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Life Cycle Assessment for the Design of Chemical Processes, Products, and Supply Chains

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Abstract

Design in the chemical industry increasingly aims not only at economic but also at environmental targets. Environmental targets are usually best quantified using the standardized, holistic method of life cycle assessment (LCA). The resulting life cycle perspective poses a major challenge to chemical engineering design because the design scope is expanded to include process, product, and supply chain. Here, we first provide a brief tutorial highlighting key elements of LCA. Methods to fill data gaps in LCA are discussed, as capturing the full life cycle is data intensive. On this basis, we review recent methods for integrating LCA into the design of chemical processes, products, and supply chains. Whereas adding LCA as a posteriori tool for decision support can be regarded as established, the integration of LCA into the design process is an active field of research. We present recent advances and derive future challenges for LCA-based design.

1. MOTIVATION

Life cycle assessment (LCA): ISO-normed environmental assessment method that considers all life cycle stages and various impact categories

Chemical engineering is one key discipline to approach many of the United Nations' Sustainable Development Goals (1). Sustainability is therefore becoming a main objective for the design of chemical products, processes, and supply chains (2–9). Although the term sustainability implies environmental, economic, and social aspects (10), in this review, we focus on chemical design approaches for minimizing environmental impacts.

Environmental assessment has been recognized as an important part of chemical process design for more than 20 years (11). Various performance indicators and metrics for environmental assessment have been proposed and used in process design (12). Frequently, these metrics assess environmental impacts as part of a hazard assessment on environment, health, and safety, and impact categories include acute toxicity, biodegradability, and ozone depletion potential (13, 14). The focus of environment, health, and safety metrics is often limited by both data availability and the scope of current regulations and is therefore confined to the process of interest itself. Expanding the scope beyond the process is desirable for considering potential trade-offs along the complete life cycle.

Environmental impacts can be quantified along the complete life cycle in a standardized way by life cycle assessment (LCA) (15). As its first holistic feature, LCA considers the entire life cycle of a process or a product (e.g., a chemical or chemical device), starting from the provision of raw materials and energy, via manufacturing and product use, to its recycling or final disposal at the end of life (**Figure 1**). A second holistic feature of LCA is that multiple environmental impacts are evaluated. Through these holistic features, LCA aims to avoid problem shifting between both life cycle stages (e.g., where more efficient production might lead to more harmful waste treatment) and environmental impacts (e.g., where climate-friendlier production might require more toxic materials). As a consequence, LCA enables a balanced assessment of changes across all life cycle stages and impact categories.

Compared with chemical process design, LCA is a relatively young methodology, whose standardization began in the 1990s (16). LCA therefore was not considered in process design for a

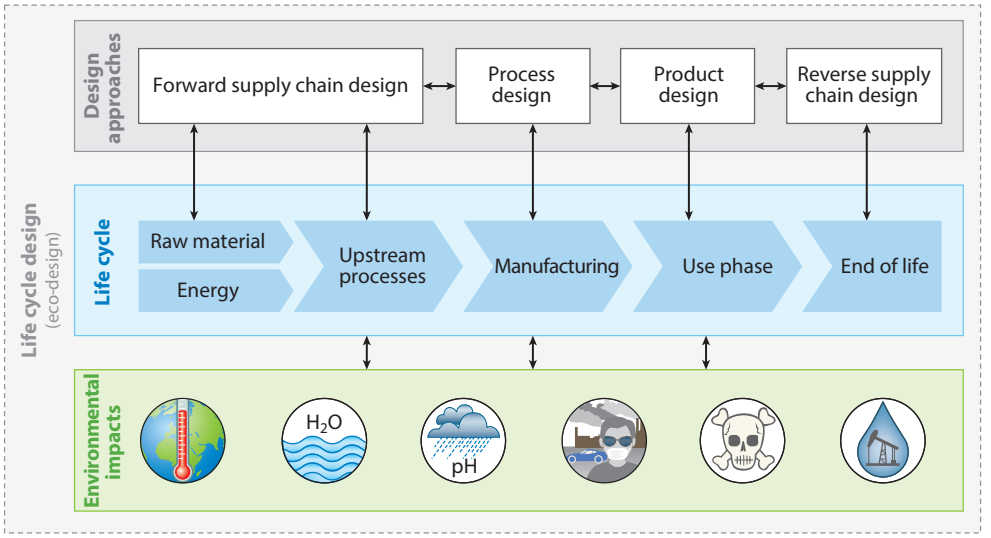

Figure 1

Life cycle design, in which different design approaches cover different parts of the life cycle, while considering multiple environmental impacts.

long time. For example, classical textbooks on conceptual process design focus solely on economic evaluation and do not teach LCA (17, 18). First approaches toward integrating sustainability were proposed using qualitative design principles, such as avoiding toxic by-products and minimizing waste streams (19, 20). Azapagic and colleagues (21, 22) reviewed the first systematic approaches to integrating LCA directly into process design in the late 1990s. Nowadays, it is common practice to formulate environmental impacts as design constraints in economic optimization problems (23). A recent development is to consider environmental impacts as a sole or additional objective in design.

From the life cycle perspective, chemical design approaches should be restricted not to parts but rather to the entire life cycle. In early pioneering work, Grossmann and coworkers (24) already identified that optimizing toxicity for parts of the life cycle may outsource toxicity impacts outside the considered system boundaries. Although expanding the scope beyond the manufacturing process of chemicals toward the entire life cycle has been and remains challenging, a complete life cycle design is highly attractive because it provides the opportunity to exploit newly introduced degrees of freedom (**Figure 1**): Alternative raw materials can be employed, such as biomass or renewable energy; supply chain structure can be redesigned to identify more sustainable suppliers; products can be designed to maximize fitness for purpose; and the end-of-life by chemical waste management can be addressed from the very beginning by designing molecules that degrade faster or are easier to recycle in efficient reverse supply chains.

In this review, we analyze how chemical engineers currently integrate LCA into product and process design and even beyond. Our vision is to exploit all possible degrees of freedom for an environmentally optimal design of complete chemical life cycles by gradually expanding the scope of the design space. For this purpose, we first introduce the fundamentals of LCA (Section 2). We present estimation methods for closing data gaps (Section 3) and begin the literature review with the integration of LCA into process design (Section 4). Here, we define process design as the conceptual engineering of a chemical plant for the process under study, e.g., the choice of equipment and process settings. The design space is then expanded to include the use phase, which is mostly affected by product design (Section 5). Further expansion of the design space covers supply chains (Section 6). In contrast to process design, supply chain design is defined as the design of a network of upstream and downstream processes, considering also the reverse supply chain. Ultimately, we extend the scope of the design space beyond physical supply-and-demand relationships by considering market-mediated effects (Section 7). Due to its holistic ambition, LCA is inherently uncertain, and validation is challenging. These topics are therefore discussed in the concluding Section 8.

2. FUNDAMENTALS OF LIFE CYCLE ASSESSMENT

The LCA methodology has been standardized by the International Organization for Standardization (ISO) for environmental assessment of products along their entire life cycle, including the supply of raw materials, the production process, product use, and final disposal (15). The basic concept of LCA is to collect all mass and energy flows that are exchanged with the environment over the life cycle of a product and to translate the resulting inventory into environmental impacts. Owing to its holistic approach, LCA identifies impact shifting between different stages in the life cycle and between different environmental impacts.

The LCA methodology consists of four phases (15):

1. Goal and scope definition: The first step defines the goal of the study (the "why?" and "what?" questions the study should answer), as well as the scope [what is (not) included?]. The scope definition includes key LCA elements, such as the functional unit (the

quantitative basis of the study), suitable benchmarks in comparative studies, the system boundaries, and the considered environmental impact categories.

2. Inventory analysis: Mass and energy flows are collected for all processes within the system boundaries.
3. Impact assessment: The inventory data collected in phase 2 are translated into environmental impacts.
4. Interpretation: Results are verified by uncertainty analyses, and compliance with the intended goal and scope is checked before conclusions are drawn.

