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Research Article

Circular Economy Sustainability Analysis Framework for Plastics: Application for Poly(ethylene Terephthalate) (PET)

Ulises R. Gracida-Alvarez, Hui Xu, Pahola Thathiana Benavides, Michael Wang, and Troy R. Hawkins*

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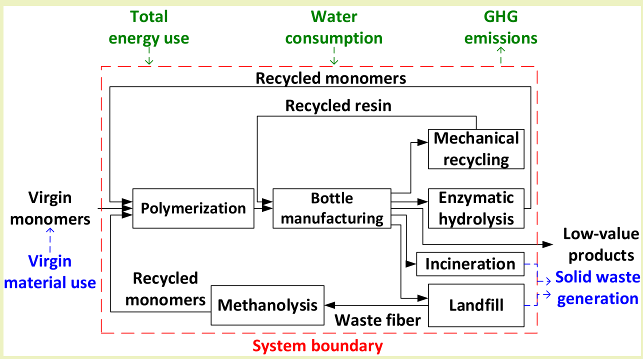
ABSTRACT: The establishment of the circular economy (CE) for plastics aims to reduce material losses and dependence on virgin materials; however, this practice does not necessarily imply reduction of life-cycle impacts. In this study, a CE sustainability analysis framework combining life-cycle assessment (LCA) and material flow analysis (MFA) was developed to simultaneously evaluate the life-cycle impacts and circularity metrics of implementing different CE strategies of production of plastic packaging, using poly(ethylene terephthalate) (PET) bottles as an example. The strategies included increasing the recycling rate of PET bottles and integrating two chemical recycling technologies in industrial development: enzymatic hydrolysis and methanolysis. The energy use of enzymatic hydrolysis and methanolysis was estimated to be 57 and 38 MJ/kg PET, respectively, while the two technologies accounted for greenhouse gas (GHG) emissions of 3.0 and 2.0 kg CO_2 e/kg PET, respectively. The analysis at the system level demonstrated that compared to the current practice, relying on 97% virgin PET resin, the joint implementation of these strategies generated similar GHG emissions (3.2 kg CO_2 e/kg bottle) but reduced virgin material use and solid waste generation by 56 and 64%, respectively. Based on present technology development, increasing the share of mechanically recycled resin in bottle manufacturing and using a decarbonized electricity grid resulted in 14 and 9% lower GHG emissions, respectively, than the current supply chain.



KEYWORDS: *circular economy, chemical recycling, upcycling, poly(ethylene terephthalate), life-cycle assessment, material flow analysis, enzymatic hydrolysis, methanolysis*

INTRODUCTION

Plastics have become one of the most essential products for our society as they are durable, lightweight, and functional. These attributes led to a global production of 368 million tonnes (MMT) of plastics in 2019 that is expected to double in the next 20 years.[1,2] As of 2020, plastics for packaging comprise around 42% of the global plastic production.[3−5] The growing production of short-lifetime plastic packaging has led to an increase in the generation of plastic waste, accounting for around 13% of the total amount of municipal solid waste in the United States (U.S.)[5,6] of which only 9% is recycled.[6] The rest is landfilled (75%) or incinerated (16%). The mismanagement of landfills is responsible for plastic pollution in terrestrial and marine ecosystems and has generated increased attention to recycling as a strategy to reduce plastics pollution and promote a circular economy (CE).

The reutilization of waste plastics through mechanical and chemical recycling technologies, which are discussed in Section S1 in the Supporting Information (SI), reduces solid waste generation and unnecessary use of virgin feedstocks, which are among the objectives of the CE. CE has gained relevance due to the current policy initiatives that propose it as a strategy to reduce pollution and promote decarbonization. CE is a strategy for production systems that aims to increase the value and use of materials by inducing their retention and restorative flows in the economy.[7,8] This material retention can be achieved through reducing waste by increasing recycling yields and collection rates, improving current recycling technologies, or developing new technologies that convert waste materials into new products of higher (upcycling), similar, or lower value (downcycling). However, the implementation of these strategies will rely on how effectively they fulfill their goals.

The selection of the methodologies and metrics for evaluation is an important step in the assessment of CE strategies. Some approaches[9−11] evaluate circularity through product attributes, like recycled content, lifetime, and quality,

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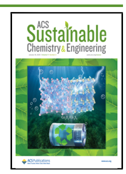

