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Circularity and Sustainability – an approach for the integrated assessment of life cycle resource, environmental, techno-economic, and circularity performance, and its application to solar photovoltaic systems

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Summary

Continually increasing consumerism since the first industrial revolution coupled with population growth have led to levels of resource consumption, waste generation, and pollution that the planet can not sustain. As a result, it may not be possible to achieve the goals of the Paris Agreement to limit global warming to no more than 1.5°C above pre-industrial levels. We are, in fact, approaching several tipping points beyond which abrupt and irreversible changes in climate with severe societal impacts become dangerously likely. Feeding into addressing this challenge, the goal of sustainable development is to meet current needs without leaving the planet in a state that would prevent future generations from meeting their needs. It is a multifaceted concept but is usually described in terms of its three main dimensions—environment, economy, and society. The United Nations have adopted seventeen interconnected Sustainable Development Goals (SDGs) to ensure future prosperity, and the European Commission considers the Circular Economy (CE) concept as a valuable tool to achieve these goals, especially for ensuring sustainable production and consumption.

A large contributor to achieving sustainable development is the decarbonisation of electricity grids—reducing the generation of electricity from fossil-based resources in favour of increased renewable energy generation using, among other renewable sources, solar and wind energy. The solar photovoltaic (PV) sector is set to grow significantly between now and 2050 and beyond. This will require proportional growth in infrastructure and in the consumption of materials and energy needed for their production. Furthermore, there will be a proportionate increase in the generation of waste when these systems come out of service after a lifetime of 25 or more years. PV systems are complex, not only their value chains but also in terms of the substantial number of specialty metals and other materials used, depending on the specific technology. Intuitively, circular material flows in a CE paradigm have a key role to play in keeping materials in circulation and out of landfill.

Attached to all these growth activities and circular strategies, however, are their own resource, environmental, economic, and societal impacts and a need for sophisticated recycling processes. Somewhat surprisingly, this intersection between sustainability and circularity is still rather blurry—concrete information and quantitative methods that assess the links between sustainability and circularity are lacking, i.e. whether circular approaches do, in fact, enhance sustainability, and to what extent. It has been stated that the sustainability of CE needs to be evaluated, as positive impacts are not guaranteed. These research gaps are addressed in this dissertation, which has the overall objective of quantifying the links between sustainability and circularity by advancing the state-of-the-art for the sustainability assessment of complex product systems with an emphasis on including potential CE strategies. As the electricity sector is key in achieving the SDGs,

there is a focus on PV-generated renewable energy. This is achieved in stages, as evidenced by four published journal articles that accompany the dissertation.

The first article assesses the prospect of simulating a very large, closed loop life cycle system comprising several base metal production and recycling systems that also produce the metals needed to produce cadmium-telluride PV modules as by-products. Process simulation, which considers the laws of mass and energy conservation as well as the second law of thermodynamics to account for entropy generation is used, as it is the only way to track the minor elements (including critical raw materials) through large life cycles. Article 1 shows the benefits of mapping all the physical flows in a single simulation platform—the product is an approach that first creates a highly detailed, disaggregated process simulation model of the entire technology life cycle to quantify material and energy flows, as well as the enthalpy, entropy, and free energy of all elements, compounds, and mixtures to quantify exergy dissipation (due to entropy generation). The detailed inventory then serves as a single source of input data for assessments of resource, environmental and techno-economic life cycle performance.

Building on the same principles, a monocrystalline Si-based PV system is assessed in Article 2. The approach is expanded by incorporating two closed recycling loops in the assessment. Article 2 represents the first analysis of the interactions between sustainability and circularity, in this case only considering environmental sustainability. Modelling the effects of circularity is a computationally intensive task, especially in the case of large process simulation models with recycling loops and several parameters changing independently. As a methodological contribution, this challenge is addressed by creating artificial neural networks (NNs) as surrogate functions that act as proxies for the simulation models. These enable efficient analysis of the responses of resource consumption, carbon footprint, and other indicators to changes in the degree of circularity. In doing so, model runs that would otherwise have taken days are done in seconds. Results show both the directions and relative magnitudes of change in the system's carbon footprint as a function of the degree to which the two circular strategies are implemented. Among others, a key finding is that increased circularity lowers the carbon footprint in the Si PV system, primarily due to avoiding energy-intensive Si production processes, the caveat being that recyclates are of high purity, which requires the appropriate infrastructure.

A further development in Article 3 is the development of a bottom-up cost model that also responds to changes in circularity, revealing how minimum module prices might respond. This body of work is concluded in Article 4 with the further development of cost models and the simultaneous assessment of resource, carbon footprint, and techno-economic performance and their responses to circularity. It is applied to the aforementioned silicon-based PV system and two promising emerging technologies—a single-junction perovskite system and a perovskite-silicon tandem system, both of which are receiving significant academic and industrial attention at present. Additional assessments, such as on the effects of substituting production technologies, changing material intensity, changing supply chain locations, and carbon taxation are also conducted. A key finding from Article 4 is that there is no one conclusion about whether circularity increases sustainability—while circularity benefits both environmental and techno-economic performance in the silicon system, it has the opposite effect in the perovskite system. In the tandem system, a trade-off is found to exist between the environmental and techno-economic dimensions, highlighting that further investigation and optimisation is necessary.

Although the term *sustainability* is used throughout this dissertation, it is acknowledged that, to gain a more comprehensive understanding of sustainability performance, other environmental impact categories and social impacts need to be considered. In the majority of this work, carbon footprint is taken as the main environmental impact. Although this is considered acceptable, the risk of shifting burden between impacts or life cycle stages still exists and should be investigated further. Furthermore, the findings presented cannot be applied to PV systems other than those selected for this dissertation.

This dissertation contributes to methodological advancement by creating harmonised, physics-based inventories at the process level as a basis from which to conduct resource, environmental, and techno-economic performance assessments. Furthermore, exergy analysis is used to account for the 'invisible' resource loss that occurs due to the unavoidable generation of entropy. In addition, NN-based surrogate functions are uniquely employed to facilitate efficient incorporation of the effects of circularity into assessments. It contributes to sustainability and CE research by quantifying the links between circularity and resource efficiency, carbon footprint, and techno-economic performance. This enables assessments for which information from more than one dimension is needed, such as quantifying the combined effects of policy measures like carbon taxation and CE, which requires fully aligned emissions and techno-economic performance data, as well as how they respond to variations in circularity. The dissertation provides PV-specific insights that researchers, industry, and policy makers can use to guide their decisions about future directions.

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From day one, I wanted to do something that would make a tangible difference and not end up in a library somewhere, never to be read again. I hope that I have achieved that. One thing that is certain regardless, is that I have changed and grown. It wouldn't have been possible without the support of the people that surround me.

