ACS Sustainable Chemistry & Engineering

pubs.acs.org/journal/ascecg

Research Article

Designing Value Chains of Plastic and Paper Carrier Bags for a Sustainable and Circular Economy

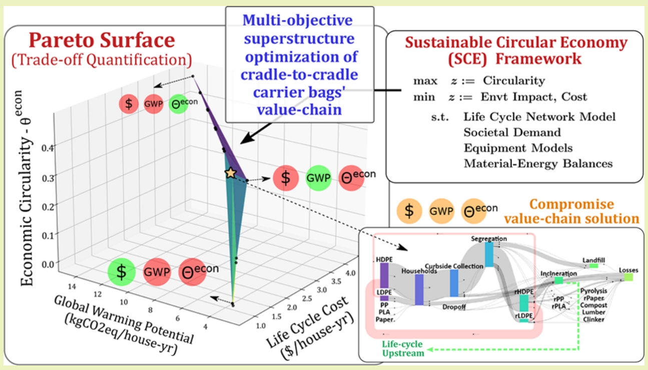
Vyom Thakker and Bhavik R. Bakshi*

Cite This: *ACS Sustainable Chem. Eng.* 2021, 9, 16687−16698

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information

ABSTRACT: Progress toward a sustainable and circular economy (SCE) is crucial to mitigate the large-scale exploitation of natural resources and the pile-up of man-made materials in the environment. Carrier bags form a large component of discarded and mismanaged plastic waste whose value can be retained through a SCE. A novel design framework developed in our previous work, has been applied to design optimal value chains based on a superstructure network containing cradle-to-cradle life cycles of some currently available carrier bag alternatives. Employing the design framework allows systematic exploration of a large number of scenarios with combinations of numerous alternatives, by virtue of using global optimization methods. Life-cycle metrics such as global warming potential and life-cycle cost have been supplemented with a novel general circularity metric that is formulated to measure (a) economic returns and (b) ecological regeneration. These metrics constitute the objectives of the framework, and pareto optimal solutions are found to quantify trade-offs present while designing for these SCE objectives, without assigning arbitrary weights. In doing so, the long-standing paper versus plastic dilemma is quantified using systematic cradle-to-cradle design approaches, and optimal value-chain reforms for SCE are found. The prospects of biobased polylactic acid bags are also studied. We are also able to affirm that advancing technologies for mechanical recycling and policy action for societal-change and value-chain reforms can lead to win−win solutions in the SCE objective domains. Some general conclusions derived from the study are as follows: low-density polyethylene bags (with 10 reuses) perform best in minimizing global warming potential and life-cycle cost, single-use high-density polyethene bags with recycling give the highest economic circularity, whereas paper bags being landfilled lead to the most ecological circularity as they readily compost to return nutrients to the environment. Trade-off solutions are found from pareto surfaces in order to achieve the best possible compromise, and it contains combination of paper and reusable plastic bags. It is observed that increasing substitutability of virgin material with recycled material favors use of biobased polylactic acid bags. Hot spot sectors for atmospheric emissions and circularity are also found for optimal value chains, which can help direct research toward activities of importance.



KEYWORDS: *Sustainable circular economy, Superstructure value-chain optimization, Cradle-to-cradle life-cycle design, Multiobjective optimization, Carrier bags network*

INTRODUCTION

Around 20 million metric tons of plastic is estimated to have entered aquatic ecosystems,[1] and over 100 million metric tons are discarded either to landfills or mismanaged as litter in a single year,[1,2] for example, during 2016. 46% of this plastic waste is generated from the packaging industry[3] including grocery carrier bags, films, containers, and so on. This is motivating the development of approaches for avoiding detrimental effects to marine biodiversity, subsequent food chain, and exploitation of natural resources for virgin plastics, while meeting human needs. The linear nature of today's plastic economy has promoted economic progress, while excessively taxing natural resources and polluting the environment. Creating circular loops for products, components, and materials within the economy is considered to be a promising approach for reducing the load on natural capital and pollution of the environment. This can be achieved by designing value chains for meeting the goals of a sustainable circular economy (SCE).[4,5]

Carrier bags or sacks constitute 25% of all postconsumer plastic waste,[6] and various organizations including grocery stores, local businesses, and governments are often baffled[7,8] by the decision of choosing the right environmentally friendly

Received: August 15, 2021
Revised: November 18, 2021
Published: November 30, 2021

bags. The prevalence of partially disintegrated bags and films in marine ecosystems and habitats has inspired public interest and empathy in solving the plastics problem.[9] Various stores and municipal bodies have therefore imposed bans on single-use plastic, favoring paper bags and other reusable plastic alternatives. However, various life-cycle studies[10,11] have denounced this decision by arguing that the paper life cycle results in a larger environmental impact in terms of air and water quality. These reasons motivate our effort to apply the SCE framework to design the carrier-bags value chain to understand the difference between alternatives and the extent to which they meet the goals of sustainability and circularity, as well as to identify areas needing further work.

Waste management of bags and aggregated household waste is also a long-standing problem that municipalities and governments face.[8,12,13] Various academic, governmental, and institutional studies have looked at this value chain, however from an assessment perspective.[14,15] This has led to solutions and insights riddled with selection biases in scenarios and technological pathway combinations evaluated. For instance, the Boustead report[16] compares recyclable plastic bags with paper and compostable plastic bags for the criteria of life-cycle emissions, resource use, and energy requirements. They find that paper bags (30% recycled fiber) lead to around 2 times the amount of CO_2 equivalent emissions, 3 times the energy usage, and 5 times the waste generated as compared to polyethylene bags. While it considers a few end-of-life scenarios for each of the alternatives, it assumes a certain degree of recycling and discrete pathways (e.g., 13% incinerated and 70% landfill). Khoo et al.[17] compared the cradle-to-gate impact of conventional plastic and biobased carrier bags to find that an advantage in CO_2 emissions appears only when energy was obtained from renewable sources. Civancik-Uslu et al.[18] assessed carrier bags with respect to the marine littering impact using scenario analyses and found plastic has the highest littering impact but the lowest greenhouse gas emissions. Nielsen et al.[19] summarized the need for and the effect of public policy on the use of carrier bags and their effect on the environment, bolstering the need for calculated value-chain reforms. While extremely insightful, these studies are still limited in scope due to the limited number of scenarios that are typically considered in an assessment problem, and do not integrate decision making at different stages of the value chain. Selection of manufacturing technologies seldom considers end-of-life scenarios or consumer behavior and vice versa. For instance, the presence of biodegradable polymers like polylactic acid (PLA) will tremendously affect the recyclability of fossil-based polymers.[20] Thus, the design of segregation and recycling systems has to consider the nature of mixed streams, and polymer manufacturers must consider such end-of-life constraints when designing polymers. Furthermore, evaluating pathways for single objectives may not directly appeal to the diverse stakeholders of a SCE. In our previous work,[21] we have developed a general framework for SCE which uses systems engineering involving optimization over numerous life-cycle pathway solutions, constituting what is called a "superstructure" network of alternatives. This framework allows systematic exploration of a large number of life-cycle alternatives and their infinite combinations using global optimization methods. Formulation of distinct objectives of SCE, followed by multiobjective optimization within the framework permits inclusion of various stakeholder preferences and calculation of trade-offs between them.

In addition to presenting the first realistic application of the SCE framework, another novel contribution of this paper is finding SCE-optimal cradle-to-cradle pathways from various alternatives in the carrier-bags value-chain network utilizing the SCE design framework.[21] The trade-offs between the metrics developed to achieve the SCE goals are also quantified. For sustainability, the metrics used are global warming potential (GWP) and life-cycle cost (LCC), which belong to the environmental and economic domains, respectively. GWP is calculated as the impact caused by greenhouse gas emissions from the network, whereas LCC is calculated as the total cost of natural resources incurred through operation of the network.

We have also developed a novel metric to quantify the circularity within the network using life-cycle flows. This metric is further characterized by the type of restoration "value" or "regenerability" of interest, for example, economic, ecological, exergetic, etc. In this study, we consider two submetrics for circularity, which include (i) economic circularity representing monetary value restoration due to circular flows and (ii) ecological circularity, which represents the ecological or nutrient value restored from the material after the physical transformations it undergoes in its habitat[22] for a year. However, some of the other types of circularity that could be of interest but are not included in this study are social value restoration, exergetic recovery using circular flows, and so on. These circularity metrics along with economic and environmental metrics are used as objectives of the study.