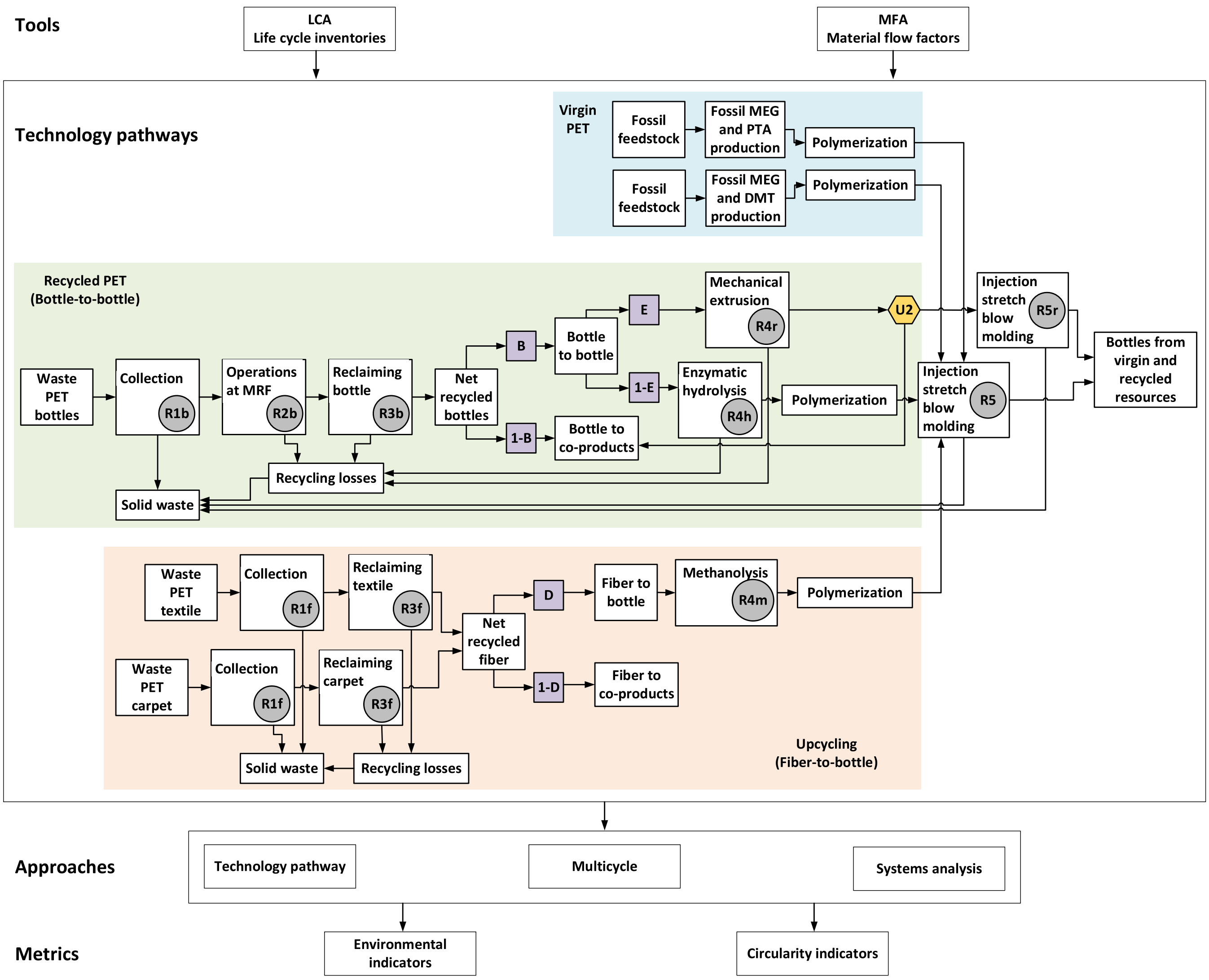

Figure 1. CE sustainability analysis framework. Material flows are represented by arrows and flow factors are enclosed in circles, squares, and hexagons. R1b: Recycling rate of bottles, R2b: Yield of sorting at material recovery facilities (MRF), R3b: Yield of bale-to-flake processing, R4r: Yield of mechanical recycling, R5r: Yield of bottle manufacturing of mechanically recycled resin, R4h: Yield of enzymatic hydrolysis, R5: Yield of bottle manufacturing of virgin and chemically recycled resin, R1f: Recycling rate fiber, R3f: Yield of fiber reclaiming, R4m: Yield of methanolysis, B: Percentage of recycled flakes to bottle production, E: Percentage of flakes to mechanical recycling, D: percentage of densified fiber-to-bottle production, U2: Recycled content limit of mechanical recycling, LCA: life-cycle assessment, MFA: material flow analysis, MEG: mono ethylene glycol, PTA: purified terephthalic acid, DMT: dimethyl terephthalate.

whereas others focus on the comparison with other end-of-life (EOL) options.[12] More comprehensive approaches should also evaluate material retention and waste minimization, which are key objectives of circularity. For example, a methodology like material flow analysis (MFA) could evaluate the resource consumption reduction and solid waste generation of CE strategies. MFA estimates the external and internal flows in a system and evaluates the interactions of these flows.[13−15] Furthermore, due to the growing concern that higher circularity does not always lead to better sustainability outcomes,[16,17] a sustainability assessment is necessary to ensure CE goals are met without increasing the environmental impact. Cumulative energy demand, water and carbon footprint, sustainable process index, and life-cycle assessment (LCA) are among the methodologies used to evaluate the environmental impact of CE strategies.[13] Of these, LCA is the most comprehensive as it encompasses other methodologies like cumulative energy demand, and water and carbon footprint. LCA also uses resource consumption data to calculate the environmental impacts during the different stages of the process.[18] The integration of multiple methodologies for the evaluation of CE—for instance, combining MFA with LCA—provides analysis capable of identifying trade-offs between circularity and environmental impacts,[13,14,19] as suggested by earlier studies evaluating the sustainability of CE strategies for plastic packaging.[16,20,21]

Most studies have evaluated the impacts of CE strategies as alternate waste management practices rather than as production technologies to translate from linear to circular manufacturing. To date, of the few studies analyzing the environmental and circularity impacts of CE strategies in the plastic packaging supply chain,[16,20,21] only one[20] considered chemical recycling and none incorporated upcycling into their assessments. For recycled resins, like recycled poly(ethylene terephthalate) (rPET), that are primarily downcycled,[22] there is an important need to evaluate CE strategies that could

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potentially divert their use toward high-value applications like food packaging. Therefore, this study analyzed the environmental and circularity impacts of increasing the recycling rates of PET bottles and implementing bottle-to-bottle chemical recycling and fiber-to-bottle upcycling to further understand the effects of increasing the use of recycled resins in the supply chain of PET bottles. To do so, a CE sustainability analysis framework was developed. The framework integrated LCA and MFA and employed detailed life-cycle inventories of all of the stages of the recycling process and key factors influencing the material flows.

■ METHODOLOGY

Goal and Scope. The objective of this study was to quantify and compare the environmental and circularity metrics for conventional and circular pathways of plastic packaging production, using PET bottles as a case study. To do so, we developed a modeling framework that considers the degree of circularity and the resulting life-cycle impacts of CE-enabling technologies and also identifies barriers that obstruct material restoration and process stages in need of improvement. The framework was built using an expanded version of the Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET) model. It represents the supply chain inputs of the various recycling and manufacturing technologies of PET bottles in the United States.[23]Figure 1 shows the structure and the components (tools, technology pathways, approaches, and metrics) of the CE sustainability analysis framework, which are described in the following sections.

Tools and Metrics of the Analysis. The LCA tool in the framework analyzed the life-cycle impacts of the recycling and manufacturing technologies on energy use and the environment, including greenhouse gas (GHG) emissions and water consumption. The life-cycle impact of a technology was calculated by summing the products of the impact per unit of input material (i.e., MJ per kg of waste PET) as estimated in GREET and the amount of material processed for each process stage of the technology, as represented by eq 1

$$I_{et} = \sum_{s}^{n} M_{st} \cdot I_{es} \tag{1}$$

where I_{et} is the life-cycle impact e (i.e., total energy use, water consumption, and GHG emissions) of technology t (e.g., mechanical recycling, enzymatic hydrolysis), M_{st} is the amount of input material processed in stage s of technology t, and I_{es} is the life-cycle impact per input material of stage s in technology t. The environmental impact on a system composed of multiple technology pathways was estimated by summing the products of the material flow of bottles (M_t) and the environmental impact for each technology, as described in eq 2

