nevertheless exhibit GHG emission reductions of 48% and 20%, respectively, with renewable alternatives. Plant size has a large impact on MSP (+23%/–5% for enzymatic hydrolysis, +21%/–4% for glycolysis, and +16%/–5% for methanolysis) as the capital investment and utilities required for chemical recycling do not scale linearly. Our previous studies on enzymatic hydrolysis have shown that variables such as solids loading, removal of mechanical pretreatment, and alternative monomer recovery techniques can also reduce MSP.^{14,46} While glycolysis

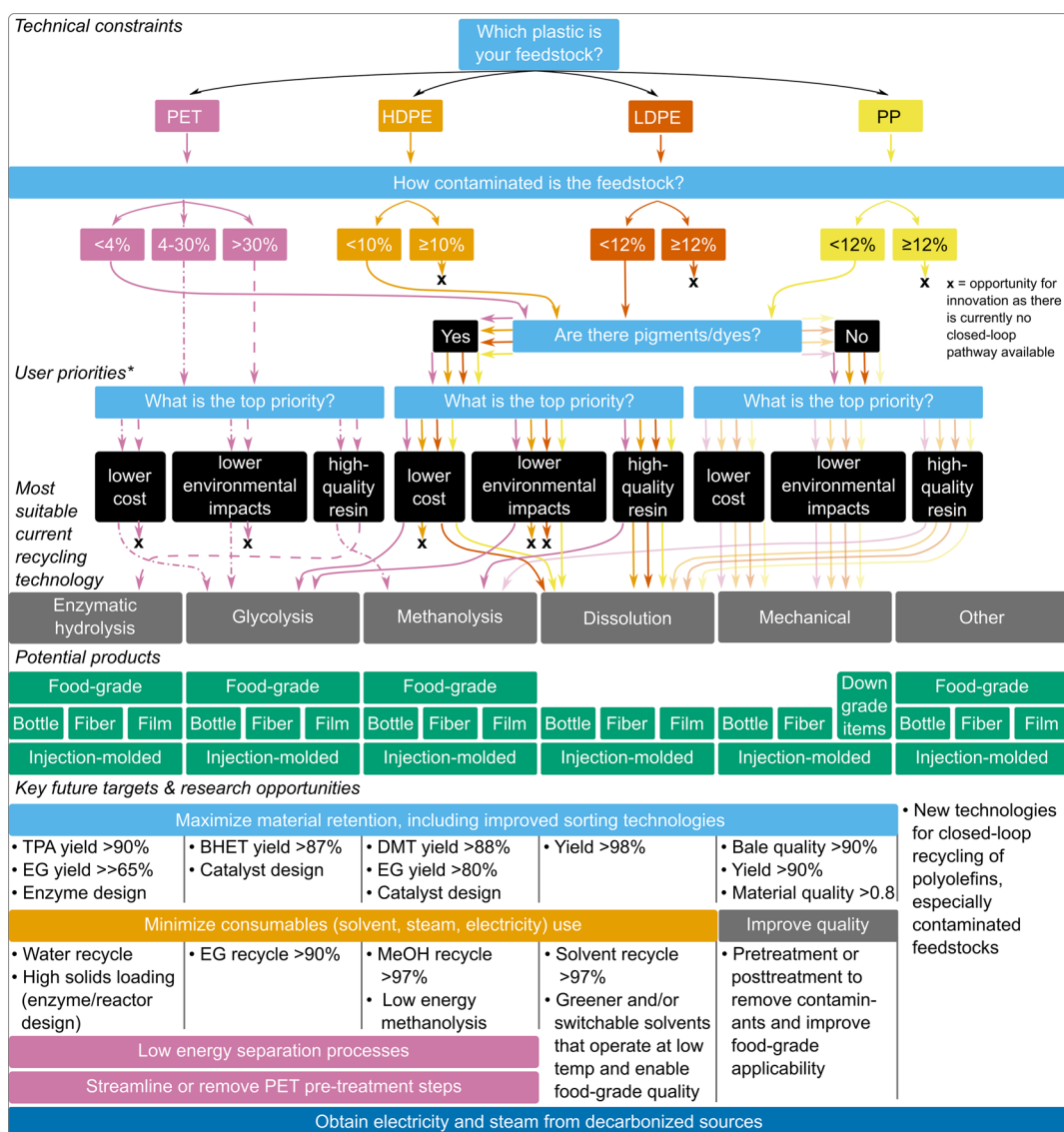


Figure 6. Decision tree for determining the most suitable recycling technology for a given feedstock given a set of criteria, as determined from this analysis. Categorizations and recommendations are based on average metric values and most likely scenarios and will therefore have exceptions. “Lower” cost and environmental impacts are benchmarked against virgin polymer manufacture and any other applicable recycling options. Research recommendations for each technology are listed in the bottom half of the figure.

outperforms virgin PET across most metrics, methanolysis and enzymatic hydrolysis remain above virgin levels across sensitivity scenarios (excepting MSP for methanolysis and renewable electricity impacts on GHG emissions). This suggests that multiple aspects of these technologies must be improved simultaneously to achieve viability.

Multicriteria Decision Analysis. MCDA, a systematic technique for evaluating and ranking options across priorities, can enable visualization of the overarching performance of closed-loop plastic recycling options (Figure 5, Table S38). Three different scenarios were assessed, including “equal weighting” where economic, environmental impact, resource consumption, and technical scores are given equal importance; an “economic and technical” scenario to reflect likely industrial priorities; and an “environment and resources” option that represents a future prioritization of sustainability. In all cases, a higher score indicates better overall performance.

PET mechanical recycling and glycolysis outperform virgin PET for all scenarios (Figure 5A–C). Methanolysis and

dissolution improve their scores to above virgin levels for the “economic and technical” weighting. This is because both processes have high economic scores (lower MSPs than virgin PET) and technical scores (material quality similar to virgin PET) but lower environmental and resource scores (high GHG emissions, toxicity, land use, and water use). Enzymatic PET hydrolysis does not currently surpass the virgin baseline under any modeled scenario.