The optimal value chains for carrier bags are found under three different scenarios: business-as-usual, societal-change, and technological-change, which represent the increasing flexibility of value-chain network modules and connections from first to third. These scenarios are used to capture conditions of the technology and society and are different from the scenarios typically used in LCA to select between multiple alternatives. Trade-offs between value chains optimized for different objectives are quantified using multiobjective optimization methods for Pareto front generation. Pareto surfaces containing trade-off or compromise solutions are plotted for all three scenarios. From these pareto surfaces, we observe that the societal-change and technological-advances scenarios allow us to explore regions of the pareto surface that are infeasible under business-as-usual scenario, thereby yielding win−win solutions. This reassuring result can be used to incentivize policy action and additional research toward SCE. We also discover that the business-as-usual scenario is dire, with a maximum possible circularity of 15%. We also acknowledge that the technological-change scenario is very ambitious as it would require advanced technologies for complete substitutability of virgin material by recycled material. Therefore, in this manuscript we focus on providing detailed results for the societal-change scenario.

The optimal value chains for each of the SCE objectives under the societal-change scenario are found and used to derive useful inferences about performance of bag types and end-of-life options on improving SCE objectives. Compromise solutions for the three objectives are found from the pareto surfaces. The trade-off between economic and ecological circularity perspectives is also explored, which quantifies the long-standing paper vs plastic dilemma, prevalent in the literature and mass media. For the optimal value chains, we also find emission and circularity hot spot sectors. We can infer from this study that no single bag type or end-of-life strategy from the currently available options, can achieve both sustainability and circularity. This would require innovations, novel technologies and stringent policy action. Through hot spot analysis, we are able to demonstrate how hotspot analysis on SCE designs can help in this effect. The results of this study are dependent on sources of data and the regions for which they are collected, and are subject to change when better data is made available. For this study, we use global values for littering rates, bag attributes, and end-of-life technologies, as well as the U.S. national average values data sets for the life-cycle inventory.

The rest of this paper is organized as follows. The "Case-Study" section describes the carrier-bags value chain and the problem statement. This is followed by a brief description of our SCE design framework[21] along with its optimization formulation in the 'Methods" section. The "Results" section begins with the transition scenarios of societal and technological change are compared against the baseline scenario to derive insights about possible win−win solutions. Furthermore, we probe the societal-change scenario for detailed results in the form of optimal value chains for each of the SCE objectives are, followed by trade-off exploration using pareto surfaces. Finally, we present the discussion on inferences drawn from the case-study results, especially regarding the paper versus plastic dilemma, enabling SCE through biobased PLA bags and hot spots of the carrier-bags value-chain network.

CASE STUDY

Description. The goal of this case study is to explore the 120 odd alternative pathways in the carrier-bags value chain and their infinite combinations, which form the "super-structure" network of interest. A superstructure is a large space of structural alternatives,[23] each corresponding to a feasible solution pathway. For our study of carrier bags, the superstructure is shown in Figure 1. The manufacturing sector in the network takes in inputs from the life-cycle upstream and can produce the following types of bags:[18]

(i) Single-use, high-density polyethylene (HDPE) bags

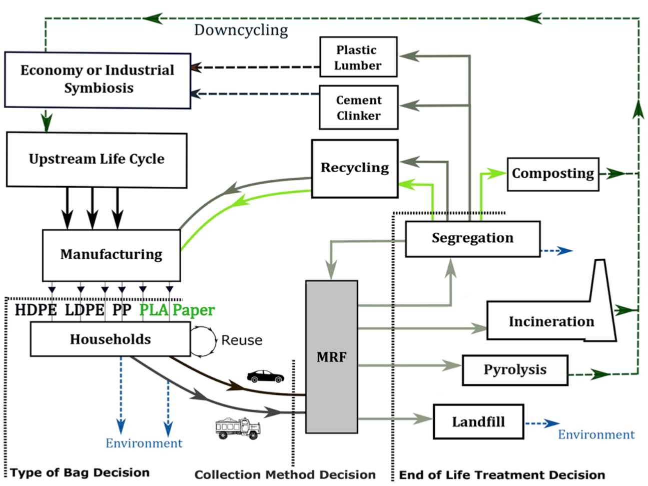


Figure 1. Carrier bags' value-chain superstructure network considered for the case study. Solid arrows indicate cradle-to-gate flows, whereas dashed arrows indicate circular or gate-to-cradle flows. Green and blue arrows represent biodegradable flows and flows to the environment, respectively. Dotted lines indicate decision variables in SCE design.

(ii) Thick, low-density polyethylene (LDPE) bags, reusable 10 times

(iii) Woven polypropylene (PP) bags, reusable 20 times

(iv) Single-use, biobased compostable PLA bags

(v) Single-use, biodegradable paper bags

These bags can be taken up by a single representative household, which disposes its mixed household waste either through curbside collection or drop-off to a center. Therefore, the functional unit (or basis) of this study is the annual volume carrying capacity of a household (4000 L/year).[18] This waste is then transported to a material recovery facility (MRF). At the MRF, the waste can either be segregated and mechanically recycled to substitute virgin plastic/paper, or downcycled to lumber, or cement filler. The mixed waste can also be directly landfilled, incinerated to produce electricity (returning to the grid), or pyrolyzed to fuel. The credits for creating downcycled products and electricity are obtained by displacement of their conventional value chains, assuming substitution at point of source. For instance, electricity from incineration displaces grid electricity, plastic lumber displaces wood-based lumber, and so on. The reason for choosing these technologies for manufacturing, logistics, and end-of-life is partially because they are prominent and mature among conventional technologies and additionally due to data availability restrictions for newer technologies. All these alternative value-chain pathways and linked upstream life cycles form the cradle-to-cradle superstructure network shown in Figure 1. The system boundary is made holistic by including the upstream activities all the way up to extraction of natural resources (see Supporting Information section S.1 for expanded boundary diagram). Notably, the data used to represent the superstructure, including bag attributes, littering behavior, and business-as-usual scenario correspond to global averages, but the life-cycle inventory data is specific to the United States. Therefore, the results are likely to change when more region specific data becomes available. The SCE design framework is used to find optimal pathways from this value-chain superstructure network for multiple objectives of SCE. Furthermore, the trade-off between these objectives is quantified for satisfying stakeholder preference.

Scenarios and Assumptions. To understand opportunities for future advances toward a SCE of carrier bags, we consider three unique scenarios corresponding to advancements of technological systems and societal acceptance of recycling. These scenarios are described in Table 1 along with their respective significance, actions required and associated numerical constraints. The three chosen scenarios are of practical relevance because the baseline scenario explores manufacturing alternatives with constraints in changing MRF and EoL of value chains, whereas the subsequent scenarios investigate the effects of relaxing constraints (i.e., increasing flexibility) of the value chain through societal and technological change.

The assumptions made in this case study are as follows: (i) The network is scaled according to the annual volume carrying requirements of a single representative household in the United States (~4000 L/year).[17,18] (ii) Plastic waste is mixed with organic waste from households which is directed to same end-of-life (EoL) technology. The impact associated with separation of organic waste and EoL is also considered as impact of the value chain. This means that there is no allocation done for organic waste byproduct. (iii) Products

https://doi.org/10.1021/acssuschemeng.1c05562
ACS Sustainable Chem. Eng. 2021, 9, 16687−16698

Table 1. Scenarios Considered for SCE Design of Carrier-Bags Value-Chain Network

scenario name	description	significance	numerical constraints	source
business-as-usual	The current state of collection, where MRF and EoL are imposed as a constraint. The choice between manufacturing options flexible.	baseline scenario used to compare other scenarios against	EoL constraints: 60% landfill, 24% incineration, 12% recycling recyclability caps: 36% for HDPE, LDPE and PP, 10% for PLA, 100% for paper	3
societal-change	Manufacturing, collection, MRF and EoL options are all made flexible, but are under technology constraints of recyclability	requires policy action or behavioral change, and capital investment on MRFs to redirect waste to optimal circular solutions from conventional technology space	same recyclability caps: 36% for HDPE, LDPE and PP, 10% for PLA, 100% for paper	24, 25
technological-change	All options retain a flexibility of choice, and technologies advance to allow larger substitutability of virgin material by recycled.	requires newer technologies, such as extruders and additives to increase permissible recycled content	no recyclability caps: complete substitutability	

with embodied biogenic carbon (e.g., paper and PLA) still contribute to the GWP, because of CO_2 sequestration occurring at different locations and time scales than emissions. This includes biomethane emission from anaerobic digestion of products in landfills. Complying with product carbon footprint and ISO 14 040/44 standards,[26] we also report results without this assumption (i.e., net zero emissions from products with biogenic carbon).