$$I_{e_{\text{system}}} = \sum_{t}^{n} M_t \cdot I_{et} \tag{2}$$

Circularity was evaluated through MFA. Virgin material use and solid waste generation were chosen as circularity metrics as they are directly dependent on the restorative flows of materials in the system. The establishment of flow factors for tracking the material flows along the different processes and technologies is an important component in estimating the circularity metrics. Section S2 in the SI presents the data sources and definitions of these flow factors. Figure 1 shows the connections between the material flows along the supply chain of PET production, which were used to estimate the amount of solid waste generated and the mass flow of bottles from each technology contributing to the bottle production supply. Solid waste generation for each technology was calculated using eq 3

$$\text{SWG}_t = \sum_{s}^{n} M_{st} \cdot (1 - F_{st}) \tag{3}$$

where SWG_t is the solid waste generation of technology t, M_{st} is the material flow through the process stage s in technology t, and F_{st} is the recycling rate or yield factor of the process stage s in technology t. The total solid waste generation in the system was estimated by the sum of the SWGs of all of the technologies involved. Virgin material use was estimated with eqs 4 and 5

$$M_t = \sum_{s}^{n} M_{st} \cdot F_{st} \tag{4}$$

$$\text{VMU} = \text{TM} - \sum_{i}^{n} M_t \tag{5}$$

where M_t is the total mass flow of technology t, VMU is the virgin material use of the system, and TM is the total mass flow of the system.

Conventional versus Circular PET Production Technologies. In the CE framework (Figure 1), we included five technologies for producing PET resin: two fossil or virgin PET (vPET) production pathways and three recycling pathways. Conventionally, PET can be produced via (1) esterification of purified terephthalic acid (PTA) with mono ethylene glycol (MEG) or (2) transesterification of dimethyl terephthalate (DMT) with MEG. The three alternative pathways included were (1) mechanical recycling, (2) enzymatic hydrolysis (chemical recycling), and (3) upcycling (fiber-to-bottle) via methanolysis. Inventory data for vPET production were updated and disaggregated[24,25] while inventories for the stages in recycling technologies were added to the GREET model. Section S3 in the SI provides descriptions and material and energy inputs for each stage of the technologies included in the analysis.

The system boundaries for each of the five technologies were cradle-to-gate, and their process flowcharts are presented in Figures S1 and S2 in the SI. Stages of the recycling pathways included collection, sorting at the material recovery facility (MRF), reclaiming (bale-to-flake) and pelletizing (flake-to-pellet) operations, and transportation among the different stages. The upcycling pathway was similar to the recycling pathways except that fiber products (carpets and textiles) were sent to a collection site, where fibers were taken apart and reclaimed, instead of to an MRF. The analysis of recycling and upcycling pathways considered a cutoff approach in which waste materials were burden-free at the collection site (i.e., energy and emissions associated with virgin product productions were not allocated to recycled materials) as well as the avoided emissions from the traditional EOL waste management practices.[6] The boundary of virgin manufacturing considered upstream production of chemical intermediates, polymerization, and transportation.

Technology Comparison Approaches and Scenarios. Life-cycle impacts and circularity metrics of the recycling

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ACS Sustainable Chem. Eng. 2023, 11, 514−524

technologies were analyzed using three approaches: (1) PET resin production for the comparison among different technologies (cradle-to-gate), (2) multiple cycles of PET bottle recycling to estimate the effects of recycling cycles, and (3) systems analysis of PET bottles to evaluate the collective effect of the different technologies in the supply chain of recycled PET bottles in the United States. The study considered two functional units: one kg of PET resin, for the cradle-to-gate comparison, and one kg of PET bottle, for the multicycle and systems analysis approaches. The functional unit was modified to one kg of PET bottles in the multicycle and systems analysis approaches because their system boundary included the bottle manufacturing stage, through injection blow molding (pellet-to-bottle).

Technology Pathway Approach. The first approach consisted of a cradle-to-gate comparison of the two vPET routes, mechanical recycling, enzymatic hydrolysis, and methanolysis. This analysis used a traditional LCA evaluation of one production cycle and considered the material yields of the different process stages. In the analysis of GHG emissions, all three recycling pathways considered the avoided emissions from conventional EOL management of plastic bottles (e.g., incineration with energy recovery), which contrasts with other approximations that added the avoided emissions of EOL management to the virgin production pathways.[26] This study derived its assumptions about conventional EOL management from U.S. EPA,[6] which reported that 80% of waste PET packaging went to landfills and 20% was incinerated with energy recovery. Since plastic combustion generates electricity, net avoided emissions were accounted for by comparing the GHG emissions associated with U.S. grid electricity generation with the GHG emissions associated with electricity generation via plastics incineration.[23] Life-cycle impacts of the transportation of generated solid waste to a landfill were also included in the EOL management comparison. The transportation assumed similar distance and mode of transportation to those used for collection. Section S4 in the SI shows a sample estimation of these avoided GHG emissions. Additionally, for enzymatic hydrolysis and methanolysis, no credits were accounted from co-produced MEG as this is used in the subsequent polymerization stage.

Systems Analysis Approach. The third approach consisted of a system-level analysis evaluating the transition from the U.S. market of PET bottles in 2017 (assumed as the current state),[27] which relies mostly on vPET, to a market implementing CE strategies to increase the use of rPET. As in earlier studies,[16,20,21] the systems analysis was achieved through use of scenarios that in this study progressively incorporated different CE strategies. The "mechanical maximized" scenario explored the impact of expanding the production of rPET through mechanical recycling by increasing the recycling rate of PET bottles from 29 to 90%. This strategy has been applied by State governments in the United States and could leverage from existing mechanical recycling infrastructure. The "mechanical and chemical" scenario evaluated the combined effects of maximizing recycling rates and implementing enzymatic hydrolysis to recycle half of the waste PET bottles collected. Through this scenario, it is possible to understand the life-cycle impacts of increasing reclaiming yields and producing higher-quality rPET through enzymatic hydrolysis. Finally, the "chemical, mechanical, and upcycling" added the conversion of low-value feedstock (fiber) to high-value application (bottle), which provides both increased feedstock availability and the added value of upcycling. Table 1 provides more detailed information of the scenarios, and Section S5 in the SI presents the values of the flow factors in each scenario.