First, I'd like to thank Markus Reuter for the opportunity to come to Germany to take on this challenge, and for trusting me with this important topic. I'm glad to have contributed to making your vision and the work you started in the early nineties a reality. Our connection might have begun even earlier in the late seventies when my mother was working for your father-in-law at the academic hospital in Bloemfontein when I was only three or four years old. Little did we know our paths would eventually cross in 2017, of all places, in the small town of Freiberg, where we started the work in this dissertation. By chance, we also ended up living in the same street in Dresden where the many gatherings provided a welcome taste of home and home language with delicious boerekos dinners prepared by the lovely Karen, great conversations and advice about work and life in general, and reminiscing about the old days in South Africa and Australia where we had previously lived and worked, but never met. Among many other things, you taught me the finer details of process simulation and neural networks, and how to utilise the results effectively, also in terms of sustainability and circularity. You raised my awareness of how things fit together in large integrated systems and the importance of quantifying them properly using physics, contrary to many mainstream approaches that consider it "too much work". We did it anyway and showed that it isn't. Whether serendipitous or by the hand of something greater beyond our understanding, I value the connection we have made which, in a way, represents a much deeper level of circularity with things coming full circle over the last four and a half decades. I look forward to future catch-ups and collaborations. Many thanks to Magnus Fröhling for taking on a "mature-aged" foreigner with an industry background as a student, for taking me along with you to TUM, and for being the voice of reason when faced with challenges, especially those that inevitably come with being in the academic world. Your thorough reviews of my work, advice and guidance on many topics have been tremendously helpful in developing my work and my way of thinking. I'm sure our paths will cross again in the near future. Many thanks also to Rutger Schlatmann, my "honorary" supervisor, for all your guidance and support, not just through sharing your knowledge, giving ideas, and contributing to publications, but also through flexibility and allowing me time to complete this work. I consider myself fortunate to now be working for you as a result of our collaboration.

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If I survive the defence, this dissertation proves that being in your forties doesn't mean that it's too late to take on the doctorate challenge. On the contrary, my life and work experience have given me broad perspectives that only come with experience. Learning German, however, is a different story altogether!

This dissertation is dedicated to my mother, Catheleen, and brother, Etienne,

and to those who are no longer with us: my father, Cecil,

and my four grandparents, Maxie, Alta, Cecil, and Willie

Contents

1 Introduction

1.1 Background

Natural resource consumption, waste generation, and pollutant emissions have increased to levels that cannot be sustained by the planet. As a result of sustained greenhouse gas (GHG) emissions over many decades, we are now nearing several tipping points beyond which the effects of global warming could lead to abrupt and irreversible climate effects with severe societal impacts (Lenton et al. 2019).

Economic and technological growth resulting from the Industrial Revolution have led to significant global population growth, and population growth generally leads to increased negative environmental impacts (Goodwin et al. 2020). According to the United Nations (UN), the global population could reach 8.5 billion by 2030, 9.7 billion by 2050, and 10.9 billion by 2100 (UN 2019), which undoubtedly points to consumption growth and proportional increases in GHG and other pollutant emissions if no remedial actions are taken. Furthermore, most household and industrial technologies, the demand for which will increase with continuing population growth, urbanization, economic development, and globalization, make use of numerous minor metals and other special elements that provide specific functionalities. In addition to geopolitical sensitivities, some of these elements are becoming increasingly difficult to come by from a technical perspective because of decreasing ore grades, which necessitates the use of more energy-intensive extraction processes. Even if, hypothetically, the consumption of primary resources could be stopped, we would still have to contend with the unavoidable material and energy degradation and losses.

Together, continuous growth, the consumption and potential depletion of limited primary resources, and unavoidable losses make it clear that reducing consumption of resources to levels within planetary limits, minimising material losses, and running processes at their thermodynamic limits are key.

1.2 Sustainable development, life cycle thinking, and circular economy

Sustainable development (SD), life cycle thinking (LCT), and CE are among the concepts that have received significant attention in academic research, public policy development, and implementation in industry to address some of the challenges described above (Beaulieu et al. 2015; Geissdoerfer et al. 2017; Kara et al. 2022).

SD is a broad concept that seeks to "meet the needs of the present without compromising the ability of future generations to meet their own needs" (WCED 1987) in three interdependent development dimensions: environmental sustainability[1], aimed at preventing damage to the environment needed for quality of life and economic activities; social sustainability, which aims to preserve human rights, equality, and diversity, among others; and economic sustainability to maintain the human and natural capital needed to provide income and decent living standards (Klarin 2018). These are also referred to as the three pillars of sustainability and the triple bottom line.

LCT is a related concept with a focus on products and services, defined as *"going beyond the traditional focus on production site and manufacturing processes to include environmental, social and economic impacts of a product over its entire life cycle"* (Life Cycle Initiative 2017; Mazzi 2020). Its primary objectives are to increase resource efficiency, decrease environmental emissions, and to improve the life cycle social and socio-economic performance of a product or service (Ibid.). The life cycle includes all stages from acquisition of primary and/or secondary materials, processing and manufacturing to usage, collection, end-of-life (EoL) treatment, recycling, and final disposal. The main purpose of adopting a life cycle perspective is that it facilitates the identification and prevention of 'burden shifting' between life cycle stages, i.e. when an intervention that reduces the impacts in one life cycle stage or process results in an increase in the impact of another stage or process (Bjørn et al. 2018). Current and prospective life cycle performance in the individual sustainability dimensions are usually assessed using established methods based on the Life Cycle Assessment (LCA) standards and amendments (ISO 14040:2006/Amd 1:2020 and ISO 14044:2006/Amd 2:2020) (ISO 2020a, 2020b) for estimating potential environmental impacts, Life Cycle Costing (LCC) and Techno-economic Assessment (TEA) (Mahmud et al. 2021) for economic performance, and social LCA (S-LCA) (UNEP 2020) for social impacts.

Numerous attempts have been made to formulate a clear and universally applicable definition of CE. Based on their analysis of ninety-five unique published definitions, Kirchherr and colleagues formulated the following comprehensive description:

"A circular economy describes an economic system that is based on business models which replace the 'end-of-life' concept with reducing, alternatively reusing, recycling and recovering material in production/distribution and consumption processes, thus operating at the micro level (products, companies, consumers), meso level (eco-industrial parks) and macro level (city, region, nation and beyond), with the aim to accomplish sustainable development, which implies creating environmental quality, economic prosperity and social equity, to the benefit of current and future generations." and *"It is enabled by novel business models and responsible consumers."* (Kirchherr et al. 2017).

[1] *Sustainability* and the phrase *sustainable development* are used interchangeably in this dissertation.