The sensitivity of results to these assumptions has been discussed briefly in the discussion section and more deeply in Supporting Information section S.2, which describes the results without the assumptions (ii) and (iii).

METHODS

Framework for SCE Design. The carrier-bags problem posed in the previous section is solved using the SCE framework,[21] developed to assess and design life-cycle value chains of products with the goal of an SCE. It is based on modifying life-cycle assessment (LCA),[27] a linear algebra model to holistically evaluate the impacts of value chains. LCA has been modified to include alternative value chains to create a superstructure network, which is optimized to find the best possible value chain. Allocation and displacement methods are used along with material-energy balances, consumer utility satisfaction, and other physicochemical governing equations to create the feasible design space. Various life-cycle-based SCE objectives can be used and trade-offs can be quantified using pareto fronts. This framework is general and usable for any product value-chain network, and its features are demonstrated in this study of carrier bags. The design framework can be represented as follows:

$$
\begin{aligned}
\min_{s, a_{im}, a_{nj}} \quad & z := Z(\cdot) \\
\text{s.t.} \quad & As = f && \text{Life-cycle model, Allocation} \\
& g = Bs && \text{Life-cycle model} \\
& Hs \geq u && \text{Utility satisfaction} \\
& \mathcal{F}(\cdot) = 0 && \text{Governing equations} \\
& s \geq 0, f \in \mathbb{R} && (1)
\end{aligned}
$$

Here, z denotes the SCE objectives which can be minimized or maximized using the SCE framework. They are formulated as functions ($Z(\cdot)$) of the decision variables of the superstructure network, and are described further in the next subsection. The technology (A) and intervention (B) matrices represent the value-chain superstructure network of carrier bags shown in Figure 1. Technology matrix A contains all possible flows and connections within the value chain, whereas matrix B has flows to and from the environment. The indices of the technology matrix are i and j, where i belongs to the set of products or flows within the network and j belongs to the set of nodes or modules constituting the network. k is the row index of emission and resource use flows to or from the environment in the intervention matrix B. The amount of physical flow of a product i to (− sign) or from (+ sign) a node j is expressed as $a_{ij}s_j$, where s_j is the scaling factor multiplied to all flows to or from node j. Therefore, the constraint $As = f$ ensures selection and scaling of technologies for flow conservation and to meet a final demand f requirement outside the network. The second constraint uses the same scaling vector, s, to calculate environmental impact and resource use, g. More information about this LCA methdology can be found in the literature.[27,28] For conventional LCA, the A matrix is square and provides a single solution. However, in the SCE design framework, this matrix is likely to be rectangular to capture the presence of multiple alternatives. The utility satisfaction constraint ensures that each household's requirement of carrier bags is met. The governing equations impose material, energy and component balances across value chain modules and sections. These equations also capture physicochemical phenomena of various steps such as the separation efficiency of the segregation unit.

Objectives. The degrees of freedom in the superstructure network are utilized by the SCE optimization framework to find optimal value-chain pathways. The SCE objectives used in this case study consider environmental, economic, and circularity aspects. Life-cycle midpoint indicators can be considered as objectives within the environmental domain. These are computed as a dot product of emission flows and characterization factors (ϕ_k), which convert emission flows in equivalent units of midpoint indicator $\left(M = \sum_k \phi_k^M g_k\right)$. For this case study, we consider Global Warming Potential (GWP) as a relevant indicator of environmental impact. GWP signifies the net CO_2 equivalent of all emissions arising from the operation of the value chain and is therefore an important metric of the value chain. It is expressed as follows:

$$\text{GWP}: = \sum_k \phi_k^{\text{GWP}} g_k \quad \forall\ k \in \text{Emission flows} \tag{2}$$

A viable objective within the economic domain for evaluation of value chains is the LCC. This metric is expected to cover all direct, indirect, and capital costs[29] incurred within the life-cycle of a product with value addition at each stage. However, for this study we only consider the cost of directly and indirectly used natural resources as an estimator of LCC[30] due to scarcity of value addition, capital, and operating cost data for each technology and value-chain activity in the network. The LCC metric used in this work is formulated as follows:

$$\text{LCC}: = \sum_k -c_k g_k \quad \forall\ k \in \text{Natural resources} \tag{3}$$

Parameter c_k denotes the cost per kg of natural resource k or price of technological flow of i per unit flow (e.g., \$/kg for material flow, \$/MJ for energy flow).

Within the circularity domain, we formulate a novel metric θ using life-cycle flows of the network to quantify the circularity of the network. It is calculated as the ratio of the value of circular flows within the system to total value of manufactured products. The value of circular flows can be retained or restored through both technological flows and interventions to the environment. The general expression for θ is as follows:

$$\theta = \frac{\sum_i \sum_e \gamma_i a_{ie} s_e + \sum_k \gamma_k g_k}{\sum_i \sum_m -\gamma_i a_{im} s_m}$$
$$\forall\ e \in \text{EoL processes},\ k \in \text{losses of product } i \text{ to envt.,}$$
$$\text{and } m \in \text{manufacturing processes} \tag{4}$$

In the numerator of the circularity metric (θ), the first term signifies the *value* of material flows retained within the economy or technosphere as down-cycled or up-cycled products. The second term captures the *value* of flows leaving the technosphere to the environment via landfills, litter, emissions, and so on, and are prone to biophysical transformations. The denominator contains the *value* of manufactured products, derived from both virgin and recycled material. The type of value (e.g., monetary, exergetic, etc.) and corresponding units are decided by the parameter γ, which in turn determines the nature of the circularity metric, θ. In this study, we consider two perspectives of circularity: (i) economic circularity (θ^{econ}) and (ii) ecological circularity (θ^{ecol}). Economic circularity (θ^{econ}) is determined as the monetary value regenerated by circularity minus the cost of externalities such as plastic litter, divided by the monetary cost of manufacturing and raw material procurement. In this case study we ignore the cost of externalities ($\gamma_k = 0$) to calculate θ^{econ} from eq 4.

Ecological circularity (θ^{ecol}) is found as the ratio of mass regenerated from physical transformation of end-of-life flows after 1 year in its native environment, to the mass of manufactured product. This metric requires information about whether the products discarded to the landfills, seabeds, or islands provide some value to the ecosystem in which it resides after one year. In general, readily biodegradable materials such as paper provide value to the environment, while nonbiodegradable materials do not provide any value. Ecological value of technological flows is assumed to be 1 kg/kg of material EoL ($\gamma_i = 1$) regardless of whether its upcycled or downcycled, whereas for the flows to the environment, γ_k is the fraction of EoL material which gets regenerated or decomposed in the environment it resides for 1 year after disposal (lets say η_k^{env}). Employing a simple mass balance (equation S.4) to relate technological flows to environmental flows, the final expression of ecological circularity θ^{ecol} is found as the following.

$$\theta^{\text{ecol}}(\cdot) = 1 - \frac{\sum_k [1 - \eta_k] g_k}{\sum_i \sum_m -a_{im} s_m}$$
$$\forall\ k \in \text{Losses of material flows},\ m \in \text{Manufacturing} \tag{5}$$

Here, the parameter $\gamma_k = \eta_k^{\text{env}}$ is the fraction (between 0 and 1) of i regenerated or biodegraded under the environment in which it resides for a year. For instance, $\eta_{\text{PLA}}^{\text{l}}$ for PLA bags in a landfill is around 0.6, whereas in marine environments $\eta_{\text{PLA}}^{\text{m}}$ is around 0.8.[31,32] These parameter values are listed in Table S.4. The proposed ecological metric does not capture all the important aspects of plastic pollution to the environment. However, since state-of-the-art LCA studies do not include these impacts at all, θ^{ecol} is a step forward.

Each objective function can propagate unsustainable or uneconomic behaviors which are captured in other objectives. For instance, minimizing cost may lead to decisions which are not environmentally sustainable and can lead to global warming. This indicates that a single objective that does not provide perverse results or cause unintended harm for any other SCE requirement is nearly impossible to devise. Thus, the SCE objectives in our study have to be simultaneously optimized using a multiobjective optimization approach, as elaborated in Supporting Information section S.1.4.