The addition of the environmental and circularity metrics for each of the technologies involved provided the total life-cycle impacts and circularity metrics for each scenario, which were divided by the total amount of bottles supplied in the market (2.7 MMT) to obtain the results on a per kg of PET bottle basis. The contribution of EOL treatment of the solid waste generated in each scenario was only significant in the evaluation of GHG emissions. In the systems analysis approach, the EOL treatment GHG emissions were obtained by subtracting the GHG emissions from displaced electricity produced during incineration from the GHG emissions of the transportation to landfill and incineration of the solid waste. Details of the EOL management emissions estimations are presented in Section S4 in the SI.

This study also analyzed the effects of multiple recycling cycles in the GHG emissions and retention of the original material used for recycling. Methods and results for this analysis are presented in Section S6 in the SI.

Data Sources for Material Flow Estimations in the Systems Analysis Approach. The MFA of each scenario was based on the U.S. supply chain of PET bottles in 2017 according to the National Association of PET Container Resources (NAPCOR).[27] Values for the recycling rate and the shares of rPET to different products in the current state were also sourced from NAPCOR.[27] The sorting yield of PET in MRFs was taken from Resource Recycling Systems[31] while data from Franklin Associates[32] and industrial equipment specifications[33] provided information for the yields of bottle-to-flake conversion and mechanical extrusion, respectively. The yield for the enzymatic hydrolysis process was obtained from the process modeling reported by Singh et al.[34] Bottle manufacturing yields from virgin and recycled PET were shared by an industry partner. The values and variations of the recycling rate, shares, and yields for the different scenarios are presented in Section S5 in the SI. The estimations of EOL flows of PET waste to landfill and incineration with energy recovery were calculated with the share reported by EPA.[6,35]

In the upcycling pathway, the amount of waste PET fiber from textiles was estimated by multiplying the total generation of waste textile in the United States in 2017 (15.3 MMT), as reported by EPA,[35] by the share of polyester (52%) in the global production of textiles.[29] The amount of waste PET fiber from carpets was calculated from data of the Carpet America Recovery Effort (CARE),[30] indicating that 1.5 MMT of waste carpets were discarded in 2017 in the United States. This amount was multiplied by the composition of fiber in carpets estimated at 45%.[36] With these estimations, the total generation of waste fibers resulted in 8.2 MMT, from which we assumed a recycling rate of 10% based on the average value for carpets and textiles.[30,35] The yield of fiber reclaiming operations (i.e., separation, shredding, densification) was obtained from CARE,[30] and the yield for the methanolysis process was provided by Eastman Chemical Company (details are provided in Section S2 in the SI).

Although downcycling and fiber and film production were not within the LCA system boundary, their material flows were included in the MFA to provide a complete perspective of the supply chain. Downcycling produces mechanically recycled PET resin of low quality not suitable for food packaging;[16]

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ACS Sustainable Chem. Eng. 2023, 11, 514−524

Table 1. Scenarios Used in the Systems Analysis Approach

scenario	description
current state (reference scenario)	Market consisting of 2.7 MMT of PET bottles with a bottle recycling rate (R1b) of 29%. Of the bottles recycled, only 21% are recycled back into bottles (B), through mechanical recycling.[27]
mechanical maximized	Increased recycling rate of PET bottles (R1b) to 90% due to improved collection practices.[28] This increased quantity of collected bottles sent to recycling is entirely used in bottle production. As a result, the percentage of recycled bottles recycled back into bottles (B) increases to 74%. Mechanical recycling is the only technology used for bottle-to-bottle conversion.
mechanical and chemical	Similar to the mechanical maximized scenario but with the assumption that bottle-to-bottle conversion is carried out 50% through mechanical recycling and 50% through chemical recycling.
mechanical, chemical, and upcycling	Similar to the "mechanical and chemical scenario" but with the added introduction of recycled PET produced from polyester fiber. This scenario considers an increase from 10 to 20% in the current waste fiber recycling rate (R1f)[6,29] through the implementation of fiber collection programs.[30] This additional amount of recycled fibers (0.8 MMT per year) is entirely used in fiber-to-bottle conversion.

therefore, recycled PET from downcycling is distributed only among fiber, film, and other products (i.e., sheet and strapping). The distribution share of downcycled PET resin among fiber, film, and other products was estimated at 59, 8, and 33%, respectively, from the data reported by NAPCOR.[27] The amount of vPET used in fiber and film production was 1.2 and 0.3 MMT, respectively, as reported by Heller et al.[37] and Lonca et al.[16] The total amount of PET entering each manufacturing process was multiplied by the corresponding yield of film extrusion[38] and yarn spinning[39] to obtain the amount of fiber and film produced (1.7 and 0.3 MMT, respectively). The produced fibers are assumed to stay in use for more than a year as they are categorized as nondurable goods.[5,37]

RESULTS AND DISCUSSION

Material Flows in the Supply Chain of PET Bottles. In the MFA, the material flow of waste PET to downcycling between scenarios did not change; therefore, increased flow of recycled material to bottle production was driven by modifications of recycling rates and reclaiming yields. The material flows of the current state scenario and the mechanical, chemical, and upcycling scenario are shown in the Sankey diagrams in Figure 2A and Figure 2B, respectively, which shows that reliance on vPET for bottle production decreases when transitioning from the current state to a system using multiple CE strategies. Solely increasing the recycling rate from 29 to 90% (mechanical maximized scenario) decreases vPET use by 31%. With the incorporation of chemical recycling and upcycling, vPET is further reduced (56% less compared to current state). Under this scenario, the shares of virgin, mechanically recycled, chemically recycled, and upcycled (see Section S5 in the SI) resin are 42, 23, 18, and 17%, respectively, which shows a considerable improvement compared to the 97 and 3% shares of vPET and mechanically recycled PET, respectively, observed in the current state scenario. To entirely substitute vPET with PET from upcyling (see Section S7 in the SI) and reduce the share of vPET to 0%, the recycling rate of waste fibers would need to increase from 10 to 45%. Solid waste generation is another important factor for circularity that can be tracked with MFA. In the mechanical maximized scenario, solid waste generation is reduced by 36% compared to the current state scenario; in the mechanical, chemical, and upcycling scenario, solid waste generation is reduced by 64%. The Sankey diagrams of the mechanical maximized and mechanical and chemical scenarios are presented in Figures S4 and S5 in the SI, respectively.