Mechanical recycling of HDPE outperforms virgin production under “equal” and “environment and resources” weightings, although its low technical (low material quality) and economic (high MSP) scores reduce its performance under the “economic and technical” scenario (Figure 5D–F). Dissolution does not reach the virgin threshold for any weightings because of its higher MSP, energy use, land use, toxicity, and water use. Similar results are observed for LDPE and PP (Figure 5G–I and Figure 5J–L, respectively), although mechanical recycling performs better under the “economic and technical” weighting than the HDPE case due to the lower

postconsumer LDPE and PP bale prices and correspondingly lower MSPs.

According to this analysis, mechanical recycling and PET glycolysis offer overarching opportunities for supplanting virgin polymer production. The other emerging recycling technologies perform worse under the “environment and resources” weighting than the “equal” or “economic and technical” scenarios. This can be attributed to high GHG emissions (electricity, steam, process solvents), toxicity (waste disposal), and water use (cooling water). By streamlining these processes with a combination of the optimistic cases used in the sensitivity analysis, all technologies can improve to levels approaching (HDPE dissolution, PET enzymatic hydrolysis) or exceeding (all others) virgin manufacture (black circles in Figure 5). Under this optimized scenario, mechanical recycling scores increase by 3%–20%, dissolution by 5%–60%, glycolysis and methanolysis by 5%–15%, and enzymatic hydrolysis by 10%–40%, depending on the polymer and score weighting. Furthermore, the MCDA does not necessarily indicate that low-scoring technologies should be abandoned. These techniques may not be advantageous across all metrics but could provide benefits in specific priority areas, especially with continued improvements as the technologies mature.

Research Recommendations. The reported analysis enables the recommendation of closed-loop plastic recycling applications and research opportunities. The decision tree in Figure 6 is based on current technology capabilities and is designed to clarify connections between a given feedstock, its suitable recycling technique(s), and potential polymer products. The decision tree considers average metric values and most likely scenarios, and each categorization will have exceptions.