In the following, we briefly discuss key elements of LCA that we believe are particularly important for conducting and integrating LCA into chemical design approaches, including definition of functional unit and benchmarking, selection of environmental impact categories, definition of system boundaries, and contrasting of foreground versus background systems.

2.1. Functional Unit and Benchmarking

The functional unit is the reference basis to which all LCA results are related. The functional unit quantifies the function or service of the system under study. Typically, the function of a chemical process is to provide products. Thus, a common functional unit is simply "1 kg of a chemical."

The definition of the functional unit can become more complex if processes provide more than one product. These so-called multiproduct or multifunctional processes are common in the chemical industry, e.g., in (bio)refineries or water electrolysis producing both hydrogen and oxygen. Although the functional unit can be defined as a bundle of products (e.g., x kg of chemical A and y kg of chemical B), this bundle approach becomes impractical in highly integrated chemical networks with many by-products. Product-specific impacts are therefore often desirable, where overall emissions must be allocated to each product. Allocation rules have been developed, and a hierarchy is specified in the ISO standard. For detailed guidance on allocation rules, interested readers are referred to Reference 25.

Functional units are particularly important in comparative assessments (26). For a sound comparison, the final functions must be identical for both the assessed process and its benchmark. For example, if two process alternatives produce the same chemical, it is meaningful to compare the processes based on 1 kg of the chemical. In contrast, if, e.g., chemicals with different heating values are compared as fuel alternatives, a comparison based on 1 MJ of provided energy is preferable.

The choice of benchmark processes and products is crucial and depends strongly on the application. For example, methanol produced from CO_2 can be used as chemical feedstock or synthetic fuel. As chemical feedstock, CO_2-based methanol competes with fossil-based methanol and thus should be compared with methanol production from steam reforming of natural gas. In contrast, when using methanol as synthetic fuel, CO_2-based methanol would probably not substitute fossil-based methanol but rather gasoline, and thus should be benchmarked against gasoline production (27).

2.2. Environmental Impact Categories

An ideal LCA should assess all environmental impact categories of concern (25, 28). If instead only a few impact categories are considered, the LCA practitioner might miss increased impacts in other categories that are out of scope, and problems might be shifted to other impact categories. In practice, however, it is not always obvious which impacts are relevant and must be included in the assessment. Selecting impact categories and corresponding methods (called classification in ISO) for a reliable assessment faces further challenges from limitations in inventory data, lack

or immaturity of impact-assessment methods, or even the choice between multiple assessment methods for the same impact category. To support LCA practitioners in this challenge, guidelines on selection of impact categories have been developed for specific impact categories (29), geographical contexts (30), and applications (31).

The standard approach to quantifying environmental impacts is to use impact scores IS_j obtained by multiplying the flows $Q_i = \sum_k Q_{k,i}$ of each resource and emission i (as sum over all processes k in the entire life cycle) with a characterization factor $CF_{i,j}$ for each environmental impact category j (28):

$$IS_j = \sum_i CF_{i,j} \cdot Q_i.$$

Global warming potential (GWP): a measure of the molecule-specific contribution to global warming relative to CO_2

Global warming impact (GWI): a process property that describes the cumulated GWP of all emissions in a product's life cycle

The most prominent impact category is probably climate change (CC), which considers the global warming effect caused by greenhouse gas emissions.[1] The standard characterization factors $CF_{i,CC}$ for impacts on climate change are the global warming potentials (GWP) published by the Intergovernmental Panel on Climate Change (32). GWP quantifies the radiative forcing induced by an emission of a substance relative to the radiative forcing induced by 1 kg of CO_2. The GWP of a substance is measured in kilograms of CO_2 equivalents (kg CO_2e). If GWP values are multiplied with the corresponding inventory results, the global warming impact (GWI) is obtained (33). The distinction between GWP ($=CF_{i,CC}$) and GWI ($=IS_{CC}$) is essential: The characterization factor GWP is an intrinsic, molecular property reflecting the strength of emission to absorb radiation. We refer to properties such as GWP as product properties because they depend exclusively on molecular structure. In contrast, GWI is a process property that describes the cumulated GWP of all emissions in a product's life cycle (also known as carbon footprint) (34). For example, the GWP of the refrigerant R134a is 1,300 kg CO_2 e/kg R134a, whereas the GWI for the production of R134a is only 6.6 kg CO_2 e/kg R134a (35).

2.3. System Boundaries

The system boundary defines which life cycle stages are included in the assessment. In principle, LCA studies should always consider the entire life cycle (**Figure 2**), which is the so-called cradle-to-grave approach. However, in specific cases, narrower system boundaries can be sufficient: cradle-to-gate, gate-to-gate, or gate-to-cradle. A cradle-to-gate approach neglects all emissions after a certain factory gate, i.e., all emissions caused in downstream processes and the end-of-life phase. Such a cradle-to-gate approach is appropriate in comparative assessments if the life cycle after the gate is identical for the compared processes. For example, if the same chemical is produced, the system boundaries need to include only the processes from the extraction of raw materials to the factory gate. Similarly, a gate-to-grave approach can be chosen for competing uses of a scarce product (36) or for alternative waste treatment options. The gate-to-gate approach is typically employed for compiling data for LCA databases (37). A gate-to-gate analysis can be used in comparative assessments only in rare and very special cases and is generally not recommended.

A common pitfall when assessing the environmental impacts of individual processes is the use of inconsistent system boundaries. An assessment accounting for environmental impacts of the electricity supply while excluding the supply of raw materials, for example, will always imply that a shift in energy supply from electricity towards more energy-intensive raw materials is environmentally beneficial. In reality, however, the production of raw materials may have

[1]The terms impact on climate change, global warming impact, greenhouse gas emissions, and carbon footprint are often used equivalently.

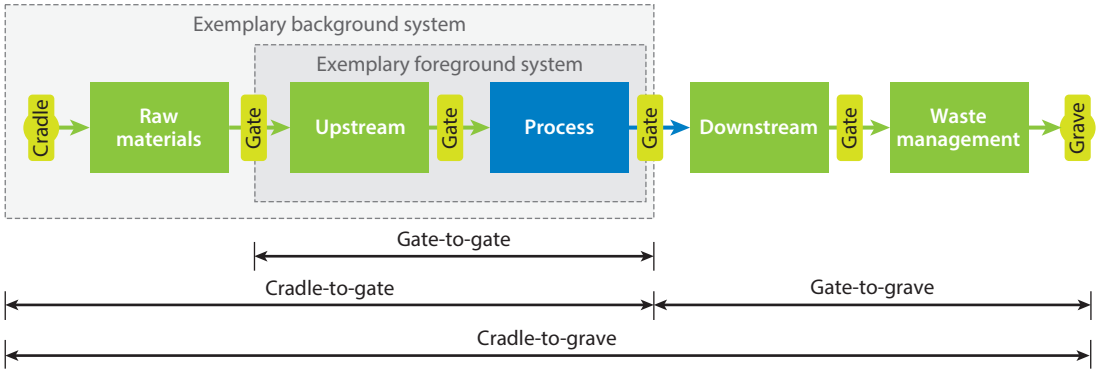

Figure 2

System boundaries in life cycle assessment. Ideally, all processes are included in a so-called cradle-to-grave boundary. The system boundary can further be divided into foreground and background systems. The foreground system is here defined as the part that is "under control," i.e., within the design space.

substantial environmental impacts that need to be accounted for to understand the environmental implications of such a shift. It is therefore crucial to include the provision of both energy and raw materials into the system boundaries (38, 39).

2.4. Foreground versus Background System

The system boundary is often further divided into the foreground and background systems. However, these terms are not part of the ISO standard, and there is no common definition. The foreground system can simply be viewed as the processes "of direct interest" (40). It can also be defined as the part for which specific data are available, e.g., own-company data or supplier-specific data for raw materials and energy (41). Here, we adopt Frischknecht's (42, p. 57) definition: "The foreground system consists of processes which are under the control of the decision-maker." In the context of design approaches in this review, the foreground system thus corresponds to the design space.