RESULTS

The nonlinear optimization problem in eq 1 is solved using the BARON solver[33] in GAMS language.[34] The objectives are formulated using eqs 2−5, and optimal pathways are found from the value-chain superstructure network shown in Figure 1. Figure 3 contains the expression of each of these objectives explained in words and optimal value chains for each of them represented as Sankey flow diagrams. Corresponding values of these objectives are listed in Table 2. Since the SCE design problem has multiple objectives, there are bound to be trade-offs and win−win solutions. These are quantified using pareto optimal points, which represent the best possible "compromise" solutions without bias to any one of the three objective domains: circularity, economic, and environmental. More details about the formulation, parameters in the objectives, ϵ-constraint method[35] for pareto surface generation and solver time requirements are in Supporting Information section S.1.3.

Scenarios. With so much interest in increasing sustainability and circularity of the business-as-usual practices, many advances are likely in the near future. We created three scenarios, as described in the Case-Study section, to quantify these advances, from the business-as-usual scenario to the societal- and technological-change scenarios. The significance of scenarios and actions required for transitions between them are stated in Table 1. The technology-change scenario does not consider the potential innovations in advanced recycling such as depolymerization, solvent-purification, and so on, since their scalability needs to be probed before inclusion in the network. Under each scenario condition, we optimize the superstructure network for the three objectives: θ^{econ}, GWP, and LCC. Following this, we construct pareto surfaces in the three-dimensional objective space to quantify the trade-offs between

https://doi.org/10.1021/acssuschemeng.1c05562
ACS Sustainable Chem. Eng. 2021, 9, 16687−16698

Table 2. Values of Objectives for Various Optimal and Pareto Optimal Solutions[a]

value chain	GWP ($kgCO_{2eq}$/house-year)	LCC ($/house-year)	θ^{ecol} (0, 1)	θ^{econ} (0, ∞)
		Edge Points		
min LCC (Figure 3a)	2.96	0.786[a]	0.005	0.000
min GWP (Figure 3b)	2.75[a]	1.18	0.600	0.396
max θ^{econ} (Figure 3c)	14.52	4.42	0.608	0.474[a]
max θ^{ecol} (Figure 3d)	97.56	12.66	0.968[a]	0.000
		Trade-Off Solutions		
θ^{econ}–GWP–LCC (Figure 4a)	5.34	1.51	0.602	0.449
θ^{ecol}–GWP–LCC (Figure 4b)	21.67	3.77	0.903	0.000
θ^{econ}–θ^{ecol} (Figure 5)	39.66	6.39	0.940	0.412
		Scenarios of Operation (max θ^{econ})		
business-as-usual	20.55	5.66		0.141[a]
base-case	14.52	4.42		0.474[a]
flexible-design	15.73	9.50		0.633[a]

[a]Optimal value for objective column.

these objectives. The optimal solutions corresponding to the three objectives form the edge points, with economic circularity (θ^{econ}) on the z-axis. This exercise of multiobjective optimization yields a "pareto surface" which contains all the "compromise" or trade-off solutions. All the points lying under this surface are suboptimal solutions and above it are infeasible.

Pareto surfaces for the three scenarios are shown in Figure 2 and dominance of scenarios over one another is probed. Pareto dominance of a point 'P_1' over another point 'P_2' in the objective space (plotted in Figure 2) indicates that all the values of objective functions are better for P_1 than P_2, thereby leading to a win–win in all objectives while transitioning from P_1 to P_2. It can be seen that the *societal-change scenario* design pareto set (blue surface) presents "win–win" or pareto dominant solutions over the *business-as-usual scenario* (red surface), as its pareto surface lies in a region which was infeasible under the business-as-usual scenario. Similarly, the *technological-change scenario* presents win–win over the other two scenarios. This indicates that there exists huge scope of improvement in all the three domains of SCE for the carrier-bags case study, which can be exploited by the technological- and societal-change scenarios, facilitated by value-chain reforms and flexible designs. For instance, increasing the recyclability of biobased PLA bags can increase circularity tremendously with relatively little compromise in the other two objective domains. Additionally, current practices of landfilling and incineration from the business-as-usual scenario are proved to be suboptimal in the societal-change scenario designs. These outcomes motivate research toward creating more efficient end-of-life technologies such as advanced recycling and provides incentives for value-chain disruption using policy intervention.

This scenario-based design approach can be made more rigorous by performing a systematic sensitivity analysis on the

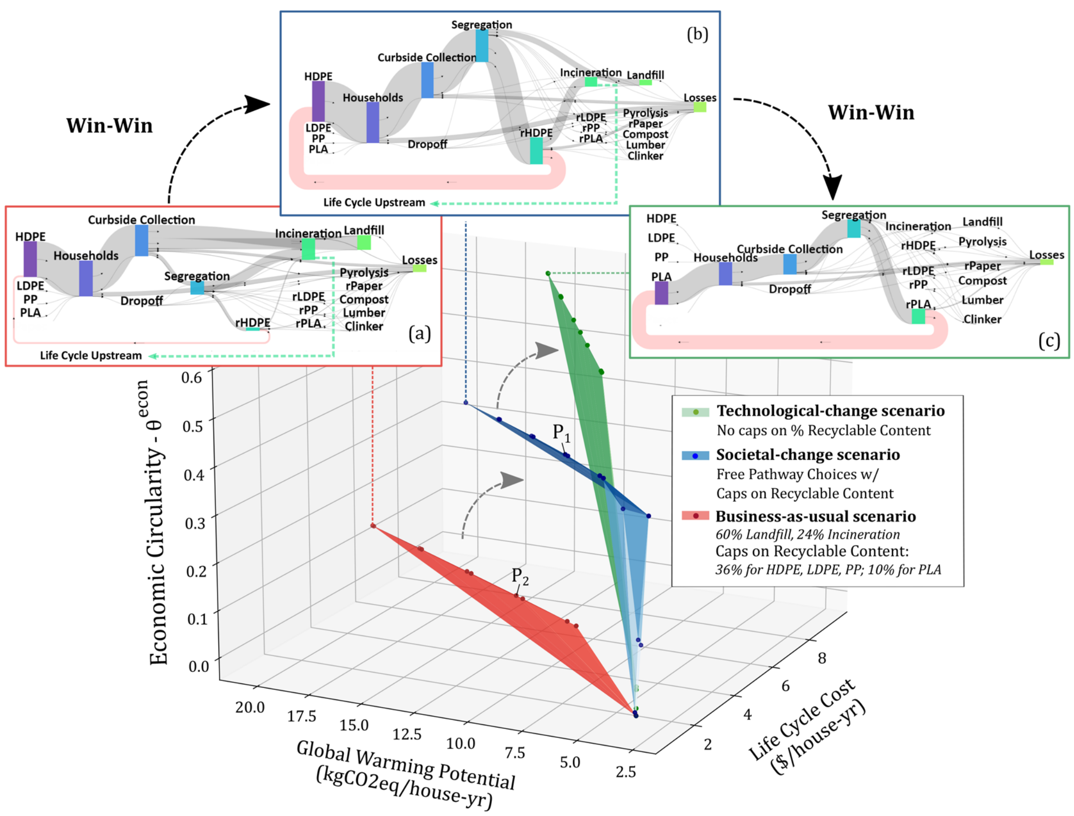


Figure 2. Societal-change and technological-advances scenarios offer win–win solutions over the business-as-usual scenario. Inset: Material flow (or Sankey) diagrams for the optimal circularity value chains under (a) Business-as-usual, (b) Societal-change, and (c) Technological-change scenarios.

https://doi.org/10.1021/acssuschemeng.1c05562
ACS Sustainable Chem. Eng. 2021, 9, 16687–16698