It clearly states that the goal of CE is to achieve sustainability by enhancing environmental quality, economic prosperity and social equity, now and into the future. It includes a systems perspective and the 4R framework (reduce, reuse, recycle, recover) as its core principles, and business models and consumers as its enablers (Kirchherr et al. 2017). Kirchherr and van Santen (2019) add, however, that practitioners are not interested in slight differences between the numerous definitions of CE, but rather in gaining an understanding of how to implement it in practice. Roos Lindgreen et al. (2020) identify three key ingredients of CE as a means to achieve sustainability: the retention of resource value to decouple growth and primary resource consumption, a hierarchical framework of resource management approaches, and the overarching goal to provide pathways to increased sustainability. These are all covered in the definition above. However, with continuous population growth, economic development, and the unavoidable losses of materials and useful energy that occur during each life cycle a material is used in, it is doubtful whether economic growth and the consumption of primary resources can be decoupled completely. This would only be achievable if the definition's "*reducing*" interventions exceed the increasing demand brought about by population growth and economic development. There are also limits to how much "reducing" can take place before a product loses its functionality. As Skene (2018) states: "*Circles can also never deliver growth. You need ever-increasing spirals for that*".

Implementing CE to achieve sustainability has also been written into public policy. In 2015, the UN adopted the "2030 Agenda for Sustainable Development", which defines seventeen interconnected Sustainable Development Goals (SDGs) (UN 2015). In the European Commission's "Closing the loop" communication (European Commission 2015) the further global elaboration of CE is considered central to implementing the 2030 Agenda and a valuable tool for achieving the SDGs by 2030, with specific reference made to ensuring sustainable consumption and production patterns (SDG 12). This is reiterated in the new CE Action Plan (European Commission 2020a), which aims to accelerate actions required by the European Green Deal (European Commission 2019), itself an integral part of Europe's SDG implementation strategy (European Commission 2019). From their review of the potential for CE to facilitate achieving the SDGs, Valverde and Avilés-Palacios (2021) conclude that there is a positive qualitative relationship between them. Other countries that have adopted CE regulatory policy packages include, among others, China, Colombia, Japan, and South Korea (Fitch-Roy et al. 2021; Ogunmakinde 2019).

1.3 Critiques of the 'as advertised' CE concept

The CE concept is often criticized for ignoring existing knowledge, including some fundamental physics principles (Corvellec et al. 2021). In a perfect CE world, all waste streams would become secondary resources to transform conventional linear value chains into perfect circular systems. Lazarevic and Valve (2017) argue that some of the concept's most prominent advocates work from the unrealistic belief that *"an economy-wide perfect circle of perpetual fully closed material loops"* is actually possible. However, processes that facilitate transformations from linear to circular systems also consume materials and energy, generate residues and wastes, and emit pollutants. Cullen (2017) states that advocates of CE often ignore material loss and the additional energy required to sustain circularity, and by considering only material flow solutions, merely shift burdens to the energy domain through its assumptions about renewable energy deployment without considering energy system limitations. Furthermore, without using tools capable of quantifying the movement of minor elements in mixtures correctly, the quantities and qualities of the available secondary resources are more than likely overestimated. Together, all these deficiencies could paint a picture that looks considerably "rosier" than actual physical reality.

Recycling, for example, is the final step in the resource management hierarchy and should only take place once a product can no longer be kept in the life cycle system through any of the steps higher up in the hierarchy (e.g. through *reducing* consumption, *reusing* products or components, etc.) as per the Kirchherr et al. (2017) definition quoted above. From the resource efficiency (RE) point of view, however, recycling must eventually take place and is the step that ultimately determines the degree to which the EoL material loop can be closed to physically displace primary materials in the next life cycle. From a technical perspective, its ability to close loops depends on how effectively it can reverse the mixing that occurs when products are manufactured (largely based on material and product design), and the mixing of different products during EoL collection and during the recycling process itself, to maximise the *quantities* of materials extracted at the *qualities* required for the next life cycle. It also depends on the efficiency with which the material and energy resources needed to run these 'unmixing' processes are consumed (Reuter et al. 2019). Efficiencies can be optimised through process and technology improvements but can never reach 100%. One obstacle is the second law of thermodynamics (SLT), according to which real-world transformation processes can only occur in the direction of increasing entropy and an associated dissipation of exergy (i.e. a loss in the usefulness of energy) (Dincer and Cengel 2001). Entropy increases are caused by processes involving mass and heat transfer such as, inter alia, fluid flow through a flow resistance, heat flow through a thermal resistance, heat exchange, chemical reactions, diffusion processes, friction between surfaces, and also by mixing and dissolution processes.

Entropy and exergy are not subject to the conservation principle. So, while the first law of thermodynamics states that energy balances must always hold because energy can only be transformed and cannot be created or destroyed, the second law implies that any real process causes an irreversible decrease in the *quality* of energy—a portion of the total energy is always "downcycled" into heat from which no work can be extracted, representing an 'invisible' resource loss that cannot be recovered. Furthermore, material quantities and qualities are lost through dissipation, production of residues and wastes, and through contamination and mixing during extraction, production, and recycling processes. The only way to compensate for losses is to introduce virgin materials and energy from outside the system. However, the production of external resources also generates entropy and the dissipation of exergy, leading to downward spirals of continually increasing entropy and decreasing exergy, rather than perfect circles. In other words, it is physically impossible to achieve fully closed loops—irreversible losses are inevitable (Wollants 2014). A better approach might be to reduce the rate at which new materials are required by reducing demand (for both the services provided by products, and the amounts of materials needed to make products) and by reusing materials, components, and products (Allwood 2014).

Furthermore, circular approaches like EoL recycling would not occur simply because of the presence of recyclable materials, and it is not just physical recycling that determines the magnitude of virgin raw material displacement. Economically unattractive residues from life cycle systems are major sources of open loops (Reuter et al. 2019)—there needs to be an economic incentive for recycling, which is mediated by supply and demand dynamics (Geyer et al. 2016). Zink et al. (2016) found that, to maximise displacement, the ability of buyers to substitute between alternatives needs to be increased. One way to achieve this would be to improve the technical substitutability of recycled materials, e.g. through better sorting and recycling processes that enhance the quality of secondary resources (Ibid.). Business models that incentivise the return of EoL products and ensure recycling in compatible processes will, therefore, contribute to increasing displacement. The necessary technologies and infrastructure also need to exist, which depends on attracting the required investment. For these reasons, the one-to-one displacement of virgin materials by available secondary resources is highly unlikely. These themes are more often than not ignored in the general CE advocacy discourse and also applies to LCA, as most waste management LCAs assume one-to-one displacement of primary materials (Kara et al. 2022). Adding population growth and the increased demand for products and services, e.g. food, water, energy, housing, transport, and other consumer products it brings, it becomes clear that any claims of loops being fully closed or closable are inaccurate. This does not mean that circular flows would not have benefits, but such claims must be analysed and confirmed. Korhonen and colleagues highlight that, also in light of the entropy problem, those approaches higher up in the material management hierarchy than recycling should be prioritised. The authors argue, however, that the contribution of all CE-related activities to net global sustainability should be analysed in detail, as circular flows are not automatically guaranteed to deliver sustainable outcomes (Korhonen et al. 2018).