Because the background system is the part that is "out of control," it is typically difficult to obtain data for background processes. Especially when the LCA ambition to cover the entire life cycle is taken seriously, data for practically all processes of the entire economy would be required. But even when focusing on the most important background processes, the LCA practitioner always faces data gaps. Thus, we first discuss the possibilities to obtain background data, in particular by using chemical engineering principles (Section 3). Subsequently, we review LCA-based design approaches, where we gradually extend the design space across the life cycle from process design via product design to supply chain design and market-mediated effects (Sections 4–7).

3. DATA SOURCES FOR BACKGROUND SYSTEMS

Owing to its holistic nature, LCA requires data for a huge number of processes. To ease the effort of manual data collection, various approaches are used to approximate LCA data. These approaches should provide the required data at maximum accuracy with minimum effort. Typical data sources can be organized in the following hierarchy:

1. industry data,
2. LCA databases,

3. simulation or laboratory data scaled up to industrial scale, and
4. so-called streamlined LCA approaches.

Life cycle inventory (LCI): contains all mass and energy balances, which are required for an LCA, including both technical flows between life cycle stages and elementary flows exchanged with the environment

Life cycle impact assessment (LCIA): in this assessment, LCI results are translated into environmental effects, such as global warming

Data accuracy typically decreases across these approaches from 1 to 4. The background system should preferably be modeled using firsthand industry data for the considered processes (43). However, real plant data are not always available owing to confidentiality, especially when data must be exchanged between different organizations.

The use of database values for processes in the background system is recommended if data of sufficient quality are available, e.g., anonymized industrial data. Current commercially available databases include approximately 500 commonly used chemicals, focusing on bulk chemicals and intermediates (44). Please note that LCA databases do not always use process data from industry but sometimes also use simple heuristics.

If only laboratory data are available, scaling up those data is essential to ensure sound comparisons, e.g., with industrial-scale benchmarks (45). Simon et al. (46) and Piccinno et al. (47) provide heuristics for the scaling of laboratory data. Laboratory data on substance properties and reaction kinetics can be fed into process simulations to estimate process data at industrial scale, which can then be used as input for LCA studies.

Finally, streamlined LCA approaches can be employed to reduce the required amount of data to a minimum (48). In one approach, the scope can be reduced directly by assessing fewer environmental impact categories and using correlations between impact categories for the left-out categories (49–51). Another approach is to use proxy data or prediction methods (52). These prediction methods are tailored to specific applications (53) and can be classified mainly into two approaches: predicting the life cycle inventory (LCI, the "bill of materials" of the life cycle) or directly predicting the final life cycle impact assessment (LCIA).

In chemical engineering, LCIA results have been predicted using molecular structure models. The logic of these models is that the molecular structure directly influences the complexity of its production process and its hazard and fate at the end of life, and thereby its life cycle environmental impacts. This correlation between molecular structure and the LCIA can be fitted by multi-linear regression models (54–56) or nonlinear models, such as artificial neural networks (57–60). Herein, the physical properties of the product, e.g., the molar mass or the number of functional groups, are used as input to describe the chemical of interest. The regression model is then used to predict an LCIA result based on previous training on a given number of LCA studies for chemicals. Another approach identifies typical groups of chemicals via clustering algorithms. The impact of a new chemical is then predicted using the average of the cluster (61). Similarly, decision trees have been used to classify the expected LCIA results into low, medium, and high environmental impact (62). This approach is based on if–then rules, which use a set of critical parameters of the process chain, e.g., the molecular structure of the product and process-chain-related variables corresponding to chemistry, complexity, and generic process conditions. Current approaches are summarized in **Table 1**.

In contrast to predicting LCIA results directly, other approaches estimate LCIs based on generic flowsheets (37, 63) or heuristics (64). A most simple approach roughly estimates LCI by using stoichiometric mass balances. This approach yields lower bounds for the environmental impacts (64). This simple approach can still be very useful when process candidates are screened against existing benchmarks: If a candidate's lower-bound impacts are already higher than the benchmark, this candidate can be discarded.

More recent approaches are based on advanced process calculations (65, 66) and data mining (67). Parvatker & Eckelman (44) compared these prediction methods for LCI. The authors

Table 1 Prediction methods for life cycle impacts[a]

Category	Method	Training input	Input for prediction	Impacts regarded	Prediction accuracy	Reference
Molecular structure models	Artificial neural networks	100 chemicals as training set	2–17 molecular descriptors	CED, GWP (but meant is GWI), BOD, COD, TOC, Eco-indicator 99	5.8–21% mean relative error due to leave-one-out	57
		338 inventories	10 molecular descriptors	CED, GWP, Eco-indicator 99, electricity use, heat use	0.41–0.69 coefficient of determination 20.7–94.6% mean relative error	58
		Total of 166 chemicals: 10 as test set, 16 as validation set, the rest for training	Molecular descriptors such as functional groups	CED, global warming (IPCC 2007, 100a), acidification (TRACI 2.0), and three end-point impact categories: Eco-indicator 99 (I,I, total) (EI99), ecosystem quality (Impact 2002+), and human health (Impact 2002+)	On test set: between 30 and 65% MRE and R^2 between 0.45 and 0.87	59
		3,073 organic and a few inorganic chemicals	Physical properties such as molecular weight or partitioning coefficient between octanol and water (9 in total)	Vector with the characterization factors to calculate human toxicity and freshwater ecotoxicity	R^2 between 0.46 and 0.96	60
		Total of 63 organic chemicals	185 possible descriptors in total, consisting of 178 molecular descriptors (e.g., physical and chemical properties) and 7 process descriptors (e.g., concentration of each component at the reactor outlet or the sum of the environmental impacts of the reactants)	17 Recipe v1.08 (H) midpoint categories	R^2 between 0 and 0.66	27
	Decision trees	91 study systems as training data	30 predictor variables (molecular descriptors of the FineChem tool and process indicators as proposed by Sogiyama (169) and Patel (170)	ReCiPe and CED methods (23 metrics in total)	13–40% averaged validation error	62
	Multi-linear regression	141 and 90 products	Original set of 18 variables available in the USEtox database	CED, Eco-indicator 99	Error values below 20% using 5 proxy LCIA metrics in the electricity category, and below 15% for 3 proxy indicators in the oil category	54

(*Continued*)

Table 1 (*Continued*)

Category	Method	Training input	Input for prediction	Impacts regarded	Prediction accuracy	Reference
	Multi-linear regression	Data set presented by Wernet et al. (57), but removed outliers and molecules where no thermodynamic properties are available, resulting in 83 data points	Molecular descriptors and thermodynamic properties such as reaction enthalpy or functional groups; 17 molecular descriptors, 15 thermodynamic properties	CED, GWP, COD, BOD, TOC, EI99	Relative errors in the range 20–44%, while COD, BOD5 and TOC showed much larger errors	55, 56
	Clustering	140 materials	Expert knowledge to classify the chemical of interest into 1 of 14 categories, e.g., solvent or organic aliphatic	Net mass of materials used, energy required, greenhouse gas equivalents, oil and natural gas depletion for materials manufacture, acidification potential, eutrophication potential, photochemical ozone creation potential, total organic carbon load before waste treatment	Expected error for most categories is less than 6% RMSE	61
Heuristics	Averaged LCA	No training required	LCA of inputs, stoichiometric equation	All possible	Not stated	64
		Not mentioned	Impact categories, which are correlated with the impact of interest	Damage to human health, damage to eco-system quality, and damage to resources, along with the GWP (GWP100) and the Eco-indicator 99	Not mentioned	51

[a]Eleven papers are reviewed predicting various life cycle impacts based on the molecular structure. For reasons of simplicity, the impacts considered are abbreviated as follows: biological oxygen demand (BOD), cumulative energy demand (CED), chemical oxygen demand (COD), global warming potential (GWP), global warming impact (GWI), total organic carbon (TOC). ReCiPe is a life cycle impact assessment method, which comprises harmonized category indicators and thus provides a variety of impact categories (171).