Starting from the type of plastic, the feedstock is categorized by contamination level (wt %, based on Robustness A and Robustness B). Material quality requirements and other priorities are used to guide the reader toward the most suitable recycling technology. While the presence of colorants was selected as a criterion here given its documented effect on the quality of recycled plastic,^{11,31,33} other additives such as antioxidants or plasticizers could also be included as more data become available. High purity feedstocks have an existing pathway toward low cost and low environmental impact (although not necessarily high quality) mechanical recycling. However, real-world data indicate that much collected plastic is unsuitable for this category. Postconsumer PET bottle bales are estimated to contain an average of 14.1% and 12.2% contamination in California and Belgium, respectively.^{47,48} Many bales will thus fall within the 4%–30% contamination range most applicable to chemical recycling processes, with glycolysis preferred for low cost and environmental impact priorities and methanolysis for higher quality, color-free material. HDPE bales in California have 4.4%–11.1% contamination,⁴⁷ while HDPE bottles from Belgium have an average contamination of 13.8%.^{47,48} Similarly, Belgian PP bottle bales have an average contamination of 21.4%.⁴⁸ Large portions of postconsumer HDPE, PP, and likely LDPE are expected to fall in the >10% contamination range, for which there is currently no closed-loop recycling process available. These pathways are marked with an “x” in the decision tree and represent necessary areas for innovation.

The decision tree highlights opportunities to combine recycling technologies, creating a supply chain where materials are diverted to the appropriate option based on performance

metrics, targeted properties, and contamination levels. Such a system would involve complex logistics, but it could be facilitated through local hubs in which multiple recycling technologies are operated in tandem using different portions of postconsumer plastic sourced from a nearby MRF.

Although several of the assessed recycling technologies offer pathways toward applications and advantages over virgin polymer production, there is need for improvement. The bottom half of Figure 6 lists targets and research areas for each technology. Mechanical recycling is an economically and environmentally feasible option, but it suffers from poor material retention, quality, circularity, and robustness. Sensitivity analyses showed that increasing bale quality (>90%) and polymer yield (>90%) could improve the viability of mechanical recycling. Advanced sorting technologies, such as fluorescent markers or robotic sorting coupled with artificial intelligence, could complement the techniques used today to achieve these goals and minimize problematic contamination.³¹ Innovative pretreatment or posttreatment technologies could increase material quality. Dissolution might be used to remove certain components from multilayer products, enabling mechanical recycling of the remaining polymer.⁴⁹ Catalysis could be incorporated into the extrusion step to selectively degrade dyes or other organic contaminants and produce food-grade recyclate. Stabilizers, compatibilizers, and other additives—and particularly their roles and fates over multiple recovery cycles—also warrant further exploration.³¹

Dissolution generates quality recyclate at high yields and reasonable prices but has high environmental impacts due to organic solvent use. Switching to biobased solvents such as limonene could minimize fossil resource consumption, but analyses must be conducted on their economic and environmental effects.⁵⁰ Switchable solvents, which have reversibly changeable physical properties, offer an opportunity to recycle laminated or multipolymer products.^{51,52} Ideally, the selected solvent should dissolve the polymer at low temperatures to minimize heat requirements and have a low boiling point to enable energy-efficient recovery by distillation as well as sufficient removal during drying to produce food-grade quality polymer.

Enzymatic PET hydrolysis as modeled is not predicted to compete with virgin PET production or other closed-loop recycling technologies. Monomer yields must increase, while steam, cooling water, and NaOH consumption should be minimized through unconventional product recovery techniques.^{14,46} Switching to renewable electricity and minimizing electricity consumption through energy-efficient feedstock pretreatment techniques (e.g., solvent annealing or swelling) or removal of amorphization (by developing enzymes compatible with crystalline PET) will be essential.^{14,46}

Of the PET chemical recycling techniques, glycolysis has the lowest environmental impacts and MSP. The technology would nevertheless benefit from improved monomer yields, for which new catalysts (e.g., organo-metallic, organic, ionic liquid) may be needed.⁵³ Catalyst design could also address BHET discoloration, which results in tinted PET and limits its applications.³⁶

PET methanolysis was shown to require yields of >88% for DMT and >80% for EG. Exploring catalysts beyond traditional zinc acetate could help reach these goals. Steam was also shown to dominate GHG emissions, energy use, and land use. Steam consumption is primarily linked to the product recovery stage, as the many miscible solvents and products present in

methanolysis require energy-intensive separation through distillation columns and crystallization. Incorporating further heat exchanger networks, utilizing alternative separation techniques such as selective membranes, or pursuing low energy methanolysis⁵⁴ rather than the superheated vapor process modeled here, could reduce the utilities requirements of methanolysis and improve its feasibility.