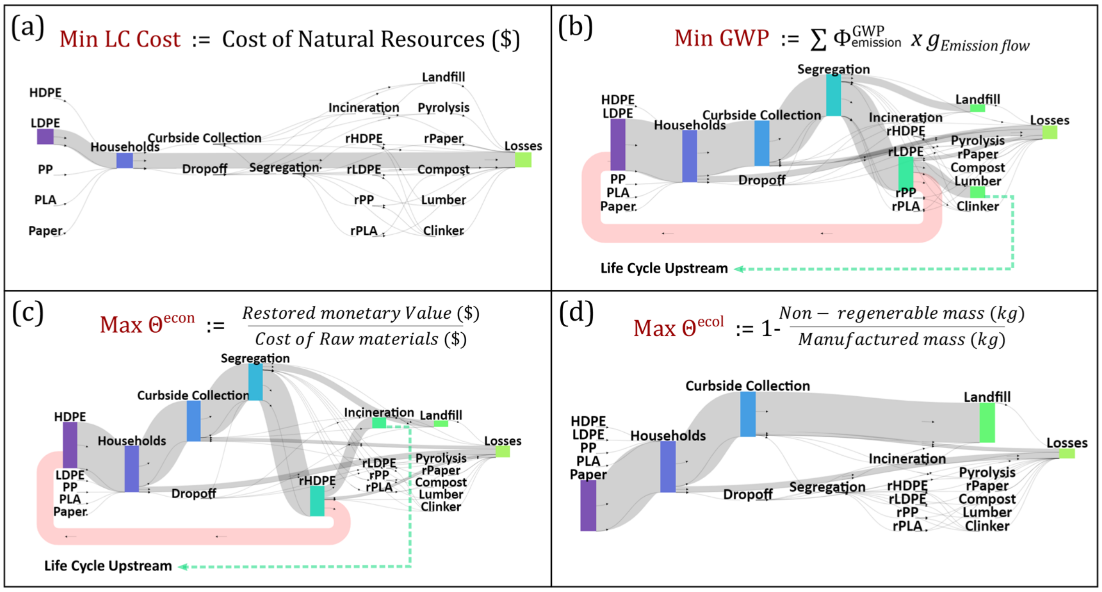


Figure 3. Optimal value chains depicted as Sankey diagrams for each of the enclosed objectives, namely, (a) LCC, (b) GWP, (c) economic circularity (θ^{econ}), and (d) ecological circularity (θ^{econ}). Cradle-to-grave flows are denoted by gray lines with thickness proportional to mass of flowing streams. Red thick lines correspond to recycled flows, and green dotted arrows represent downcycled flows returning to the upstream life-cycle processes.

parameters of the superstructure network to ascertain sectors that significantly influence the pareto surface. Although this effort will be undertaken in future work, in this paper we have conducted hot spot analyses to detect the life-cycle activities which contribute the most to GWP or reduction in circularity, as highlighted in the discussion section.

SCE Design within Societal-Change Scenario. Despite the superior metrics in the technological-change scenario, it may be difficult to achieve, since it requires significant change in technological systems to enhance substitutability of virgin material. However, the business-as-usual scenario is very dire and can lead to only 15% circularity (θ^{econ}) due to limitations at the MRF. The societal-change scenario is a "middle of the road" solution that is most likely to be practically feasible in the near future. Therefore, this scenario is emphasized in the rest of this paper.

Optimal Value-Chains for SCE Objectives. The results obtained by applying the SCE design framework for the societal-change scenario are presented in Figure 3. The value chain which minimizes LCC (Figure 3a) involves the households using reusable low-density polyethylene bags (10 uses) and disposing them to the environment as litter after use. This eliminates the economic burdens of collection and processing of waste, but it is the lowest in terms of circularity and reasonably high in terms of environmental impact in the form of GWP.

The value chain that minimizes GWP (Figure 3b) also contains reusable LDPE bags, but instead of littering, they are collected on the curbside as mixed household waste, which is segregated and mechanically recycled to generate rLDPE resin and plastic lumber. Separated organic waste is composted. Losses from collection enter the environment and waste from the material recovery facility is landfilled. LDPE is selected due to lower net impact owing to its reusability and displacement of wood-based lumber by plastic lumber. However, this value chain has lower circularity because of the nonregenerable waste entering the environment and low monetary value of rLDPE and lumber, as compared to virgin LDPE.

In terms of the life-cycle impacts of GWP and net resource use, reusable LDPE carrier bags outperform all the other alternatives including woven PP bags, biobased PLA bags, and paper bags. These observations are consistent with various recent analyses on paper and plastic products in a recycling context.[10,16,18] However, establishing a SCE requires drastic measures and disruptions within the operating value chain, by considering not only the environmental impact and cost but also circularity. Corona et al.[5] resonate with this opinion and state that CE need not be inherently sustainable, and circularity metrics must be modified to avoid burden shifting and to consider net environmental impact.

The value chain corresponding to the maximum ecological circularity suggests single-use paper bags, as shown in Figure 3d, to be disposed with mixed household waste and sent to the landfill. This is because paper is estimated to return some value to the ecosystem in which it resides as fertilizer, whether in landfill or in the environment. Even though this is not the most lucrative way to restore monetary value to the SCE it still manages to avoid harm to the ecosystems that tends to occur due to nonbiodegradable plastic waste and lock-in of valuable biological and technological nutrients.

The economic circularity (θ^{econ}) metric contrasts the ecological regeneration metric (θ^{ecol}), by crediting the monetary value of circular flows rather than discrediting nutrient pile-up. The economic circularity optimum, therefore chooses single-use HDPE bags, with curbside collection, segregation, mechanical recycling, and the remnant waste plastic being incinerated to generate electricity (Figure 3c). This inadvertently leads to some losses and landfilling of HDPE which remains unusable, or nonregenerable, in any form for a year or even decades. These negative impacts are not being monetized and therefore are not included in the metric. Under this societal-change scenario, maximum circularity optimal value chains, while aligning with goals of

https://doi.org/10.1021/acssuschemeng.1c05562
ACS Sustainable Chem. Eng. 2021, 9, 16687−16698

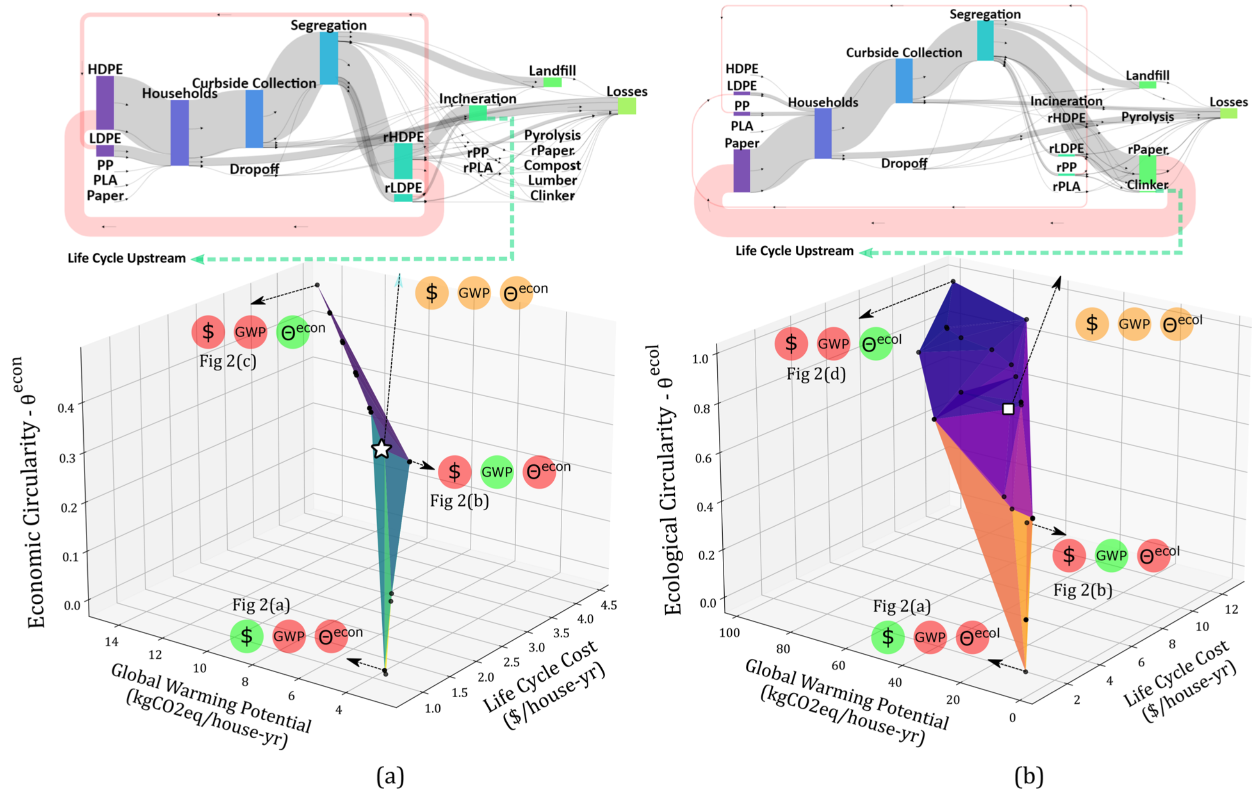


Figure 4. Pareto surfaces for three objective domains; with circularity objectives from (a) economic and (b) ecological perspectives.

CE, are not the most sustainable alternatives. They have suboptimal values of net GWP and LCC, indicating trade-offs among the objectives in the three domains.