applied five prediction methods to two case studies for the production of styrene and the subsequent product acrylonitrile butadiene styrene. The results are compared with an LCA study based on process simulations (according to Step 3 of the hierarchy) and an LCA study based on real plant data (according to Step 1 of the hierarchy). In conclusion, process simulations were perceived as the favored method to approximate LCIs, although out of 18 estimated impact

categories, only 4 categories were predicted within a 10% range compared with the full LCA conducted with real plant data. Stoichiometric calculations should be used only when no information regarding the process is available. Molecular structure models are found to underestimate the results for global warming significantly but predict the cumulative energy demand within a 3% range compared with the LCA based on plant LCI. Recent work from our own group (68) has shown that combining molecular structure models with simplified process data (e.g., using ideal thermodynamics) can improve prediction accuracy with low modeling effort. The development of prediction methods for LCIs and life cycle impacts remains an important field for future research. A key challenge for this research is to provide sufficient and reliable data at industrial scale. Therefore, chemical engineers and the chemical industry should work on developing platforms for the anonymous exchange of original plant data. Additionally, uncertainties in the provided LCA data must be quantified and considered when using the data.

4. PROCESS DESIGN

In designing a sustainable chemical process, the most natural starting point is the design of the process itself, i.e., the engineering of a single-site chemical plant. In LCA terminology, this means that the foreground system is represented by the process only. However, even the design of such a single plant itself involves decision-making on multiple scales (69, 70): On the smallest scale, molecules must be selected as catalysts, solvents, or working fluids. On the process scale, equipment types and operating conditions are determined for optimal process performance. Finally, decisions extend to plant-site scale with close links to supply chain design, such as the selection of a reaction network or separation sequences.

On all of these scales, process design can consider environmental aspects, whereby we can distinguish three levels of integration (**Figure 3**): As a first level, LCA can be used for process selection. Here, LCA is applied to fixed process alternatives. Process modeling provides the LCI. LCA results serve as decision criteria for or against selecting a specific process design. However,

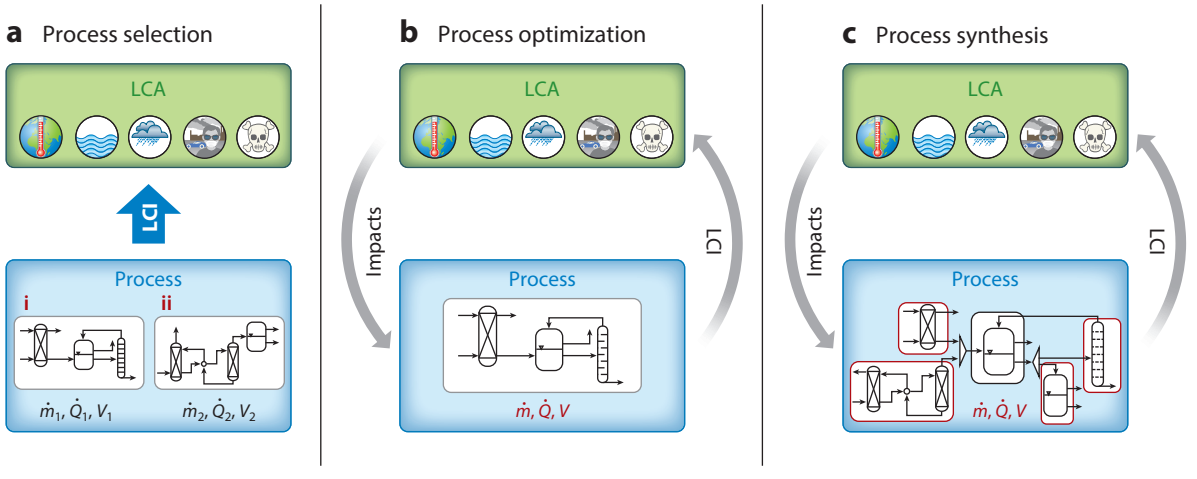

Figure 3

Integration of life cycle assessment (LCA) and process design ranges over various levels, from (*a*) evaluation of fixed process structures by LCA in process selection to (*b*) integrated feedback loops of environmental impact on process design in process optimization and (*c*) process synthesis. On all levels, the integration of LCA is enabled by the provision of life cycle inventory (LCI) from the process model. Red font and boxes indicate optimization variables, i.e., process settings or flowsheet alternatives.

there is no feedback loop from LCA results to process design. As a second level, LCA can provide feedback to process design by integrating LCA into process optimization. Here, mass and heat flows as well as temperatures and pressures are typically optimized for minimal environmental impacts, but the flowsheet is fixed. Finally, as a third level, the flowsheet itself can be generated under environmental consideration so that LCA is integrated into process synthesis.

In this review, we highlight contributions of integrating LCA into process design on all three levels of integration. Because of the large number of contributions in this field, we focus on more recent developments published since 2012. Earlier contributions are summarized in excellent previous reviews (71–73).

4.1. Process Selection

Most contributions to LCA-conscious process design apply LCA after process synthesis and process optimization. These contributions can be categorized as process selection based on comparative LCA (26) (see **Figure 3**). The integration of LCA into process design is rather low, as environmental impacts serve only as decision criteria to choose among fixed process designs and do not provide feedback to process design.

Still, LCA for process selection has been applied for important problems such as the selection of feedstocks (74), catalysts (75), products (76, 77), and process routes (78, 79). Major interest in comparing process alternatives arises from the assessment of novel process designs to conventional processes, e.g., recently in the area of bio-based production of chemicals (80) or carbon capture and utilization (27). Typical tasks include the selection of more sustainable pathways to produce a given product or to utilize a limiting feedstock.

The idea of comparing available options has been extended to provide feedback to process design by analyzing the LCA results of every option, e.g., via hot-spot analysis (81, 82). The identified hot spots provide valuable insights for process designers on where to set a focus in the design of new, improved alternatives. Ott et al. (81) demonstrated this approach to improve a pharmaceutical process. Gear et al. (82) formulated this approach as a standardized flow scheme and showed how the approach leads to more environmentally friendly process decisions for a thermal cracking process for mixed plastic waste.

4.2. Process Optimization

The above-described selection of fixed process designs via comparative LCA requires considerable manual effort and still runs the risk of missing beneficial options. Therefore, it is desirable to consider LCA simultaneously with process design (53). LCA has been incorporated into well-known process design methods, such as optimization by mathematical programming (21, 71). Mathematical programming can include environmental considerations, such as LCA impacts, as constraints or as objective function yielding one integrated, multi-objective optimization problem. Integrated process optimization with LCA commonly optimizes process settings and equipment sizes for a fixed process flowsheet to find economic and environmentally benign solutions (83–88).

Depending on the question at hand, various solution strategies have been developed that can be grouped based on the level of detail for process modeling: Frequently, processes are modeled in an equation-oriented way by formulating mass and energy balances and cost correlations (83–85). The resulting optimization problems can be solved via deterministic mathematical optimization. Most often, multi-objective optimization has been limited to one economic and one environmental objective by focusing on only one LCA impact category, such as the GWI, or by using aggregated metrics, such as the Eco-Indicator 99 (89). Problems that have been solved in this way include optimization of a coupled solar desalination facility for minimum cost and GWI (83), design of

environmentally conscious absorption cooling systems via minimization of Eco-Indicator 99 and cost (84), and optimization of design and operations of a hydrocarbon biorefinery by minimizing GWI and maximizing net present value (85). The optimization typically leads to so-called Pareto optimal solutions, which represent optimal trade-off solutions between the conflicting environmental and economic objectives (22).