A subsequent challenge that arises is the detail level needed to analyse the flows of materials and energy. Cullen (2017) rightly states that *"What is required, is more detailed systems analysis of material and energy flows in linear and circular economies, and a deeper understanding of the practical limits to circularity imposed by thermodynamics and recovery processes. Without such analysis and insight, the CE vision risks becoming just another perpetual motion dream"*. Material flow analysis (MFA) is most often used and touted as the best tool for mapping resource flows (Graedel 2019). While suitable for bulk, single-element flows, MFA is inadequate when complex material and minor element combinations such as those found in PV and other technologies need to be traced through processes, as it does not consider solution chemistry and cannot predict the distribution of minor elements between process outputs (Reuter et al. 2019). These limitations hamper its use in prospective assessments when not integrated with high-detail approaches like process simulation (Baars et al. 2022).

In summary, while the concepts are promising, the effects of any taken or planned actions can only be appraised if rigorous, physics-based methods are used to assess baseline and/or future life cycle performance. The methods used to analyse material and energy flows through life cycles must be able to handle the complexity of the system at hand. While methods for assessing performance in the individual sustainability dimensions are established, their integration for more holistic sustainability assessments is only now gaining more traction. However, as highlighted by Roos Lindgreen et al. (2020), the connections between CE and sustainability, more specifically the environmental, economic and social impacts of CE at the micro level, are "fuzzy" and have yet to be delineated. There is a need for methodologies that quantify and demonstrate the interactions between circular approaches and sustainability in all its dimensions.

1.4 Minerals and metals from geological and urban mines

Complex combinations of various precious and specialty metals and other materials, even if in exceedingly tiny amounts, allow most technologies used today to perform specific functions. The combinations refer to mixtures of alloys, pure metals, compounds, plastics, and other materials, whether chemically bonded or, for example, glued, riveted or bolted together (UNEP 2013 p.63). This includes household electronic devices and also the technologies that allow us to harness and store renewable energy. A smartphone, for example, typically contains up to forty different metals. Wind turbines make use of neodymium magnets (a rare earth element). Between first-generation crystalline silicon (c-Si), second-generation cadmium telluride (CdTe), and third-generation perovskite-based PV systems, the elements used include, among others, aluminium (Al), boron (B), cadmium (Cd), caesium (Cs), copper (Cu), indium (In), lead (Pb), phosphorous (P), selenium (Se), silicon (Si), silver (Ag), tellurium (Te), and tin (Sn) in various combinations to be able to generate renewable electricity. Lithium-ion batteries, to name but one of many types, use Al, cobalt (Co), Cu, graphite, lithium (Li), manganese (Mn), and nickel (Ni), among others.

The production and EoL treatment of these technologies depend on the availability of the required metals and other materials, and the infrastructure needed for their production, supply, and recycling, which is achieved through complicated networks of physical separation, minerals processing, and pyro- and hydrometallurgical processes. These processes require various external material and energetic inputs and are connected to one another through supply chains that are often geographically dispersed. The extent to which metals and other materials are mixed[2] strongly influences the ability to separate them at EoL and hence, the extent to which their material loops can be "closed" from a technical perspective, as alluded to in the previous section. Therefore, product design and the design, optimisation and operation of production and recycling processes are central to optimal recovery and waste reduction and rely on in-depth knowledge of metal and compound properties, and their interactions during processing (Reuter 2016). Treating any of the metals or materials as if they flow individually in parallel rather than in their mixed state must be avoided by adopting a product-centric approach that considers all the materials in their combined form (Reuter and Kojo 2012).

1.5 By-product metals

A further complication is that many of the metals used in modern technologies are by-products[3] of other metal production systems (Bleiwas 2010). Te, for example, is primarily produced as a byproduct of Cu—only if the so-called anode slimes residue from the last step in the production and refining of Cu is processed further can Te be produced. Similarly, Cd and In mostly originate from the zinc (Zn) system, to name but a few examples. These metals are generally present in small quantities in minerals compared to their carrier metals, usually making their independent extraction economically unjustifiable (Fortier et al. 2018). Their supply is, therefore, dependent on the market dynamics surrounding their carrier metals, which could lead to significant price fluctuations when production cannot respond quickly enough to sudden changes in demand (Nassar et al. 2015).

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[2] In addition to their conventional meanings, the terms *mixed* and *mixture* in this dissertation refer to the way materials and components are connected to each other to manufacture a product, be it through nuts and bolts, welding, chemical bonding, gluing, molecular deposition methods, or others. That is, finished products can also be seen as mixtures of materials, which, at EoL, become "urban minerals", analogous to geological minerals.

[3] The terms *by-product* and *co-product* are used interchangeably for the purposes of this work.

Manufactured urban minerals can contain even more complex combinations of metals and other materials. Urban 'mineral deposits' of electronic waste can contain in excess of fifty interconnected metals and other compounds (Van Schaik and Reuter 2010; Reuter et al. 2019). Depending on the intended use, inefficiencies during collection and sorting, mechanical separation processes, metals ending up in incompatible recycling processes, or simply the thermodynamic limits of extraction processes necessitate the dilution of impurities with virgin materials to meet product quality specifications, as mentioned earlier. For this reason, primary resource extraction cannot be eliminated completely. Ignoring the effects of mixing and dilution would lead to an underestimation of the quantities of materials and energy needed for high-quality recycling processes. Even bulk metals like steel and Al, often described as being infinitely recyclable, cannot be recycled without quantity and quality losses (Reuter et al. 2019).

1.6 Critical raw materials

Further adding to the importance of making a concerted effort to recover these minor metals is the fact that a number of them are considered critical raw materials (CRM). As defined by Schrijvers et al. (2020), the study of raw material criticality "… *evaluates the economic and technical dependency on a certain material, as well as the probability of supply disruptions, for a defined stakeholder group within a certain time frame*". Several factors are considered in the methodology used to calculate economic importance and supply risk indicators for raw materials in the European Union (EU). Detailed descriptions are beyond the scope of this work and can be found in Blengini et al. (2017). Among the purposes of the European CRM list are that it serves to support trade policy development, to guide investment decisions, to guide research and innovation in new technology, substitution and recycling, and to promote responsible and sustainable sourcing (European Commission 2020b).

CRM lists are dynamic—the latest iteration of the European Commission's three-yearly update includes thirty materials or material groups[4] (Ibid.), of which at least a third can be considered by-product metals (cf. Nassar et al. 2015). Recycling is a crucial tool for countering criticality as it could complement primary production and reduce the risk of supply disruptions (Tercero et al. 2020). This depends, however, on the availability of recyclable materials, compatible recycling processes and infrastructure, and favourable economics for the products of recycling (UNEP 2013; Reuter et al. 2019). The ability to separate the metals and other materials of interest from the mixtures in which they are present, is crucial.

[4] The groups referred to are the light and heavy rare earth elements (REE), and the platinum group metals (PGM).