Pareto Surfaces and Compromise Solutions. Figure 4 contains the pareto surfaces for the societal-change scenario considering two different perspectives of circularity. Figure 4a quantifies the trade-offs between economic circularity (θ^{econ}), GWP, and LCC, whereas Figure 4(b) pertains to ecological circularity (θ^{ecol}), GWP and LCC. Notably, the pareto surface in Figure 4a is identical to the blue pareto surface corresponding to societal-change scenario in Figure 2. The edge points in Figure 4a correspond to the value chains in Figure 3a−c; the edge points in Figure 4b correspond to Figure 3a,b,d. The trade-offs are evident and represented as red, green, or orange circles annotated to each of the points.

We observe that both the pareto surfaces contain a steep rise in circularity, with marginal increase in impact and cost. This can be attributed to the nature of the superstructure network and the underlying data of technologies and logistical life-cycle modules. Beyond this steep rise, increase in cost or permitted GWP impact, does not yield significant improvement in circularity. This is a good sign for SCE, as one can look at implementing an intermediate or "compromise" solution at the breakpoint of the contour cliffs to capture the trade-offs between objectives. These are annotated as star and square on the pareto surfaces in Figure 4a,b, respectively. The corresponding value chains enclosed within the figure, and the objective values are listed in Table 2.

The compromise solution is indicated by the star symbol in Figure 4a, and includes a combination of reusable LDPE and woven PP bags. These bags are disposed as the mixed household waste, which is collected curbside and segregated at the MRF, and mechanically recycled to maximum capacity. The remaining plastic waste is diverted to cement clinker and lumber production. The value chain corresponding to the intermediate point in Figure 4b (labeled as a square) includes a combination of paper and reusable LDPE and woven PP bags. These bags are also disposed as mixed household waste, which is collected curbside, segregated at the MRF, and mechanically recycled to maximum capacity. The remaining plastic waste is diverted to a cement clinker plant, and the biodegradable waste is landfilled.

DISCUSSION

Paper versus Plastic. Results stated in the above sections employ multiobjective optimization to systematically evaluate paper and plastic carrier-bags value chains, thereby quantifying the long-standing paper−plastic dilemma.[16,18,36] One key inference is that paper bags are not an attractive option to reduce life cycle environmental impact. This is with the assumption that biogenic carbon dioxide sequestration due to wood cultivation must be excluded from calculation of GWP (assumption (ii) from the "Case-Study" section). This assumption is commonly made because of varied opinions about biogenic carbon,[11,37] partly because of the difference in temporal scales of paper use, growth of trees, and seasonality of CO_2 capture. We do find the results without making the assumptions for the societal-change scenario in the case study and provide them separately in Supporting Information section S.2.2. This is compliant with the ISO 14 040/44[38] standards of accounting for biogenic carbon in LCA. In that case, the lowest GWP optimal value chain involves single-use paper bags being disposed to a landfill. In contrast, LDPE bags with 5−10 reuses, followed by recycling and downcycling to lumber, is the least GWP alternative with the assumption of getting credits for biogenic carbon sequestration.

https://doi.org/10.1021/acssuschemeng.1c05562
ACS Sustainable Chem. Eng. 2021, 9, 16687−16698

If circularity has to be considered in designing a new value chain for carrier bags, then the choice between paper and plastic is not very obvious. If one wants to minimize nutrient pile-up and harm to marine ecosystems (θ^{ecol}), then paper bags should be chosen. Plastic bags promise more economic circularity, with higher monetary value restoration (θ^{econ}). However, the societal damages due to plastic use are excluded from θ^{econ} and can be included as cost of externalities in γ_k when data becomes available. As evident from the trade-off in Table 2, the former value chain (max θ^{ecol}) performs poorly in monetary restoration (θ^{econ}), whereas the latter with plastic bags and recycling leads to excessive nonregenerable litter. This trade-off is quantified in the pareto curve plotted in Figure 5, and the compromise solution containing an optimal ratio of paper and plastic bags is suggested (highlighted circle). This compromise solution also has a reasonable GWP and LCC (Table 2).

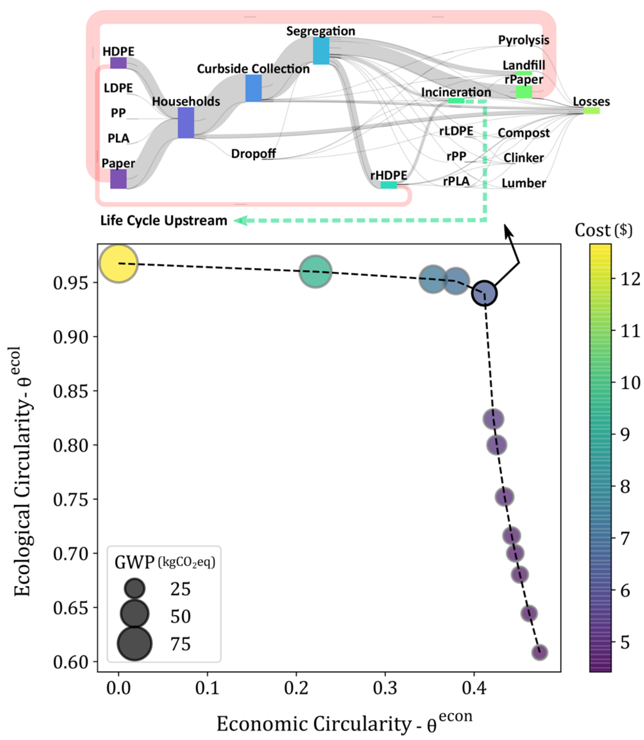
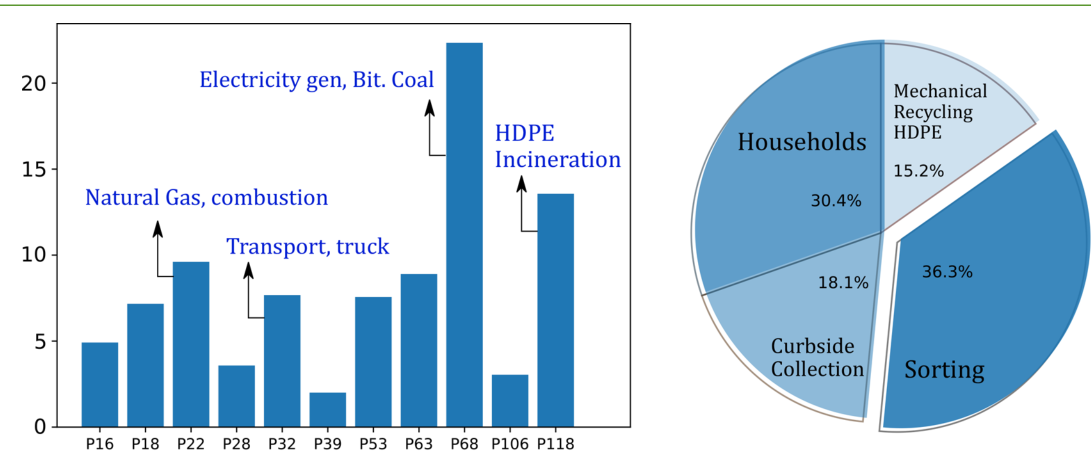


Figure 5. Pareto curve describing the trade-offs between different perspectives of circularity and compromise value chains.

We are therefore able to emphasize the utility of these novel circularity metrics based on product life cycles, which can capture contrasting perspectives of circularity for any value-chain network. However, we suggest inclusion of both these metrics using multiobjective methods such as pareto front development as done in Figure 5. Alternatively, this could also be achieved by employing weighted objective algorithms like analytic hierarchy process (AHP) and multiobjective particle swarm optimization (MOPSO)[39] and to carefully aggregate these perspectives according to stakeholder preference. As demonstrated in this study, the two metrics capture trade-off between ecological and economic views to solve practical challenges in reducing plastic litter for circularity. Unfortunately, in the current scenario, economic burden of circularity (θ^{econ}) often outweighs the urgency of plastic waste accumulation in the environment (θ^{ecol}). Using such metrics and methods can help systematically tackle this issue.