Technology Comparison Approach. The total energy use, GHG emissions, and water consumption of each PET production pathway are shown in Figure 3. Virgin production from the DMT route has the highest total energy use (71 MJ/kg PET) due to the high energy requirements in the production of DMT compared to the production of PTA and DMT in the other pathways. Of the recycling pathways, mechanical recycling has the lowest energy use (13 MJ/kg PET) and enzymatic hydrolysis has the highest value. Compared to methanolysis, enzymatic hydrolysis uses 50% more energy because of the high energy demand of the reclaiming process, which requires the use of cryogenic temperatures and energy-intensive separation processes to obtain PTA.[34] The analysis of net (cradle-to-gate) GHG emissions shows that enzymatic hydrolysis has the highest net GHG emissions (3.0 kg CO_2 e/kg PET) of the production

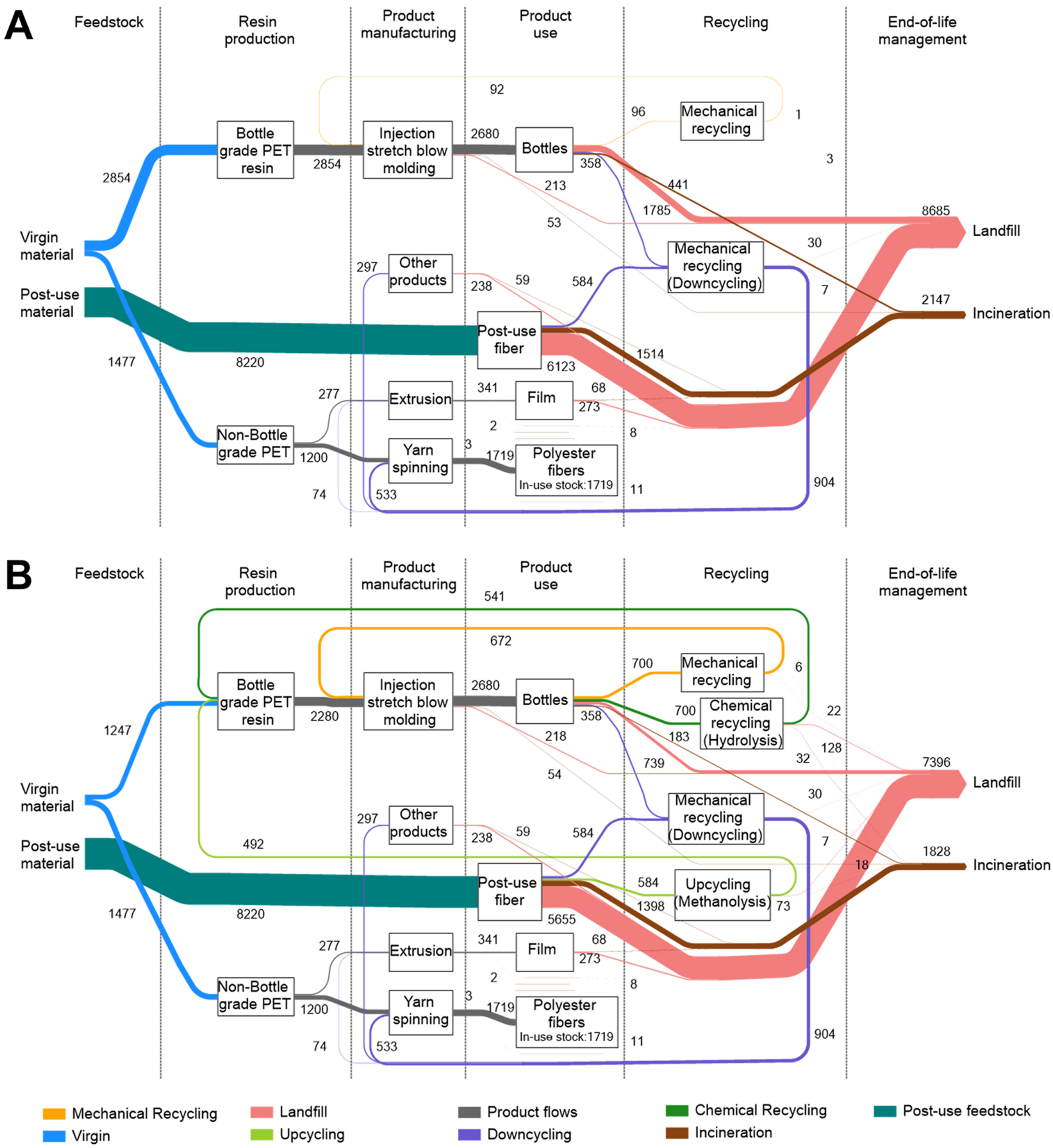

Figure 2. Sankey diagrams of the U.S. supply chain of PET bottles in 2017 for the (A) current state scenario and (B) mechanical, chemical, and upcycling scenario. Numbers indicate thousand tonnes per year.

pathways analyzed, 44 and 71% higher, respectively, than those from vPET production via the DMT (2.1 kg CO_2 e/kg PET) and PTA (1.7 kg CO_2 e/kg PET) routes. For enzymatic hydrolysis and methanolysis, the process stages that convert waste plastic into monomers account for most of the GHG emissions, contributing 65 and 69% of the pathways' total cradle-to-gate GHG emissions, respectively. The analysis of water consumption shows that rPET production via enzymatic hydrolysis consumes approximately three times more water than vPET production via DMT or PTA. The enzymatic hydrolysis process occurs in an aqueous solution from which only 52% of the process water is recovered. In contrast, methanolysis and mechanical recycling have the lowest water consumption of all of the analyzed PET production pathways. Comparison among all analyzed production pathways (Figure 3) demonstrates that mechanical recycling is the pathway with the lowest life-cycle impacts due to its low material and energy requirements. Due to the higher material and energy