If more detailed modeling of the process is required, e.g., by including more detailed thermodynamics, mass and energy balances are often not sufficient, and equation-oriented approaches frequently become computationally demanding because of the highly nonlinear models. For such cases, process simulators and LCA software have been combined with surrogate modeling approaches and derivative-free optimization (86–88). For instance, Gonzalez-Garay & Guillen-Gosalbez (86) developed the SUSCAPE framework for multi-objective design of chemical processes. SUSCAPE employs process simulation and surrogate process models with a genetic algorithm for optimization. At the same time, SUSCAPE aims at accounting for more than one LCA impact category without using aggregated indicators. Instead, SUSCAPE minimizes the number of objective functions with an objective-reduction algorithm (90). The resulting Pareto frontier is explored by multi-criteria decision analysis using data envelopment analysis, which has been extended to include improvement targets (87). To realize the integration of LCA into process optimization, systematic approaches such as the SUSCAPE framework are highly desirable, as they try to capture all facets of the integrated problem.

4.3. Process Synthesis

As a further extension to process optimization, process synthesis not only optimizes given process designs but also derives novel process designs, e.g., by selecting options from a superstructure or by altering the design via evolutionary algorithms. Applications include synthesizing a process flowsheet by selecting from a set of unit operations, selecting an optimal type of equipment for a given task, or choosing a reaction route for a given product (91, 92).

Similar to process optimization problems, synthesis problems have been expanded by LCA. Contributions to LCA-based process synthesis can be distinguished by solution approach and application: Firstly, much work has been performed on formulating equation-oriented superstructure optimization problems for optimal process flowsheets (93–95). Wang et al. (93) developed a superstructure model to determine technology, operational settings, and flow rates of a hydrocarbon biorefinery for maximum net present value and minimum GWI. Similarly, Gong & You (94) optimized a microalgae-to-biodiesel process for minimum GWI and cost. In both works, the results show distinct trade-offs between net present value and GWI. Large savings of GWI can be achieved, but only at higher cost, which highlights the need for multi-objective optimization of conflicting objectives. Demirhan et al. (95) minimized the costs of ammonia production by solving a process superstructure under strict restrictions on GWI. They found that an optimal process design can reduce both greenhouse gas emissions and cost.

Bakshi and colleagues (96) extended the scope of superstructure flowsheet design to include ecosystem services. Environmental impacts have been taken into account by limiting the process design to not exceed natural resources. The capability of this approach has been shown for a biodiesel production facility that can operate sustainably with only small economic losses. Later, Gopalakrishnan et al. (97) extended this work by including the ecosystem as a unit operation to design both process and ecosystem in biodiesel production.

Apart from finding optimal equipment or a process flowsheet, LCA has also been integrated in process pathway design. A topic of major recent interest is the design of reaction networks, e.g., for biorefineries. König et al. (98) investigated the production of bio- and e-fuels to find reaction

pathways that are cost optimal and have low GWI. They formulated an optimization problem including various reaction pathways to various fuel candidates from lignocellulosic biomass as well as from CO_2 and renewable hydrogen. The nonlinear optimization problem was solved for optimal reaction pathways using the method of reaction and process network flux analysis (99, 100). Similarly, Balakrishnan et al. (101) optimized the product portfolio of a sugarcane biorefinery. They developed a novel heterogeneous catalyst and showed the effects of employing the novel catalyst in a biorefinery by optimizing environmental impact as well as production of fuel or lubricant. Furthermore, process pathway design can be closely linked to product design, as shown by Dahmen & Marquardt (102). They incorporated the model-based prediction of fuel properties into pathway design for optimal fuel blends from biomass. Caldeira et al. (103) recently presented an approach to further integrate LCA. They designed a biodiesel blend from waste-based feedstocks for minimizing environmental impacts and production cost constrained by technical fuel requirements.

This type of superstructure optimization often leads to computationally demanding mixed-integer nonlinear programs (MINLPs), in particular when combined with multi-objective optimization. Because the resulting MINLPs are sometimes not solvable with commercial multipurpose solvers, many researchers work not only on the problem formulation but also on tailored solution algorithms (94, 95).

In contrast to formulating an equation-oriented mathematical optimization problem, Maréchal and colleagues (104) created a process synthesis framework based on flowsheeting and process integration software and combined it with an evolutionary, multi-objective optimization algorithm. Their so-called thermo-environomic design explores trade-offs not only in environmental (eco-indicators) and economic objectives (profit) but also in thermodynamic objectives (thermodynamic efficiency). They found that neither objective alone is sufficient for a balanced process design by applying their framework to CO_2 mitigation in chemical processes and oil and gas plants (105–107). Pavão et al. (108) also chose a derivative-free optimization approach, using a meta-heuristic approach with simulated annealing and particle swarm optimization, to synthesize a heat-exchanger network with minimum cost and environmental impacts.

In conclusion, pioneering work has extended the methods of process systems engineering by integrating environmental assessment at all levels of process design. Future work should focus on the key features of LCA: Because LCA is a holistic method, systematic approaches must be further developed and applied for including multiple environmental impact categories. Additionally, the choice of system boundaries needs to be presented consistently for meaningful and comprehensible results.

5. PRODUCT DESIGN

The goal of chemical product design is to find a product that fulfills desired properties and functionalities for an intended application (109). "[Product design] decides ***what*** to make" (110, p. 319). To identify the relevant properties, the function of the product must be known. In this sense, product design is similar to LCA, where the LCA practitioner also first identifies the product's function to define a functional unit (see Section 2). On the basis of the functional unit, environmental impacts of alternative products can be compared to identify the most environmentally friendly product. Given these similarities, it seems natural to merge the typically retrospective LCA approach with predictive product design into LCA-based product design.

Chemical products can be roughly classified into molecular products (single species and blends thereof, e.g., solvents or fuel blends), formulated products (mixtures, e.g., sunscreen lotions), functional products (e.g., controlled-release herbicide granules), and chemical devices (e.g., inhalers

CAMD: computer-aided molecular design

for drug delivery) (111). The design of molecular products is an inherent part of the design of all chemical product classes (110) and is thus most advanced. The key element in computer-aided design of molecular products is the prediction of molecular properties. Therefore, we focus on property prediction in the context of LCA-based (molecular) product design.

In LCA-based product design, two types of product properties are considered: On the one hand, traditional physicochemical properties are considered to maximize product performance and quality. On the other hand, environment-related properties must be considered to ensure an environmentally benign application. For example, the design of a working fluid for organic Rankine cycles should consider not only physicochemical properties, such as enthalpies and thermal conductivity, but also environment-related properties, such as toxicity, ozone-depletion potential, and GWP (112). Whereas traditional product design often optimizes properties for the product-use phase only, product properties can also be tightly coupled with environmental impacts during manufacturing (e.g., fugitive emissions owing to high vapor pressure), during product use (e.g., wear emissions), and in the end-of-life phase (e.g., as persistent waste or difficult-to-recycle composites). To expand the scope of product design, Kümmerer (113) suggested a benign-by-design approach to include the end-of-life stage and to identify properties necessary for easy and fast degradation.

Several advances have been made in computer-aided molecular design (CAMD) methods toward environmentally relevant properties and combinations of CAMD with LCA. Although fully integrated approaches of CAMD and LCA are yet missing, we present recent advances in CAMD in which LCA-relevant properties are considered. Mehrkesh & Karunanithi (114) presented an LCA-based CAMD approach to minimize potential downstream environmental impacts from ecotoxicity of solvents. The ecotoxicity characterization factors were obtained from the USEtox LCA model (115), where the required physicochemical properties and toxicity data were computed with group contribution (GC) models. Schilling and coworkers (116) proposed a CAMD approach for a working fluid for organic Rankine cycles and considered not only conventional physical properties but also nonconventional product properties, such as flammability and autoignition temperature, as well as LCIA characterization factors, such as toxicity, ozone-depletion potential, and GWP. They showed the importance of integrating nonconventional properties into CAMD, as constraints on nonconventional properties excluded otherwise-optimal molecules from the design space. Papadopoulos et al. (117) demonstrated an integrated sustainability assessment and CAMD approach for the design of solvents for chemisorption-based CO_2 capture. The considered sustainability metrics include LCA process metrics computed from a molecular-based neural network (58; see Section 3) as well as environment-relevant product properties computed from GC models. As expected, their comparison of two CAMD approaches with and without LCA objectives showed that considering LCA process metrics reduces environmental impacts. Remarkably, the authors were able to link these impact reductions to the chemical groups involved (e.g., the CAMD approach with LCA constraints favors OH-containing structures) and potential phenomena (e.g., OH groups hinder desired solvent-water immiscibility and phase-change behavior). Von der Assen et al. (118) proposed an LCA-based approach for the design of an environmentally optimal polymer structure and supply chain. The focus of this approach was not on the employed GC model for polymer properties but rather on how alternative LCA choices for by-products along the supply chain affect the optimal polymer structure. The work shows that a deep understanding of the LCA methodology, in this case of allocation methods, is essential in LCA-based product design.