Case for Biobased Bags. Figure 2 shows that the circularity optimal for the technological-change scenario involves the use of biobased PLA bags, followed by segregation and mechanical recycling. In the business-as-usual and societal-change scenarios, this value chain is not chosen because of the low substitutability of virgin PLA by recycled PLA (~10% as compared to 36% for PE).[25,40] Enhancing the recycling technologies for these bags specifically and promoting their value chain can prove to be a viable SCE alternative in the future. This is evident from Figure 2 in which PLA-based value chains with enhanced recycling offer win−win solutions over the business-as-usual and base case designs. The lower GWP of these value chains is because of the inherent benefits of using corn and its byproducts (e.g., stover) instead of fossil-based resources for the plastics. They also perform well in terms of circularity, as recycled resin has good monetary value (θ^{econ}) and the losses of PLA to land-based and marine ecosystems can partially regenerate circularity (θ^{ecol}) and provide some ecological value as compost.

Hot Spots in Sustainability and Circularity. It can be very useful to identify nodes of the value chain that can be enhanced or transformed to increase the sustainability and circularity metrics the most, at the lowest expense. Our



(a) GWP Contribution (%) (b) Loss to Envt. Contribution (%)

Figure 6. Hot spots for θ^{econ} optimal value chain (Figure 3c); based on (a) GWP and (b) circularity.

https://doi.org/10.1021/acssuschemeng.1c05562
ACS Sustainable Chem. Eng. 2021, 9, 16687−16698

framework employs simple linear algebra and life-cycle allocation-based techniques to find such hot spots in the value chain. This method simply ranks contributions of various sectors to GWP and loss of ecological circularity (θ^{ecol}), thereby identifying hot spots in the value chain. Details are elaborated in section S.1.3. Figure 6 depicts the hot spots in GWP and θ^{ecol} for the value chain corresponding to maximum θ^{econ} (Figure 3c). It is evident from Figure 6a that for the optimal θ^{econ} value chain the largest emissions contributing to GWP come from coal-based electricity production at power plant, followed by incineration of nonrecyclable HDPE. In addition, from Figure 6b we see that for the same optimum value chain, the biggest circularity hot spot occurs in the sorting stage, followed by household losses. This leads us to the inference that working on sorting techniques and incentivizing good human behavior can enhance circularity the most, whereas GWP can be reduced by shifting to cleaner energy sources and shifting from incineration of residual nonrecyclable HDPE to plastic lumber production. This exercise of finding hot spots can identify sensitive and vulnerable sectors of the value chain, thereby guiding innovations, future research, industrial symbiosis agendas, and new policies for broader SCE goals.

CONCLUSIONS

In this paper, we focused on developing a sustainable and circular economy of plastic and paper carrier bags by using our previously developed design framework.[21] We are able to achieve the following goals to advance to a SCE for the carrier-bags value chain: (a) the design of value chains with an integrated holistic system boundary; (b) the consideration of various objectives of SCE through careful evaluation of multiple stakeholders; and (c) the ability to find critical sectors which could benefit from innovation and intervention efforts.

For this SCE design, we construct a "superstructure" network for the carrier-bags value chain which comprises of all the alternative pathways. This includes options in the manufacturing, collection and end-of-life steps. We explore the superstructure network under three scenarios: business-as-usual with fixed MRF operation, societal-change with flexible policy action and social change to modify EoL strategies, and technological-change with advances in technologies to allow complete substitution of virgin material by recycled content.

The nodes in the superstructure network are connected to one another and upstream processes through life-cycle models for flow conservation and technology scaling. Governing equations are also setup for ensuring material-energy balances, imposing scenario conditions, and modeling physicochemical transformations at various nodes. These models constitute the constraints in the SCE framework[21] which define the feasible structural alternatives of the superstructure network. Nonlinear optimization is used to find optimal carrier-bags value chains for environmental, economic, and circularity objectives.

Our results convey that policy action and value chain reforms can enhance all the objectives of the value chain simultaneously, as compared to the business-as-usual scenario. If this is aided by technological advances, such as larger recycled content in bags, then further betterment in the objectives can be attained through win−win solutions. This should motivate not only governmental bodies but also industrial practitioners and academicians to strive toward societal change and better technologies to reap benefits in all three objective domains. The societal-change scenario seems to be easier to realize than the technological-change scenario. For this scenario, the smallest carbon footprint is provided by the reusable LDPE bags with around 35% recycled content, while the smallest life-cycle cost of natural resources use optimal cost involves reusable LDPE bags being directly littered to the environment to avoid economic burdens of collection and processing. Circularity purely from an economic standpoint prefers the use of single-use HDPE bags with 35% recycled content and incineration of residual plastic, whereas maximizing ecological circularity involves use of paper bags and landfilling. Evidently, there are vast trade-offs between these objectives and choosing a single optimal value chain is a complex question relying heavily on stakeholder preference. Our framework provides pareto curves and surfaces to identify combinations of various carrier bags and waste management techniques that can appease most stakeholders without large compromises.

We also find that there is no single optimal bag type or EoL strategy from the currently available alternatives that can satisfy all SCE objectives: Trade-offs are unavoidable. These can only be reduced by societal and behavioral change, novel technologies, and innovations such as advanced recycling which can lead to win−win solutions and exploration of the infeasible region of the pareto surface. In this work, we demonstrate how hot spot analysis can be used to identify the sectors within the value chain which possess the largest scope of betterment of objectives through innovations, thereby guiding future research directions.

In future work, we will evaluate the uncertainty in life-cycle data sets due to variation in data for different regions, technologies, and policy and probe its impact on pareto optimal designs. We will also apply sensitivity analyses to the multiobjective optimization problem to find most effective ways to narrow the pareto surface and probe the effects of parameter values on optimal solutions. More work is also needed to improve the ecological circularity metric by including the impact of plastic pollution by more sophisticated quantifiers than mass and utilize models corresponding to ecosystem management and value generation. In addition, we may also extend the design problem to include consequential life cycle and hybrid economic models[41] to consider effects of marginal changes in supplies and demands of reagents and chemical raw materials, thereby allowing systematic ways to model transitions to a sustainable and circular economy.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssuschemeng.1c05562.

> Data sources, model formulations, life-cycle goal, and scope definition, solver times, and results exploring the effects of case-study assumptions (PDF)

AUTHOR INFORMATION

Corresponding Author

Bhavik R. Bakshi − *William G. Lowrie Department of Chemical and Biomolecular Engineering, The Ohio State University, Columbus, Ohio 43210, United States;* orcid.org/0000-0002-6604-8408; Email: bakshi.2@osu.edu

https://doi.org/10.1021/acssuschemeng.1c05562
ACS Sustainable Chem. Eng. 2021, 9, 16687−16698

Author

Vyom Thakker − *William G. Lowrie Department of Chemical and Biomolecular Engineering, The Ohio State University, Columbus, Ohio 43210, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acssuschemeng.1c05562

Notes

Sources of information for bag attributes,[18] consumption, and mismanagement[3] are from the literature, whereas upstream life cycle information is obtained from US LCI commons[42] and ecoinvent v2.2 data sets.[43] The end-of-life process information is obtained from literature, techno-economic assessments, and US EPA WARM data set.[44] The cost and price information is obtained from U.S. BEA and other industrial data sets. The code used in this case study is provided as a Mendeley Data set.[45] The life-cycle inventory data for the superstructure network is also made available in the data set as an excel workbook (CarrierBags_LCI_Data.xlsx).

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

Partial funding for this work was provided by the Global Kaiteki Center at Arizona State University, the National Science Foundation through grant EFMA-2029 397, and the Richard M. Morrow professorship fund.

■ REFERENCES

(1) Borrelle, S. B.; Ringma, J.; Law, K. L.; Monnahan, C. C.; Lebreton, L.; McGivern, A.; Murphy, E.; Jambeck, J.; Leonard, G. H.; Hilleary, M. A.; et al. Predicted growth in plastic waste exceeds efforts to mitigate plastic pollution. *Science* **2020**, *369*, 1515−1518.

(2) Geyer, R.; Jambeck, J. R.; Law, K. L. Production, use, and fate of all plastics ever made. *Sci. Adv.* **2017**, *3*, No. e1700782.

(3) The New Plastics Economy - Catalysing Action; Technical Report; Ellen MacArthur Foundation, 2017. https://ellenmacarthurfoundation.org/the-new-plastics-economy-catalysing-action (Accessed on 11/16/2021).

(4) Korhonen, J.; Honkasalo, A.; Seppälä, J. Circular economy: the concept and its limitations. *Ecol. Econ.* **2018**, *143*, 37−46.

(5) Corona, B.; Shen, L.; Reike, D.; Rosales Carreón, J.; Worrell, E. Towards sustainable development through the circular economy-A review and critical assessment on current circularity metrics. *Resour. Conserv. Recy.* **2019**, *151*, 104498.