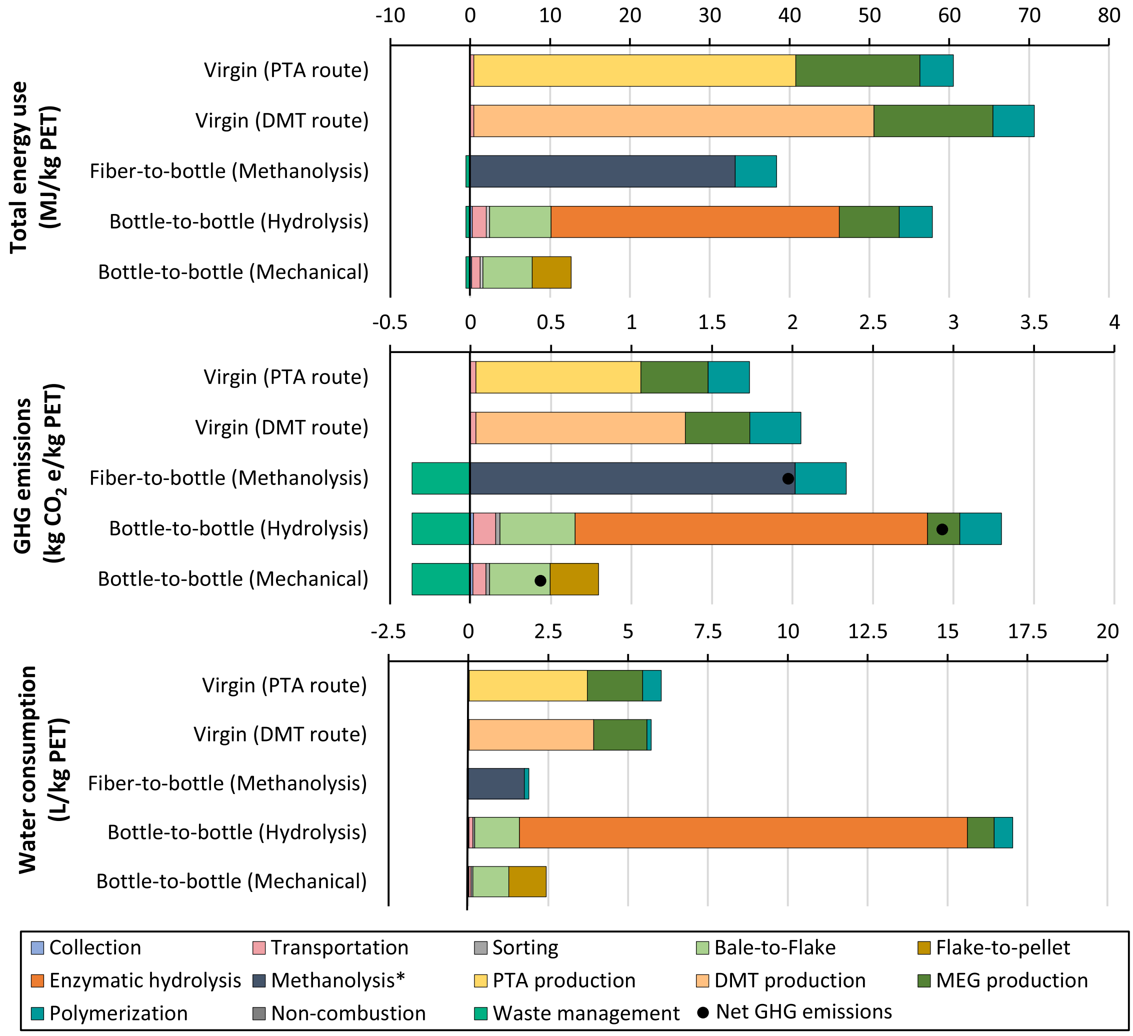

Figure 3. Comparison of life-cycle impacts among PET production pathways. Methanolysis data is aggregated from the waste to resin stages to maintain confidentiality but includes the use of make-up MEG for polymerization through transesterification of DMT.

requirements of depolymerization, enzymatic hydrolysis and methanolysis (3.0 and 2.0 kg CO_2 e/kg PET, respectively) have the highest GHG emissions of all of the pathways.

Systems Analysis. Figure 4 shows the environmental and circularity metrics of the four different bottle production scenarios considering a functional unit of one kg of bottles. The analysis assumes that 90% of vPET is produced from PTA and 10% is produced from DMT. In the total energy use comparison, all three scenarios implementing CE strategies have lower total energy use than the current state (86 MJ/kg bottle) because the energy used in vPET production includes the energy embedded in the monomers used in polymerization. This reduction in total energy use, compared to the current state, ranges from 15 to 21%. In the GHG emissions comparison, the mechanical maximized scenario generates the lowest GHG emissions (2.7 kg CO_2 e/kg bottle) compared to the current state (3.1 kg CO_2 e/kg bottle). This scenario has the highest share (34%) of mechanically recycled resin, which has the lowest GHG emissions (0.5 kg CO_2 e/kg PET) of all of the PET production pathways. Due to the higher GHG emissions of the enzymatic hydrolysis and methanolysis pathways, the GHG emissions of the mechanical and chemical and chemical, mechanical, and upcycling scenarios (3.1 kg CO_2 e/kg bottle and 3.2 kg CO_2 e/kg bottle, respectively) are similar to current-state GHG emissions despite their reduction in emissions from waste management and make-up electricity generation. This is because (1) hydrolysis uses a high amount of natural gas and electricity and employs chemicals, like sodium hydroxide and sulfuric acid, with high GHG emissions per unit of product, and (2) methanolysis also uses a high amount of natural gas. The 14% reduction in GHG emissions observed in the mechanical maximized scenario suggests that since a high share of mechanically recycled resin improves the environmental performance of the system, mechanical recycling should not be disregarded. In the water consumption comparison, the two scenarios that include hydrolysis have the highest water consumption (up to 13 L/kg bottle) compared with the current state (11 L/kg bottle). Hydrolysis consumes over three times more water (17 L/kg PET) than the other PET production pathways, which increases the water consumption in the mechanical and chemical scenario system by 15% compared to the current state.

Although the life-cycle impacts of the scenarios implementing CE strategies are relatively similar to those of the current state, their circularity metrics show significant reductions (see Figure 4). The first and most prominent reduction occurs by raising the recycling rate of PET bottles in the mechanical maximized scenario, reducing virgin material use and solid