1.7 Solar photovoltaics

In its communication on tightening Europe's climate change mitigation targets to reduce emissions by 55% compared to 1990 levels by 2030 (the so-called "Fit-for-55" package), the European Commission again highlights the importance of renewable energy development and deployment to realizing the European Green Deal and achieving climate neutrality by 2050; as part of this plan and Europe's role in promoting climate neutrality globally, it aims to create alliances that support equal opportunities around several sustainable technologies—among others, green hydrogen, solar, wind, batteries and carbon capture—as well as the CRMs needed for these technologies (European Commission 2020c). The contribution PV is required to make towards achieving this target translates to the annual deployment of at least 21-22 GW_{AC} of PV capacity per year in the EU until 2030 (Kougias et al. 2021). The EU and UK deployed 16.8 GW_{DC} of PV capacity in 2019 and 19 GW_{DC} in 2020 (IEA 2021). Besides its role in limiting the impacts of climate change, a transition to renewable energy supports CE aspirations—fossil-based energy generation consumes substantial amounts of materials that cannot be recycled. In contrast, renewable energy generation does not, thus contributing to minimising resource consumption and waste generation.

The COVID-19 pandemic and the war in Ukraine have caused significant downturns in economic activity, also strongly affecting the energy sector (IEA 2020; acatech/Leopoldina/Akademieunion 2022). However, Europe's economic response to the pandemic is seen as an opportunity to accelerate climate action and digitalization (European Commission 2020c). At the peaks of the pandemic, energy demand in developed countries decreased by 25%, coinciding with a shift from energy consumption for transport to increased electricity demand during lockdowns, a further incentive for direct electricity generation from solar PV (Rządkowska 2020). With the current focus on reducing or eliminating dependence on Russia for energy, PV and other renewable energy technologies are even more relevant, and their development and deployment even more critical. The war has pushed governments into re-evaluating the rapidity with which renewable energy technologies need to be deployed and as mentioned, PV has a significant contribution to make in future energy mixes.

First-generation solar PV technologies, based on silicon (Si) wafers, dominate the PV market with its 95% share (VDMA 2021) and is expected to continue to do so over the next decade and beyond. The raw materials needed to produce metallurgical grade Si (MG-Si) are relatively abundant, but the purity of MG-Si (typically < 99.5% Si) does not meet the minimum requirements for solar applications. Additional, often energy-intensive, processing is required to refine MG-Si into solar grade Si (SG-Si) with a purity of 99.9999% Si (also referred to as *6N*) (Chigondo 2018), making solar and higher grades of Si metal relatively less abundant than MG-Si. Whereas the cost of raw silica (SiO_2) from which Si metal is derived is less than 5 cents/kg, it is not surprising that the cost of SG-Si can be around 10 US$/kg or more (Simandl et al. 2021). In fact, it reached 28.5 US$/kg in June 2021, while 6.3 US$/kg a year prior (NREL 2021). While Si metal is not a by-product of another system, it is on the European CRM list (European Commission 2020b). Second-generation technologies are of the thin-film type and include, for example, cadmium-telluride (CdTe), copper-indium-gallium-selenium (CIGS), and amorphous Si (a-Si) cells deposited on glass substrates, as opposed to first-generation Si wafers. Between CdTe and CIGS, five of the six metals—all but Cu—are by-product metals. Used in a-Si cells, Germanium (Ge) is also a by-product metal. Furthermore, Ga, Ge and In are CRMs in the EU (Ibid.), while Te is added on the USA's list (Graedel et al. 2022). Third-generation technologies include various newer developments, among which are organic lead (Pb) halide perovskite cells, as well as various multi-junction and tandem devices that aim to overcome the Shockley-Queisser theoretical power conversion efficiency (PCE) limit of just over 33% (Shockley and Quiesser 1961) for conventional single-junction solar cells. In a four-terminal tandem configuration, two independently manufactured sub-cells are stacked on top of each other and can be operated independently to maximize performance (Leccisi and Fthenakis 2020).

Various other materials used in PV modules also contain by-products and CRMs. These include, for example, transparent conductive oxide layers such as indium-tin oxide (ITO), which is a solid solution typically consisting of 90% indium(III)oxide (In_2O_3) and 10% tin(IV)oxide (SnO_2), and the tin-coated Cu ribbons used as connectors. Tin (Sn) appears on the USA's CRM list (Graedel et al. 2022). A summary of the most important PV-relevant metals is shown in Figure 1.

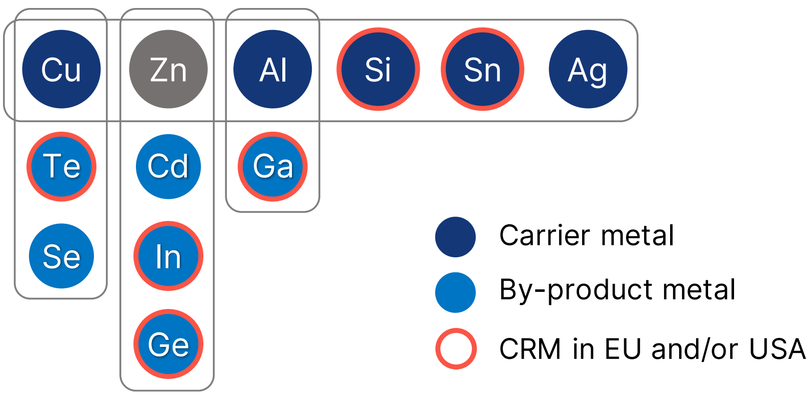

Figure 1. Carrier, by-product, and critical metals relevant to PV. Although aluminium is not listed as a CRM in Europe, the mineral bauxite from which it is extracted, is.

Because they contain numerous by-product and critical materials, PV cells and modules are referred to as *complex products* in this work. It further implies that EoL PV modules are complex urban minerals that contain several important secondary resources, if recovered. The deployment growth required between now and 2030, and onwards to 2050 and beyond will result in several-fold increases in demand for many of the minor metals used in PV (Davidsson and Höök 2017; Carrara et al. 2020). For Si-based PV recycling to support CE, Heath and colleagues recommend, among others, prioritising the development of recycling processes and purification methods that produce high-grade recyclates (as opposed to typically MG-Si recyclates), reductions in material intensity by, e.g. reducing wafer thickness and manufacturing losses (Heath et al. 2020). The authors also recommend using "anticipatory, systems-based analytical tools" for evaluating the economic-environmental trade-offs that might exist in the Si PV life cycle, and to inform recycling process design (Ibid.).

The PV case represents the confluence of several themes—the use of renewable energy to decarbonise the energy sector, the use of both by-product metals and CRMs in a technology that, at the same time, drives sustainability, the need for sophisticated recycling processes and infrastructure capable of separating several minor elements to prevent the dissipation of CRMs and other important and/or detrimental materials, and CE approaches, business models, incentives, and legislation that support such initiatives. Crucial to measuring baseline sustainability performance, or the potential performance of future systems, and to guiding development towards sustainability is being able to rigorously quantify the flows and transformations of materials and energy along the life cycles of complex products like PV, taking into consideration the aforementioned aspects in as much detail as is practicable. The resulting mass and energy balances form the basis of all sustainability and circularity assessments.