In summary, very few approaches have included LCA in product design, which is likely due to the wide range of applications of chemicals and the corresponding need for many property

predictions. Nevertheless, LCA-based product design provides a great potential to maximize the product's function and simultaneously minimize environmental impacts, i.e., to do more with less.

SCM: supply chain management

6. SUPPLY CHAIN DESIGN

The previous sections focused on the integration of LCA into process and product design. In addition, the implementation of a process typically affects upstream and downstream activities along the entire supply chain. Vice versa, the supply chain can influence the designed process. Therefore, it is desirable not to consider the supply chain as an aggregated background system but to analyze—or even design—the supply chain in detail. For example, supply chains are typically multiregional. A detailed multiregional supply chain analysis can resolve transregional effects, such as changes in transport distances and environmental impact restrictions (119, 120).

Whereas LCA was developed to measure sustainability to support a decision, the implementation of a decision was the original focus of supply chain management (SCM) (121). Owing to the different objectives, these two separate fields of research have emerged over time. Nevertheless, SCM and LCA are conceptually and technically very similar. As Blass & Corbett (121) argue, both fields use the same mapping and measurement approaches to extend the scope from a single process to multiple processes. It is therefore only a small step to add resource consumption and environmental impacts in SCM. We begin our review on sustainable SCM by introducing SCM terminology and mapping these terms to LCA. Then, we review the design possibilities and modeling frameworks in sustainable SCM. SCM is a broad field, including tactical, operational, and strategic decision levels. Here, we focus on strategic SCM, which covers long-term issues, such as facility locations and technology decisions.

SCM commonly discusses different types of system boundaries: forward supply chain, reverse supply chain, and closed-loop supply chain (122) (see **Figure 4**). Mentzer et al. (123) describe the forward supply chain as the set of processes from a source to a customer. In other words, the forward supply chain design focuses on determining the production network and logistics

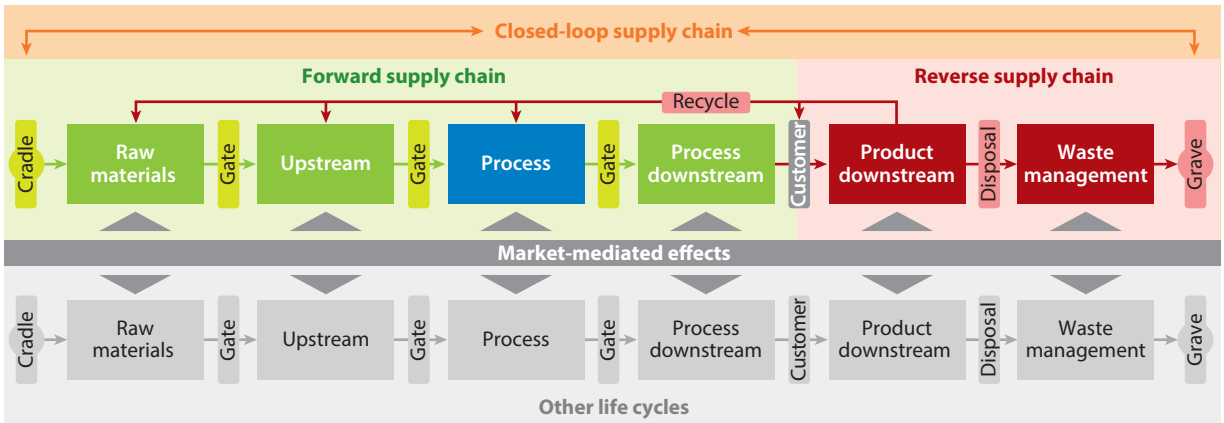

Figure 4

System boundaries for supply chain management and the integration of market-mediated effects. Typical system boundaries in supply chain management include forward supply chain, reverse supply chain, and closed-loop supply chain. These system boundaries can be expanded to account for interactions with other life cycles via market-mediated effects.

operations from raw material to customer (cradle-to-gate). A typical task when analyzing a forward supply chain is to identify the most environmentally beneficial production pathways for a product. For example, Mahbub et al. (124) compared the environmental impacts of conventional diesel production to bio-based oxymethylene ether (OME) blends. In contrast to a process-level LCA, they expanded the foreground system to the forward supply chain of OME synthesis from the supply, transportation, and conversion of biomass to the combustion of OME blends. They analyzed selected pathways toward OME blends using LCA. Because the complexity of the supply chain was manageable, the environmentally best pathway could be selected manually (124). Looking at multiple environmental indicators, von der Assen et al. (125) analyzed to what extent the use of CO_2 as a building block in the forward supply chain of polyurethanes is environmentally beneficial compared with conventional production. Although the number of production pathways is limited, they have been considered in an integrated model and are therefore interdependent. Owing to the higher complexity, an optimization-based approach was chosen to find the environmentally optimal solution. Kätelhön et al. (126) expanded this scope to the forward supply chain of 20 large-volume chemicals to quantify climate benefits and trade-offs of CO_2 utilization technologies to supply the global chemical industry.

In addition to environmental indicators, economic indicators are often considered simultaneously in forward supply chain design. For example, You et al. (127) investigated the trade-offs between economic and environmental performance of cellulosic biofuel supply chains. Yang et al. (128) compared the economic and environmental performance of three production pathways of ethylene from different feedstocks. Their detailed modeling allows analysis of additional effects, such as supply seasonality, geographical diversity, and biomass degradation, on the environmental performance of the process.

In contrast to the forward supply chain, the reverse supply chain refers to logistical activities carried out in recycling, substitution, reuse of materials, and disposal (gate-to-grave) (129). By considering the reverse supply chain, the question of whether a product should be recycled or disposed of can be answered from an economic and environmental point of view. Thereby, not only different recycling technologies but also different waste sources can be considered. Guo and colleagues (130) analyzed the environmental impact of mechanical recycling of different sorts of plastic waste using different waste management technologies and compared the impacts to virgin material production. Another use of reverse supply chains answers the question of which waste management technology should dispose of a product in the most sustainable way. For more information, see work by Laurent et al. (131, 132).

However, the isolated consideration of either forward or reverse supply chains still neglects interactions between these system boundaries. For example, changing the production pathway may reduce the environmental impact of production, but benefits may be outweighed by higher recycling efforts. To consider effects and opportunities in both the forward and reverse supply chain, closed-loop supply chains are considered (133, 134). Closed-loop SCM maximizes value throughout the entire life cycle of a product (cradle-to-grave). The design of complete, closed-loop supply chains is an approach that has hardly been implemented to date and will therefore be an important field of research in the future.

Furthermore, LCA at the process level can be linked with SCM. Hanes & Bakshi (135), for example, proposed the process to planet (P2P) framework, a multiscale modeling framework that aims to integrate process models into their supply chains. P2P uses process-level data in combination with value-chain and economy data derived from economic input–output databases. Thereby, P2P can investigate the impact of changes within the process on the overall supply chain.

Typically, there are many possible combinations of process design and SCM. Finding the best possible solution therefore requires the use of decision-making tools such as optimization.