(6) *Advancing Sustainable Materials Management: Facts and Figures*; Technical Report; U.S. Environmental Protection Agency, 2017.

(7) Ellen, G. An Inconvenient Bag. *The Wall Street Journal*, 2008. https://www.wsj.com/articles/SB122238422541876879 (Accessed on 11/16/2021).

(8) Wagner, T. P. Reducing single-use plastic shopping bags in the USA. *Waste Manage.* **2017**, *70*, 3−12.

(9) Jambeck, J. R.; Geyer, R.; Wilcox, C.; Siegler, T. R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K. L. Plastic waste inputs from land into the ocean. *Science* **2015**, *347*, 768−770.

(10) Van Ewijk, S.; Stegemann, J. A.; Ekins, P. Limited climate benefits of global recycling of pulp and paper. *Nat. Sustain.* **2021**, *4*, 180.

(11) Sun, M.; Wang, Y.; Shi, L.; Klemeš, J. J. Uncovering energy use, carbon emissions and environmental burdens of pulp and paper industry: A systematic review and meta-analysis. *Renewable Sustainable Energy Rev.* **2018**, *92*, 823−833.

(12) Singh, J.; Cooper, T. Towards a sustainable business model for plastic shopping bag management in Sweden. *Procedia CIRP* **2017**, *61*, 679−684.

(13) Ahamed, A.; Vallam, P.; Iyer, N. S.; Veksha, A.; Bobacka, J.; Lisak, G. Life cycle assessment of plastic grocery bags and their alternatives in cities with confined waste management structure: A Singapore case study. *J. Cleaner Prod.* **2021**, *278*, 123956.

(14) Muthu, S. S.; Li, Y. *Assessment of Environmental Impact by Grocery Shopping Bags*; EcoProduction Series; Springer, 2014.

(15) Kimmel, R. M.; Cooksey, K. D.; Littman, A. *Life Cycle Assessment of Grocery Bags in Common Use in the United States*; Clemson Univ. Dig. Press, Envt. Studies, 2014.

(16) Chaffee, C.; Yaros, B. R. *Life Cycle Assessment for Three Types of Grocery Bags*; Boustead Consulting& Associates Ltd., 2007.

(17) Khoo, H. H.; Tan, R. B.; Chng, K. W. Environmental impacts of conventional plastic and bio-based carrier bags. *Int. J. Life Cycle Assess.* **2010**, *15*, 284−293.

(18) Civancik-Uslu, D.; Puig, R.; Hauschild, M.; Fullana-i-Palmer, P. Life cycle assessment of carrier bags and development of a littering indicator. *Sci. Total Environ.* **2019**, *685*, 621−630.

(19) Nielsen, T. D.; Holmberg, K.; Stripple, J. Need a bag? A review of public policies on plastic carrier bags-Where, how and to what effect? *Waste Manage.* **2019**, *87*, 428−440.

(20) Gere, D.; Czigany, T. Future trends of plastic bottle recycling: Compatibilization of PET and PLA. *Polym. Test.* **2020**, *81*, 106160.

(21) Thakker, V.; Bakshi, B. R. Toward sustainable circular economies: A computational framework for assessment and design. *J. Cleaner Prod.* **2021**, *295*, 126353.

(22) Morseletto, P. Restorative and regenerative: Exploring the concepts in the circular economy. *J. Ind. Ecol.* **2020**, *24*, 763−773.

(23) Mencarelli, L.; Chen, Q.; Pagot, A.; Grossmann, I. E. A review on superstructure optimization approaches in process system engineering. *Comput. Chem. Eng.* **2020**, *136*, 106808.

(24) *National Postconsumer Plastic Bag & Film Recycling Report*; American Chemistry Council, 2014.

(25) Hopewell, J.; Dvorak, R.; Kosior, E. Plastics recycling: challenges and opportunities. *Philos. Trans. R. Soc., B* **2009**, *364*, 2115−2126.

(26) Garcia, R.; Alvarenga, R. A.; Huysveld, S.; Dewulf, J.; Allacker, K. Accounting for biogenic carbon and end-of-life allocation in life cycle assessment of multi-output wood cascade systems. *J. Cleaner Prod.* **2020**, *275*, 122795.

(27) Suh, S.; Huppes, G. Methods for life cycle inventory of a product. *J. Cleaner Prod.* **2005**, *13*, 687.

(28) Bakshi, B. R. *Sustainable Engineering: Principles and Practice*; Cambridge University Press, 2019.

(29) França, W. T.; Barros, M. V.; Salvador, R.; de Francisco, A. C.; Moreira, M. T.; Piekarski, C. M. Integrating life cycle assessment and life cycle cost: a review of environmental-economic studies. *Int. J. Life Cycle Assess.* **2021**, *26*, 244−274.

(30) Berger, M.; Sonderegger, T.; Alvarenga, R.; Bach, V.; Cimprich, A.; Dewulf, J.; Frischknecht, R.; Guinée, J.; Helbig, C.; Huppertz, T.; et al. Mineral resources in life cycle impact assessment: part II-recommendations on application-dependent use of existing methods and on future method development needs. *Int. J. Life Cycle Assess.* **2020**, *25*, 798−813.

(31) Chamas, A.; Moon, H.; Zheng, J.; Qiu, Y.; Tabassum, T.; Jang, J. H.; Abu-Omar, M.; Scott, S. L.; Suh, S. Degradation rates of plastics in the environment. *ACS Sustainable Chem. Eng.* **2020**, *8*, 3494−3511.

(32) Ho, K.-L. G.; Pometto, A. L.; Hinz, P. N. Effects of temperature and relative humidity on polylactic acid plastic degradation. *J. Polym. Environ.* **1999**, *7*, 83−92.

(33) Sahinidis, N. V. BARON: A general purpose global optimization software package. *Journal of global optimization* **1996**, *8*, 201−205.

(34) Bussieck, M. R.; Meeraus, A. General algebraic modeling system (GAMS). In *Modeling Languages in Mathematical Optimization*; Springer, 2004; pp 137−157.

(35) Mavrotas, G. Effective implementation of the e-constraint method in Multi-Objective Mathematical Programming problems. *Appl. Math. Comput.* **2009**, *213*, 455−465.

(36) Plummer, B. Plastic Bags, or Paper? Here's What to Consider When You Hit the Grocery Store. *The New York Times*, 2019. https://

www.nytimes.com/2019/03/29/climate/plastic-paper-shopping-bags.html (Accessed on 11/16/2021).
(37) Wiloso, E.; Heijungs, R.; Huppes, G.; Fang, K. Effect of biogenic carbon inventory on the life cycle assessment of bioenergy: challenges to the neutrality assumption. *J. Cleaner Prod.* **2016**, *125*, 78−85.
(38) Finkbeiner, M.; Tan, R.; Reginald, M. *Life Cycle Assessment (ISO 14040/44) as Basis for Environmental Declarations and Carbon Footprint of Products*; ISO Technical Committee 207 Workshop, Norway, 2011.
(39) Ridha, H. M.; Gomes, C.; Hizam, H.; Ahmadipour, M.; Heidari, A. A.; Chen, H. Multi-objective optimization and multi-criteria decision-making methods for optimal design of standalone photovoltaic system: A comprehensive review. *Renewable Sustainable Energy Rev.* **2021**, *135*, 110202.
(40) Cornell, D. D. Biopolymers in the existing postconsumer plastics recycling stream. *J. Polym. Environ.* **2007**, *15*, 295−299.
(41) Thakker, V.; Bakshi, B. R. Multi-scale Sustainable Engineering: Integrated Design of Reaction Networks, Life Cycles, and Economic Sectors. *Comput. Chem. Eng.* **2022**, *156*, 107578.
(42) *U.S. Life Cycle Inventory Database*; National Renewable Energy Laboratory, 2012.
(43) Frischknecht, R.; Jungbluth, N.; Althaus, H.-J.; Doka, G.; Dones, R.; Heck, T.; Hellweg, S.; Hischier, R.; Nemecek, T.; Rebitzer, G.; Spielmann, M. The ecoinvent database: Overview and methodological framework. *Int. J. Life Cycle Assess.* **2005**, *10*, 3−9.
(44) *Waste Reduction Model (WARM)*, v15; U.S. Environmental Protection Agency, 2009.
(45) Thakker, V.; Bakshi, B. R. Designing Value-chains of Plastic and Paper Carrier Bags for a Sustainable and Circular Economy - Code. *Mendeley Data* **2021**, DOI: 10.17632/6zdsk66b9t.1.