1.8 Knowledge gaps identified

1.8.1 Methodological gaps

As highlighted in Section 1.2, the method most commonly used to compile inventory data for product life cycles, MFA, inadequately quantifies the flows of materials and energy in systems that contain mixtures of by-product and critical metals, and it does not consider the SLT. Material and energy flows in these systems need to be analysed using more sophisticated, physics-based tools.

Secondly, the resource, environmental, and economic performance of emerging technologies are typically assessed in isolation with methodological challenges arising in subsequent attempts to integrate the two dimensions. The integration of assessments can be done in diverse ways and to various degrees (Wunderlich et al. 2020) but methodological challenges surface due to inconsistent system boundaries and functional units, and different assumptions when attempting to integrate standalone assessments (Mahmud et al. 2021).

There is a gap with respect to methodologies capable of analysing environmental and techno-economic performance simultaneously, while allowing for the effects of variations in process parameters to be investigated (Ibid.). Integrated assessments that make use of consistent inventory data are needed to circumvent these challenges. A potential hindrance to achieving the required depth of inventory modelling is the complexity of the system being analysed.

Lastly, with all the purported positive aspects and critiques surrounding CE, there is a need to establish whether circular interventions do, or do not, contribute positively to sustainability, and to what extent. Especially for established and emerging complex technologies, rigorous, quantitative, physics-based methods are needed to cut through the CE marketing "buzz" and to examine the relevance and significance of the critiques described earlier. To achieve this, resource flows, environmental impacts, economic performance, and circularity need to be linked. Moreover, by examining sustainability trends over ranges of circularity, as opposed to assessing single operating points, the contribution of circularity to environmental-economic trade-offs can be evaluated.

1.8.2 PV system performance assessment gaps

Efficiency improvement and cost reduction are the general foci of PV development efforts. From a sustainability perspective, however, resource efficiency and environmental (and social) impacts also need to be considered simultaneously. While numerous LCA and TEA studies have been published for a wide variety of PV types, integrated environmental-economic assessments are lacking in the published literature. As mentioned above, attempts to integrate standalone assessments are usually met with challenges. For PV systems in particular, this is exacerbated by the large number of, inter alia, configurations, materials, efficiencies, locations, and production methods used in published studies, especially for the emerging next-generation technologies.

The potential resource and environmental benefits of EoL recycling have been described for a limited number of PV systems at fixed operating points. However, assessments of the simultaneous effects of EoL recycling as a circularity strategy on the resource, environmental, and techno-economic performance of PV systems to identify potential sustainability trade-offs have not been published.

Furthermore, there are, to the best of the author's knowledge, no published studies that combine resource, environmental, and economic performance, and model closed-loop recycling in the foreground system to avoid having to make rather arbitrary, and sometimes counter-intuitive, methodological choices when a product reaches EoL.

1.9 Objectives and research questions

Based on the knowledge gaps identified, the overall objective of this dissertation is to explore and quantify the links between circularity and sustainability[5] for the life cycles of complex[6] technologies by advancing approaches for the sustainability assessment of CE concepts. The application-oriented objective is to generate physics-based inventory data for renewable energy systems, as these are indispensable for sustainable development, but themselves also have sustainability impacts. The foundation of such assessments is the rigorous quantification of resource (material and energy) flows into, within, and out of the life cycle system. Circularity also implies that the analysed life cycles contain circular flows. In this context, *rigorous quantification* refers to doing so in a manner that prioritises physics-based relationships over linear transfer coefficients when assessing the flows of materials and energy through the numerous processes that constitute the life cycle. The *physics-based relationships* refer not only to compliance with the law of mass conservation, but also to taking into account energy conservation (i.e. the first law of thermodynamics) and the changes in enthalpy, entropy, and hence, the free energy (according to the SLT) of chemical and metallurgical transformation processes, as described in Section 1.1.2. The resource flow data generated in this way is subsequently used to assess the environmental and techno-economic performance of the life cycle, which requires exploring methods and indicators used to quantify each of these dimensions and combining them to conduct fully aligned, integrated assessments that consider both sustainability and circularity. As previously described, the complex technology selected for this dissertation is *solar photovoltaics* due to its high relevance in society today.

To achieve the overall objective, three research questions (RQs), each building on the previous, have been formulated as follows:

1. Which methods can be applied and combined to analyse the resource efficiency, environmental impacts, and techno-economic performance of complex technology life cycles, considering the laws of conservation and the SLT to produce physics-based mass and energy balances?

[5] It is acknowledged that the term *sustainability* refers to all three of its dimensions—environment, society, and economy. As social impact assessment is beyond the scope of this dissertation, the term here refers to environmental and techno-economic aspects.
[6] In this dissertation, a *complex* technology refers to one whose functionality is provided by several special metals, some of which are CRMs and/or by-product metals, typically used in small quantities and highly interlinked. This applies to most electronic products, including renewable energy generation technologies such as PV.

2. What are the combined quantitative effects of circularity on the resource, environmental, and techno-economic performance of established and emerging PV systems?
3. Does EoL circularity contribute to the sustainability of established and emerging PV technologies?

Each of these questions are addressed in one or more of the four published first-author journal articles that this dissertation is based on. Article details and the questions addressed are given in Table 1. Other associated publications not included in this dissertation are listed in Table E-1 in Annexure E.

Table 1: Details and RQs addressed in each of the journal articles accompanying this dissertation

Article no.	Reference		RQs addressed 1	2	3
1	Bartie, N. J., Abadías Llamas, A., Heibeck, M., Fröhling, M., Volkova, O., & Reuter, M. A. (2020). The simulation-based analysis of the resource efficiency of the circular economy – the enabling role of metallurgical infrastructure. Mineral Processing and Extractive Metallurgy, 129(2), 229–249. https://doi.org/10.1080/25726641.2019.1685243		●		
	Impact factor	1.74			
	Author's contribution	Conceptualization and identification of CdTe PV as an appropriate case study, development of the simulation models for Cd, In, Pb, and Zn production. Article: methodology; model development, validation, formal analysis, investigation; writing of complete original draft, review, editing, visualisation.			
2	Bartie, N. J., Cobos-Becerra, Y. L., Fröhling, M., Schlatmann, R., & Reuter, M. A. (2021). The resources, exergetic and environmental footprint of the silicon photovoltaic circular economy: Assessment and opportunities. Resources, Conservation and Recycling, 169, 105516. https://doi.org/10.1016/j.resconrec.2021.105516		●	●	○
	Impact factor	12.68			
	Author's contribution	Conceptualisation and identification of opportunities for Si circularity, development of all simulation models, creation of NN-based surrogate models, generation, and analysis of all results. Article: conceptualization, methodology, software, validation, formal analysis, investigation, writing – original draft, visualisation, editing.			
3	Bartie, N., Cobos-Becerra, L., Fröhling, M., Schlatmann, R., & Reuter, M. (2022). Metallurgical infrastructure and technology criticality: the link between photovoltaics, sustainability, and the metals industry. Mineral Economics, 35(3-4), 503–519. https://doi.org/10.1007/s13563-022-00313-7		●	●	○
	Impact factor	2.48			