Optimization is often used to identify production pathways with the highest revenues and the lowest environmental impacts. As Barbosa-Póvoa et al. (136) recently demonstrated, optimization models can be used as a practical tool for designing sustainable supply chains. Optimization tools can help to identify environmental hot spots in complex supply chains. Whereas our review focuses on strategic supply chains with environmental objectives, Barbosa-Póvoa et al.'s (137) broader review summarized the main characteristics of optimization models for the design of sustainable supply chains. These characteristics include the system boundaries, decision variables, constraints, and objectives. In addition, they identified the most important research gaps in supply chain design and planning and developed a set of guidelines for the development of supply chain optimization models (137).

Aside from these guidelines, specific models for the design and optimization of sustainable supply chains have already been developed. For example, Mota et al. (138) looked into the design and planning of sustainable supply chains, adopting a multi-objective approach in which the three pillars of sustainability were considered. A closed-loop supply chain for a battery producer was studied, and different supply chain structures were obtained based on the respective objectives considered. A near-optimal solution was reached easily when considering the three sustainability pillars simultaneously. In contrast to Mota et al. (138), who implemented a bottom-up approach using only unit process data, Yue et al. (139) developed a top-down life cycle optimization framework that combines unit process data and economic input–output data. This so-called hybrid LCA approach (140) can assess interactions between the considered system and other industrial sectors in the economy.

Of course, the quality of the results of supply chain optimization always depends on the data used. However, the comprehensive representation of the supply chain, including multiple production steps, is always associated with simplifications and uncertainties (120). Therefore, the integration of uncertainty as well as risk and resilience aspects is important for the design of robust and sustainable processes and supply chains. A detailed analysis of the main sources of uncertainty arising from the consideration of the supply chain would help in understanding the impact on the quality of the LCA results (141). Other future research topics for sustainable process and supply chain design are the development of integrated decision models that consider all life phases of a product and the sound treatment of all sustainability pillars. In this context, Mota et al. (142) have already addressed the shortcomings of existing models and proposed the multi-objective optimization tool ToBLoOM for the design and planning of sustainable supply chains under uncertainty. In addition, efficient approaches are highly desirable to overcome multi-scale, multi-target, and multiplayer challenges (5, 136, 137).

7. MARKET-MEDIATED EFFECTS

In addition to the physical supply chains discussed in the previous section, process design can also affect other product life cycles via market-mediated effects (see gray boxes in **Figure 4**). The integration of market-mediated effects into LCA is often discussed under the term consequential LCA. Consequential LCA aims at capturing the environmental consequences of decisions, such as the introduction of a new technology, the purchase of a product, or changes in process design (143–146). Consequential LCA combines technical and economic modeling to trace these consequences throughout the economy, considering both technical relationships and market-mediated effects. In this section, we first review market-mediated effects relevant for process design. Subsequently, we discuss modeling approaches used to incorporate market-mediated effects into LCA-based process design.

7.1. Types of Market-Mediated Effects

TCM: technology choice model

Process design decisions can trigger various types of market-mediated effects. A commonly discussed market-mediated effect is a change in technologies that are required to satisfy an increase in a market demand. This increased demand for, e.g., raw materials or energy can be due to a change in the process design. Importantly, the additional demand is not necessarily produced by the average technology mix in the market supply (147). In Germany, for example, approximately 13% of electricity is produced by nuclear power plants (148). An increase in electricity demand, however, will not increase nuclear power production in the same way, owing to the limited additional capacity of nuclear power plants and a political commitment to phase out nuclear power generation (149). Consequently, the marginal technology mix used to satisfy an additional electricity demand will probably contain less nuclear power, and thus, the marginal differs from the average technology mix. Considering marginal technologies for the supply of energy and reactants represents the actual consequences of design decisions (147). Considering market-mediated effects through marginal technology mixes for energy and reactants may lead to different process designs. Thus, the identification of marginal technologies is of major importance for assessing the impacts of process design choices.

Other market-mediated effects relate to price changes and changes in production cost. A decrease in price commonly leads to an increase in demand and vice versa (150). This effect is denoted the price elasticity of demand. Owing to the price elasticity of demand, environmental benefits from efficiency improvements in process design can partly be offset by increased production triggered by additional demand, the so-called rebound effect. Similarly, price increases through the implementation of low-carbon technologies can even amplify environmental benefits by decreasing demand. In addition, changes in production cost due to design changes can affect the relative competitiveness of competing technologies producing the same product and hence lead to substitution effects among competing technologies (151).

7.2. Modeling Approaches

Various modeling approaches have been proposed to account for market-mediated effects in LCA (144, 152). The application of these models has been demonstrated in numerous consequential LCA studies (for a review, see 144). However, fewer examples exist for the integration of market-mediated effects into LCA-based process design.

Weidema and colleagues (143, 147) proposed a prominent method for identifying marginal technologies. This method involves a step-wise procedure in which each step focuses on a specific question: (*a*) What is the relevant time horizon? (*b*) Are specific processes or overall markets affected? (*c*) What is the trend in the market? (*d*) Which technologies are flexible? (*e*) What technology is actually affected? By answering these questions, the user is guided through a decision tree to identify marginal technologies. These marginal technologies can then be included in the system boundaries of LCA or process design models to account for changes in demand for reactants or energy. Thonemann & Pizzol (153), for example, used this procedure to identify marginal technologies for a potential increase in supply of H_2 and CO_2 owing to the introduction of novel carbon capture and utilization technologies. Advantages of the procedure include its easy applicability. However, Mathiesen et al. (154) showed for the Danish energy system that the procedure did not match the actual historical developments of the energy system.

The technology choice model (TCM) provides a more complex model enabling the simultaneous determination of both marginal technology mixes and substitution effects among competing technologies (155). TCM is a bottom-up model of industrial production systems in which technologies are represented based on engineering-level data. In TCM, each product can be

produced by more than one technology, and the choice of technology is based on economic or environmental objectives, taking into account constraints in factor availability and parameter uncertainty. Using TCM, Larrea-Gallegos et al. (156) analyzed changes in land use, water consumption, and greenhouse gas emissions owing to an increase in pisco production in Peru. Furthermore, Budzinski et al. (157) used a similar model to determine Pareto-efficient configurations and feedstock supply regions for biorefineries, considering economic and environmental objectives. The studies demonstrate the ability of TCM to model complex supply chains at a high level of detail while systematically taking into account market-mediated effects. However, individual processes are so far treated as black boxes and thus cannot be optimized simultaneously in terms of process parameters.

Other classes of models used in consequential LCA studies are partial equilibrium (PE) and computational general equilibrium models (144). These models can simultaneously determine changes in price and demand based on econometrically derived data. The complexity of PE models ranges from relatively simple models of single markets to complex models covering multiple regions (158, 159). In contrast, computational general equilibrium models cover all markets within the economy (for example, 160, 161). Voll et al. (162) combined a simple PE model of wood markets with a process model for biofuel production to consider the interdependency between wood demand and price in the design of a biofuel process. Gong & You (163) combined modeling approaches from LCA and techno-economic analysis with process and PE models to simultaneously optimize process parameters, technology choices, and markets in a MINLP model. Integrating economic equilibrium models into process design can resolve the interactions between processes and markets in settings. These interactions are important where design decisions have a substantial effect on the overall market, e.g., the effect of large-scale biorefineries on local agricultural markets. The integration of PE models, however, is constrained in practice by the availability of high-quality economic data, such as price elasticities (145, 164). Furthermore, equilibrium models are based on several simplifying assumptions that may not reflect market reality. For example, equilibrium models assume that markets are in equilibrium, economic agents act rationally under perfect information and foresight, and individual decision-making by economic agents leads to a global economic optimum (164).

To allow a more detailed representation of the behavior and objectives of economic agents, other authors explored the integration of agent-based models into LCA and process design. Agent-based models take the perspective of multiple individual agents (165). Each agent individually assesses their own situation and makes decisions based on predefined rules. By simulating the interplay between multiple agents, agent-based models can derive information on the dynamics of the overall system. Singh et al. (166) developed an agent-based model of corn markets and used this model to optimize biorefinery supply chain networks. In this model, corn prices are determined by auctions between biorefinery agents, farmer agents, and a food market agent. Bichraoui-Draper et al. (167) developed an agent-based model to investigate switchgrass production patterns for a bioenergy system.