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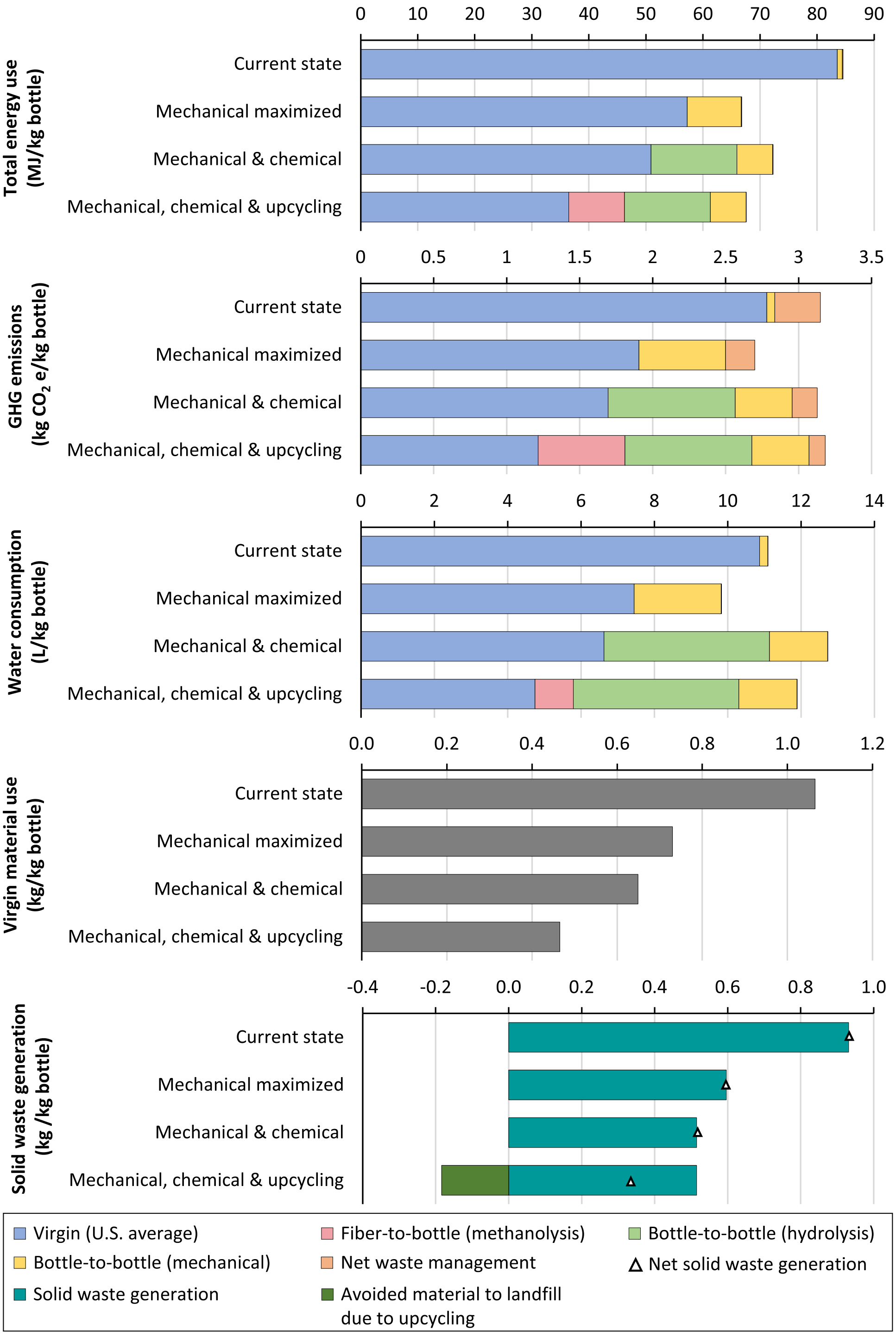

Figure 4. Life-cycle impacts (total energy use, GHG emissions, and water consumption) and circularity metrics (virgin material use and solid waste generation) of the four different bottle production scenarios in the systems analysis.

waste generation by 31 and 36%, respectively, compared to the current state. Increasing the reclaiming yield through hydrolysis in the mechanical and chemical scenario reduces virgin material use and solid waste generation by an additional 8 and 9%, respectively. Integrating upcycling in the mechanical, chemical, and upcycling scenario further reduces virgin material use and solid waste generation to 0.5 kg/kg bottle and 0.3 kg/kg bottle, respectively, corresponding to reductions of 56 and 64% compared with the current state. These large reductions result from the diversion of waste fibers from landfilling to produce bottle-grade PET resin. This diversion would result in a negative net solid waste generation if vPET were entirely substituted by upcycled PET, retaining an additional 0.1 kg of material per kg of bottle (see Figure S7 in the SI).

The results from the systems analysis indicate a trade-off between the circularity and environmental metrics of PET bottle production. For instance, it is clear that implementing CE strategies reduces virgin material use and solid waste generation, but improving the environmental performance

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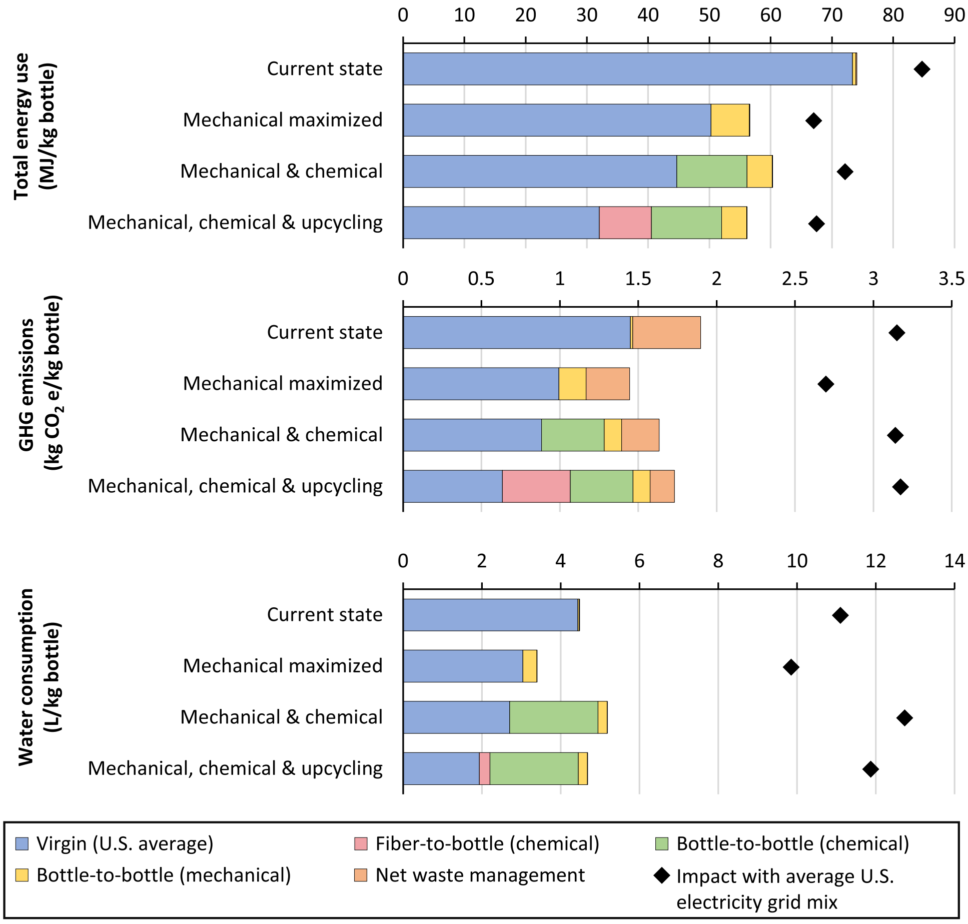