Article no.	Reference		RQs addressed 1	2	3
	Author's contribution	Conceptualisation of additional analyses (resource efficiency comparisons, material intensity, carbon taxation), development of bottom-up cost model. Article: conceptualisation, methodology, software, validation, formal analysis, investigation, writing—original draft, visualisation, editing.			
4	Bartie, N., Cobos-Becerra, L., Mathies, F., Dagar, J., Unger, E., Fröhling, M., Reuter, M. A., & Schlatmann, R. (2023). Cost versus environment? Combined life cycle, techno-economic, and circularity assessment of silicon- and perovskite-based photovoltaic systems. Journal of Industrial Ecology, 1–15. https://doi.org/10.1111/jiec.13389		●	●	●
	Impact factor	7.81			
	Author's contribution	Conceptualisation, development of simulation models and bottom-up cost models for all PV systems, creation of NN-based surrogate models, generation, and analysis of all results. Article: conceptualization, methodology, software, validation, formal analysis, investigation, writing – original draft, visualisation, editing.			

○ Partial

1.10 Dissertation outline

This chapter has introduced a number of key themes pertaining to sustainability and the CE concept, the use of metals in complex technologies, and the role of PV in the transition to sustainable energy systems. Knowledge gaps have been identified and the overall objective with associated RQs formulated. The key methods used to address the RQs during the course of this work are described in Chapter 2. Chapters 3, 4, 5 and 6 provide key information on method development and findings from Articles 1, 2, 3, and 4, respectively. While key results and findings are discussed in the main text, readers are encouraged to peruse the full publications in Annexures A, B, C, and D, respectively, for more comprehensive information. How each of the RQs have been addressed, novelty aspects, implications of the work, and limitations are addressed in the Discussion in Chapter 7. The dissertation is concluded, and future work proposed, in Chapter 8.

2 Methods

As stated in Section 1.1, potential environmental impacts are usually estimated using the standardised LCA methodology, and economic performance using TEA and/or LCC, which are established and widely applied approaches. At the core of these assessment approaches is the 'inventory' of the system being investigated, which refers to the flows of materials and energy within and through the life cycle. Various methods are available for the compilation and analysis of inventory data. Two of these, material flow analysis (MFA) and process simulation are described in Section 2.1 in the context of analysing complex product life cycles. The application of LCA to estimate potential environmental impacts and carbon footprints is described in Section 2.3, and the application of TEA in Section 2.4. The use of neural-network-based surrogate functions to enhance computational efficiency is described in Section 2.5. As social impact assessment is not within the scope of this work, it is not discussed in detail in the remainder of this dissertation.

2.1 Quantification of material and energy flows in complex product life cycles

2.1.1 Material flow analysis

The most commonly applied and cited method for mapping life cycle inventory is MFA. In essence, it refers to analysing the mass flows through a life cycle in compliance with the law of mass conservation. MFA aims to evaluate relationships between physical flows, socioeconomic activities, and environmental changes, and is applied over wide ranges of detail and completeness depending on the purpose of the investigation (OECD 2008), and mostly on the national or regional level (Gößling-Reisemann 2008a, 2008b). It is used to contribute to environmental policy development, analyse resource use patterns, and to provide estimates of recycling potential and losses to the environment (Chen and Graedel 2012). Stocks are determined using top-down (difference between in- and outflows) or bottom-up approaches (summing actual stocks of relevant materials at the time of interest), the trade-off between the two depending on precision requirements and data availability (Gerst and Graedel 2008). In general, however, MFA aims to reduce the number of substances studied to preserve manageability and clarity (Brunner and Rechberger 2017).

While suitable for bulk, single-element material flows, MFA cannot predict the distribution of minor elements between process outputs when the system contains material and minor element combinations such as those found in PV and other technologies, as it does not consider solution chemistry and thermodynamics (Reuter et al. 2019). In their analysis of mobile phone recycling, as another example of a complex product, Valero Navazo et al. (2014) highlight difficulties in allocating material and energy savings to precious metals arising from the fact that these metals are produced as by-products of others. As mentioned earlier and depicted in Figure 1, this would also be the case for all the PV technologies investigated in this work. Similarly, where materials or substances are embedded, and assumptions must be made about the composition of goods, it becomes challenging to determine resource efficiency at the micro level (OECD 2008). Bollinger and colleagues argue that, while methods like MFA can give insight into potential future material flows, their aggregated nature ignores the fact that materials are often packaged into discrete products, the properties of which influence the flow patterns of the materials within them (Bollinger et al. 2011). These are functions of product design and the chosen processing route, and directly impact the recovery of materials and energy (Reuter 2016; Reuter et al. 2019). It is, therefore, recommended that MFA be used within a wider framework of tools to identify those most suited to the required level of detail (OECD 2008). Allesch and Brunner (2015) recommend further assessments like risk or entropy/exergy analyses and Chancerel et al. (2009) recommend combining MFA with engineering simulation tools to counter the limitations of lower detail levels in MFA.

2.1.2 Process simulation and resource efficiency

It is standard practice for process engineers to employ process modelling and simulation tools to design and optimise production processes, and to construct multi-level recycling models (UNEP 2013). Process models aim to mimic the behaviour of real systems to provide mass and energy flow data at a detail level that would allow for equipment sizing to be done and for bankable feasibility studies to carried out, the goal being the eventual construction of a real chemical or metallurgical plant. Methodological reliability, therefore, is essential. In contrast to the more aggregated methods like MFA, the relationships that describe how the inputs into a process step are transformed into its outputs are not automatically assumed to be linear when developing a process simulation model. In addition to implicitly performing MFA, process simulation makes use of comprehensive thermodynamics databases to take the enthalpy, entropy, and thus, free energy of all compounds and solutions into account, which is necessary to resolve complex mass and energy balances. The free energy of a process, such as the production of a certain metal or the recovery of a certain element from a solution indicates whether the process is possible at all. If so, the amount of energy transformed into entropy, which can never be zero, indicates the thermodynamic limits of the process, as this part of the energy flow can never be recovered.

Models are built by first creating, for each individual process step, a model that describes the transformation process that occurs in that unit. Individual process steps refer to, for example, a furnace in which a Cu-sulphide concentrate is smelted to produce a high-grade Cu matte phase for further downstream processing to produce Cu, or a distillation column used to separate individual substances from a mixture based on their different boiling points. Transformation processes can be defined based on, among others, chemical reactions, thermodynamic equilibrium relationships, reaction, and mass/heat transfer rate correlations, and known industrial operating conditions, all underpinned by closed mass and energy balances. Individual steps are then connected using material and energy flows to represent specific production or recycling processes they are part of. The larger processes can then be connected to represent entire value chains (Bartie et al. 2021a). The HSC Chemistry software package (mogroup.com), in particular its process simulation module, is used for the work presented in this dissertation. It is a sequential modular simulator, which, as the name suggests, iteratively solves the predefined process units one at a time and in sequence, taking into account how the units interact with one another (e.g. via the material flows between them) and with the environment (e.g. purchased electricity, fuels, and reagents).