The consideration of market-mediated effects enables the assessment of consequences of process design beyond physical supply chains. These consequences are likely to be relevant for changes in production processes that affect the overall market, i.e., processes that either produce or consume substantial shares of the market for individual products (168). However, there is no single, commonly accepted method for integrating market-mediated effects into process design, and different modeling approaches may lead to different outcomes (145). A detailed guidance for selecting modeling approaches for market-mediated effects is strongly needed for LCA-based process design.

8. UNCERTAINTY AND VALIDATION

Because LCA is a quantitative, data-driven methodology, it is inherently uncertain. In consequence, LCA-based chemical process design should consider data uncertainty and validation. Data uncertainty can be divided into complete lack of data (i.e., data gaps) and data inaccuracy (172). Whereas the previous sections present methods to close data gaps, this section briefly highlights approaches for how to (*a*) quantify and reduce inaccuracy in LCA, (*b*) validate LCA results, and (*c*) incorporate uncertainty into LCA-based chemical design.

Quantifying uncertainty is essential for a more trustworthy evaluation of LCA results. For this purpose, the LCA practitioner must be aware of all types of uncertainty: uncertainty in parameters and models and uncertainty owing to choices as well as spatial and temporal variability (173, 174). For the different types of uncertainty, several quantification methods have been developed and applied in the context of LCA (174–176), covering both analytical (177, 178) and stochastic (179) approaches. Heijungs & Lenzen (180) argue that stochastic methods provide more information, while also requiring detailed uncertainty distributions and higher computational effort than analytical methods. Still, Lloyd & Ries (175) found that most LCA practitioners use stochastic methods. In our view, both stochastic and analytical methods ultimately allow for proper analysis of error propagation. Error propagation, however, requires knowledge about the uncertainty of the inputs. In LCA-based process design, uncertain inputs often refer to uncertain mass and energy balances for which chemical engineers have a good understanding, because they also affect process economics. However, LCA suffers from further uncertainties less familiar to chemical engineers. In our view, major uncertainties in LCA are due to the "unknown unknowns," where important elements are completely missing from the analysis and might reveal themselves only after time. Important unknowns in LCA occur in LCIA because environmental impacts are often not yet known for many chemicals, and novel impact categories are continuously developed to capture all impacts of our actions on the environment. As a prominent example, early assessment of biofuels focused on benefits for climate change and ignored potential negative consequences of land-use change. Having quantified the uncertainty in LCA results, the LCA practitioner can reduce the uncertainty by increasing data accuracy of key parameters using the methods discussed in Section 3. A powerful method to identify these key parameters is global sensitivity analysis (181, 182), which analyzes how strongly each parameter affects the overall LCA results.

Depending on the scale of the LCA study, the practitioner should validate the input data and LCA results on various levels of detail. Thinking about environmental impacts would have to start during the development of the basic chemistry, e.g., by identifying and accounting for potentially harmful side-products. On all levels, from individual unit operations to the total system, the consistency of mass and energy balances should be evaluated (174). In addition, alternative data sources can be combined, e.g., LCA databases, input–output tables, and historical emission registers (183, 184). In particular, formats are needed to share actual industrial data. A role model could be PlasticsEurope's Eco-profiles for polymers (185). Other forms of validation include critical peer review as requested by the ISO norms for different types of LCA studies (ISO 14040). Still, validation of LCA results remains a major challenge.

LCA-based design effectively compares different design alternatives. However, some uncertainties are common to all alternatives (e.g., the carbon footprint of the future electricity mix). Wei et al. (186) and Beltran et al. (187) therefore developed methods for calculating the probability that one alternative is better than others. These methods also allow for the inclusion of uncertainty in decision making.

Decision making under uncertainty is therefore important for all LCA-based design approaches for choosing between process configurations, product specifications, and supply chain

pathways. However, very little work has been done in considering uncertainty in LCA-based design. Guillén-Gosálbez & Grossmann (188) were the first to explicitly consider uncertainty in environmental impacts in optimal design of chemical supply chains. More recently, Mota et al. (141) addressed the improvement of sustainability reporting through the identification of the uncertainty sources in LCA methodologies and concluded that different LCIA methods and different normalization data sets result in significantly different supply chain business and environmental strategies.

Gavanker et al. (189) highlighted the importance and critical components for clearly communicating uncertain LCA results. In general, there is a consensus in the LCA community that uncertainty analysis is important; nevertheless, it is not yet common practice. We agree with Igos et al. (190), who called for a more detailed review of uncertainty assessment in LCA.

9. CONCLUSIONS AND THE WAY FORWARD

Today, LCA is recognized as an important part of environmentally benign process design. The key feature of LCA is its holistic approach, which considers all life cycle stages and impact categories in the assessment. Thus, LCA avoids shifting of environmental impacts between life cycle stages and impact categories. This life cycle thinking expands the design scope for environmentally benign processes to an overall life cycle design that includes not only the process but also the product, supply chains, and even market-mediated effects. Here, we have reviewed recent work toward the integration of LCA into chemical engineering design.

A key prerequisite for its successful integration is a deep understanding of LCA. In particular, LCA-based design needs a clear definition of (*a*) the functional unit, (*b*) benchmarks, and (*c*) system boundaries. Although the present review aimed at being educational regarding LCA, we strongly recommend that the chemical engineering curriculum should evolve from assessing only mass and energy balances to life cycle thinking and environmental balances using LCA.

Although balance equations are natural to chemical engineers, the evaluation of multiple environmental impacts provides novel challenges. Aggregation of multiple environmental impact categories should be avoided. However, major insight can be obtained using methods for dimensionality reduction. At the same time, the designer must be aware that uncertainties differ greatly between impact categories. LCA-based design thus requires a thorough understanding of the underlying models of cause-and-effect chains.

The reviewed literature clearly shows that the scope of environmental assessment is currently extended from solely assessing the process itself toward an integrated assessment of process, product, and supply chain. Although this extension in scope is highly desirable to exploit new degrees of freedom and to avoid problem shifting, a complete integration is often not yet possible for various reasons: One reason are gaps in reliable LCA data that cannot yet be closed accurately by prediction models. A second reason is the lack of accurate prediction models for molecular properties hindering an LCA-based molecular product design. As a third reason, the design space cannot be freely extended to cover processes, products, and supply chains simultaneously. For example, strict regulations in drug approval require a sequential design approach and do not yet allow an integrated design. Finally, a general framework for LCA-based product, process, and supply chain design is still missing. Once a scientific consensus is established, the long-term challenge is to transfer the scientific methods into industrial applications. For this purpose, user-friendly software tools are needed that support both process design and LCA simultaneously, regardless of the development phase. Even more, LCA and process design should be integrated not only in software but also in the organizational structure of companies. Our vision is that the next generation

of chemical engineers should no longer differentiate between process design and environmental assessment, but process design should always aim for sustainability.

SUMMARY POINTS

- Environmental assessment in process design gains increasing importance.
- The scope of environmental assessment in process design is currently extended from solely assessing the process itself toward an integrated assessment of process, product, and supply chain.
- LCA-based design requires a thorough understanding of LCA and the underlying models of cause-and-effect chains.
- LCA-based design needs a clear definition of (*a*) the functional unit, (*b*) benchmarks, and (*c*) system boundaries.
- Aggregation of multiple environmental impact categories into a single indicator should be avoided.
- LCA is inherently uncertain, and communication of LCA results should therefore account for uncertainty.
- The education of chemical engineers should evolve from assessing only mass and energy balances to life cycle thinking and environmental balances using LCA.
- The transfer of LCA methods into industrial application requires user-friendly software to simultaneously support process design and LCA, regardless of the development phase.

DISCLOSURE STATEMENT

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