Across all technologies, several trends emerge. First, material retention must be improved to levels approaching the yields seen in virgin polymer production (95%–99%).²⁵ This requirement could be met by improved sorting technologies and enhanced process yields, which will likely benefit from the learning effect and economies of scale. Second, there should be a focus on minimizing consumables such as solvents, electricity, steam, and NaOH, many of which are used for pretreatment and posttreatment procedures (e.g., flake production and product recovery) rather than the more heavily researched recycling step. Recycling should also be treated as a decarbonization opportunity. In contrast to a virgin polymer industry reliant on fossil fuels for feedstocks, heating, and pressurization, the assessed recycling technologies use electricity and lower temperatures and pressures, which could be more easily generated from renewable sources. Lastly, there is a need for closed-loop recycling technologies for polyolefins. Only mechanical and dissolution recycling are currently available. Pyrolysis and gasification operate on (mixed) HDPE, LDPE, and PP, but the resulting intermediates are typically reintegrated into the petrochemical industry where only a small fraction is used for closed-loop recycling. As discussed in multiple reviews,^{4,55} there is still considerable space for innovation within the circular polyolefins field.

CONCLUSIONS

Closed-loop recycling technologies are one of several pathways toward a circular economy for plastics. Here, we analyzed technical, economic, and environmental metrics for closed-loop recycling of the most common consumer polymers. Mechanical recycling outperformed all other technologies, as well as virgin plastic production across economic and environmental considerations, but it exhibited lower material qualities and other technical metrics. Of the chemical recycling techniques for PET, glycolysis offered the best economic and environmental performances. Organic solvents, steam, and electricity were identified as key drivers of the MSP, energy use, GHG emissions, land use, toxicity, and water use of the technologies. Sensitivity analysis and MCDA showcased areas for future improvement, including process yields, consumable reduction or replacement, utility decarbonization, and closed-loop recycling options for polyolefins. This work quantitatively characterized the performances of plastic recycling technologies and established a robust methodology for comparing new recycling processes as they emerge in the future.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.2c05497>.

Details on all Aspen Plus process models, MCI calculations, Figures S1–S21, and Tables S1–S38. (PDF)

AUTHOR INFORMATION

Corresponding Author

Taylor Uekert – Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States; Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States; orcid.org/0000-0001-7645-9166; Email: taylor.uekert@nrel.gov

Authors

Avantika Singh – Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States; Catalytic Carbon Transformation and Scale-Up Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States

Jason S. DesVeaux – Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States; Catalytic Carbon Transformation and Scale-Up Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States

Tapajyoti Ghosh – Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States

Arpit Bhatt – Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States; orcid.org/0000-0002-3780-1800

Geetanjali Yadav – Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States; Catalytic Carbon Transformation and Scale-Up Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States

Shaik Afzal – Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States; Catalytic Carbon Transformation and Scale-Up Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States

Julien Walzberg – Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States

Katrina M. Knauer – Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States; Renewable Resources and Enabling Sciences Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States

Scott R. Nicholson – Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States; Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States

Gregg T. Beckham – Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States; Renewable Resources and Enabling Sciences Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States; orcid.org/0000-0002-3480-212X

Alberta C. Carpenter – Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States; Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium, Golden, Colorado 80401, United States

Complete contact information is available at:
<https://pubs.acs.org/10.1021/acssuschemeng.2c05497>

Author Contributions

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Notes

The authors declare the following competing financial interest(s): G.T.B. has submitted patent applications on enzymes for PET recycling.

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ABBREVIATIONS

BHET; bis(2-hydroxyethyl) terephthalate; DMT; dimethyl terephthalate; EG; ethylene glycol; GHG; greenhouse gas; HDPE; high density polyethylene; LCA; life cycle assessment; LDPE; low density polyethylene; MCDA; multicriteria decision analysis; MCI; material circularity indicator; MFI; materials flows through industry; MFR; melt flow rate; MRF; materials recovery facility; MSP; minimum selling price; PET; polyethylene terephthalate; PP; polypropylene; TPA; terephthalic acid

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