Figure 5. Life-cycle impacts of the different scenarios of the systems analysis with electricity generated only from wind and solar power.

depends on the increased contribution of mechanical recycling in the system, which has been observed in other studies.[12,20] Also, the GHG emissions increase when the percentage of bottles recycled through hydrolysis increases (see Section S7 in the SI). Chaudhari et al.[20] reported similar findings, stating that the incorporation of mechanical recycling into chemical recycling reduces GHG emissions and material circularity compared to only relying on PET dissolution. Furthermore, this study shows that the upcycling of waste materials like PET fibers can improve the circularity and environmental performance of PET bottle production if these materials can substitute to a large extent for the use of virgin PET (see Section S7 in the SI). To establish a sustainable CE, it is therefore necessary to increase the recycling rate of bottles and other waste materials. Moreover, improvement of recycling technologies through increased processing yields and reduced energy requirements should be considered as targets for future research.[12,16,20,40]

Impacts of a Decarbonized Electricity Grid Mix. Hydrolysis is a major driver of GHG emissions in the scenarios where it is included. Because approximately 31% of the GHG emissions of the enzymatic hydrolysis process result from its use of electricity, using electricity sources with zero carbon impact (i.e., increased contribution of renewable energy sources) can reduce the GHG emissions of bottle production implementing CE strategies below current state GHG emissions. To estimate the effects of decarbonization, a sensitivity case substituting the average U.S. electricity grid mix with a grid mix of 50% wind power and 50% solar power was analyzed. The results, displayed in Figure 5, show that GHG emissions from bottle production using a decarbonized grid mix are between 40 and 48% lower for the four production scenarios compared to those using the average U.S. grid mix (shown in Figure 4 and indicated with black markers in Figure 5). In this decarbonized sensitivity case, the scenarios implementing CE strategies have lower total energy use and GHG emissions than the current state. The water consumption is still higher than the current state in the scenarios that include hydrolysis due to the low recycling of process water in this technology. The findings from this alternative analysis show that the decarbonization of electricity reduces the total energy use and GHG emissions of recycling technologies that reduce virgin material use and solid waste generation.

CONCLUSIONS

This study developed a sustainability framework for the simultaneous evaluation of the life-cycle impacts and circularity metrics of the implementation of CE strategies for PET plastic packaging. To the authors' knowledge, this is the first study comparing industrial-scale mechanical recycling, hydrolysis, and methanolysis processes. The results showed a trade-off between the environmental and circularity indicators of the recycling technologies. For example, recycling technologies with lower GHG emissions, like mechanical recycling, increase the reliance on virgin materials due to lower reclaiming yield (67%), while enzymatic hydrolysis increased the reclaiming yield (90%) but generates four times the GHG emissions of mechanical recycling. Indeed, reclaiming operations were the

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ACS Sustainable Chem. Eng. 2023, 11, 514−524

major drivers of GHG emissions in the recycling pathways, so improving their yields and energy efficiency or incorporating the use of renewable energy and bio-derived chemicals can reduce the environmental impact of CE strategies.

The systems analysis approach showed that increasing the recycling rate of PET bottles from 21 to 90% gave rise to a 14% reduction in GHG emissions compared to the current state, while implementing chemical recycling and upcycling, even with the increased recycling rate, did not give rise to a significant variation in GHG emissions. Nevertheless, the joint implementation of these CE strategies reduced virgin material use and solid waste generation by 56 and 64%, respectively. This suggests that although chemical recycling and upcycling do not reduce the life-cycle impacts of the current supply chain, these processes can provide additional benefits from the circularity perspective. Notably, increasing the rate of mechanical recycling markedly reduces the life-cycle impacts of bottle production, so unless chemical recycling and upcycling can be improved to generate lower GHG emissions and water consumption than vPET, use of mechanical recycling should continue. The results of this study also suggest the importance of improving reclaiming yields and optimizing the use of material and energy resources to reduce the life-cycle impacts of implementing CE strategies. Additionally, decarbonization of the electricity grid, which this study demonstrated using an electricity grid mix consisting of solar and wind power, could reduce the GHG emissions of a plastic packaging supply chain implementing chemical recycling and upcycling by 9% relative to the current state.

The developed framework was able to evaluate the implications of implementing CE strategies using a robust but simple approach. The analysis performed through the framework could be expanded by integrating the economic evaluation of the recycling technologies, making it possible to identify CE strategies that are both environmentally beneficial and economically viable. The framework could also be used to evaluate the life-cycle impacts and circularity metrics of other PET products (e.g., focus on the supply chain of polyester fibers), the effect of broadening the scope of this study's supply chain analysis to incorporate different PET products, or the benefits and trade-offs of incorporating recycling technologies into the production of other resins like HDPE, LDPE, and PP.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.2c04626.

> Mechanical and chemical recycling, calculations of the material flow factors for the different recycling technologies, life-cycle inventory data for the different PET production technologies, estimations of net waste management GHG emissions, additional data for the systems analysis approach, description of the multicycle analysis, and results of two additional system analysis scenarios (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Troy R. Hawkins − *Systems Assessment Center, Energy Systems and Infrastructure Analysis Division, Argonne National Laboratory, Lemont, Illinois 60439, United States;* orcid.org/0000-0001-6897-175X; Email: thawkins@anl.gov

Authors

Ulises R. Gracida-Alvarez − *Systems Assessment Center, Energy Systems and Infrastructure Analysis Division, Argonne National Laboratory, Lemont, Illinois 60439, United States;* orcid.org/0000-0002-5061-169X

Hui Xu − *Systems Assessment Center, Energy Systems and Infrastructure Analysis Division, Argonne National Laboratory, Lemont, Illinois 60439, United States;* orcid.org/0000-0003-2994-4892

Pahola Thathiana Benavides − *Systems Assessment Center, Energy Systems and Infrastructure Analysis Division, Argonne National Laboratory, Lemont, Illinois 60439, United States;* orcid.org/0000-0002-8253-7877

Michael Wang − *Systems Assessment Center, Energy Systems and Infrastructure Analysis Division, Argonne National Laboratory, Lemont, Illinois 60439, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acssuschemeng.2c04626

Notes

The authors declare no competing financial interest.

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