A good simulation model can provide element- and compound-level mass, energy, and thermochemical information for gate-to-gate industrial processes, and for complete life cycles. Utilizing the law of mass conservation and both the first (energy conservation) and second (increasing entropy) laws of thermodynamics, it can be used to evaluate compliance with these laws, account for the complexity of transformation processes that include multiple minor elements, and highlight the technical limits of production and recycling processes that result from irreversible losses (i.e. entropy creation). This gives it predictive capabilities, which is especially useful in the context of prospective assessments in which mass and energy flows cannot be drawn from historical data but need to be predicted (Bartie et al. 2023).

Transferring these capabilities into the sustainability assessment field makes it possible to, for example, quantify and allocate emissions to the correct output flows, to identify consumption and environmental impact hotspots, to maximise the recovery of materials and energy, and to minimise losses and the creation of entropy (Reuter 2016). For complex products, this is the minimum level of detail required to characterise process and recyclate streams, their contribution to improving RE and their impacts (UNEP 2013). Simulation results can be used to generate up-to-date, physics-based inventory data for both existing and potential future life cycle systems, which can then be used to rigorously calculate various resource efficiency, sustainability, and CE performance indicators. That is, it can more accurately quantify the technical boundaries within which CE must be adopted by setting constraints for the quantities and qualities of physical flows. In this dissertation, *material recovery rates* are used as the indicator of material resource efficiency. *Exergy efficiency* is proposed as a proxy for the overall, combined material and energy efficiency as it is based on the SLT. Similarly, *exergy cost*, which represents the cumulative amount of exergy dissipated to produce a product (Lozano and Valero 1993), is proposed as a proxy for the combined material and energy resource consumption. The thermoeconomics calculator in the HSC Chemistry simulator, based on the *theory of exergetic cost* (Ibid.), simplifies exergy cost calculations.

An indicator specifically used to measure the resource efficiency of electricity generation systems, the energy return on investment ($EROI_{PE\text{-}eq}$) is used to evaluate the efficiency with which the PV systems again produce all the energy that was harvested for their production. The 'PE-eq' subscript refers to 'primary energy equivalent', meaning that the EROI is calculated in terms of the primary energy consumption, and the primary energy equivalent energy delivered, rather than simply the electricity consumed, and electricity generated (Raugei et al. 2016).

The greatest challenges with the development of process simulation models are its resource and data intensity. Like any modelling exercise, process simulation is also subject to the "garbage in = garbage out" adage. Good simulation models take time to develop and rely on, inter alia, experience and in-depth knowledge of chemical/metallurgical processing options and limits, and high-quality input data (Verhoef et al. 2004; Hagelüken 2006; Reuter 2016). Breun and colleagues argue that the significant effort required for the parameterization of process simulation models can limit their application to smaller systems (Breun et al. 2016). The availability of industrial data for model validation can be problematic as most operators are reluctant to share their proprietary data. If fundamental data do not exist, first-principles modelling or experimental studies (e.g., Van Schalkwyk et al. 2018) are required to expand existing thermodynamic databases. It is almost inevitable, however, that not all relevant data will be available and that assumptions will need to be made—it is important for these to be realistic, confirmed through own experience or that of experts in the field. The direct linking of related simulation models could increase complexity and result in computational issues (Fröhling et al. 2012, Porzio et al. 2013). On the plus-side, once initially created, a simulation model can be adapted to alternative or prospective scenarios with relative ease (Casavant and Côté 2004; Bartie et al. 2023; Fröhling et al. 2010; Fröhling et al. 2012).

2.2 Quantification of environmental impacts

LCA is a standardised and widely used methodology for the estimation of the potential environmental and human health impacts of products or services along all life cycle stages under investigation. Among others, LCA can assist in identifying environmental performance improvement opportunities, identifying and avoiding problem shifting between life cycle stages, between environmental problems or between regions, aid in the selection of environmental impact indicators, and provide a means to communicate environmental impact information (ISO 2020a; Finnveden et al. 2009). LCA results are usually reported in terms of so-called problem-oriented midpoint impact categories and/or the damage-oriented endpoint categories. The former includes, e.g. climate change, stratospheric ozone depletion, acidification, eutrophication, human toxicity, ecotoxicity, photochemical ozone formation, water use, land use, particulate formation, ionising radiation, resource use, and others, and the latter refers to the potential damage done to human health, the natural environment or ecosystem quality, and natural resources (Rosenbaum et al. 2018). The midpoint indicators all contribute to the smaller set of endpoint indicators further down the cause-effect chain, the link between them represented by endpoint characterisation factors obtained through further modelling (Ibid.). For instance, climate change midpoint impact contributes to endpoint damage to human health (e.g. through heat-related illnesses) and ecosystem quality (e.g. through loss of habitat). These can be quantified, for example, in terms of disability-adjusted-life-years (DALY) and potentially-disappeared-fraction (PDF) of species, respectively (Matthews et al. 2014).

Distinction is made between two modes of assessment - attributional and consequential. Attributional LCA is used most often and focuses on the analysis of relevant environmental flows to and from the life cycle under investigation, while consequential LCA considers the wider effects of changes in these flows, e.g. on systems outside of the investigated life cycle, resulting from the selection of one alternative rather than another (Finnveden et al. 2009; Hauschild 2018). To enable direct comparisons of PV systems, the attributional approach is applied in this work.

The ISO standards (ISO 2020a, 2020b) specify four phases in the LCA process, namely goal and scope definition, the life cycle inventory analysis (LCI), life cycle impact assessment (LCIA), and the interpretation phase that runs in parallel with the first three. A crucial step during goal and scope definition is to clearly define the boundaries of the system for which the assessment is being done, as it directly impacts the scope and detail needed for the LCI phase. The 'functional unit' refers to the function performed by the life cycle being assessed, e.g. the delivery of 1 kWh of PV-generated electric energy.

Distinction is also made between the foreground and background systems. The foreground system is modelled in detail, mainly using primary data (Bjørn et al. 2018) if available, or if it can be collected or generated. The background system contains processes not specific to only the life cycle at hand and is usually modelled using environmental databases that contain region-specific average industry data for many processes (Ibid.), such as the supply of heat and electricity, and the production of numerous metals, chemicals, and whole products. To model the background system in this way, one needs access to LCA software (some of which are free of charge) and access to one or more of the environmental databases, the most popular and complete databases being rather costly. The collected data need to be compiled for analysis and to close the mass and energy balances, which is where the methods described in Section 2 (MFA and process simulation) come in. Naturally, the quality of an LCA depends on the quality of the inventory data generated using the chosen method. As stated, process simulation is, or should be, the method of choice when the subject of an assessment is a *complex* product.

Terminology from the Greenhouse Gas (GHG) Protocol, developed to guide organisations in quantifying their climate change impacts, provide useful classifications of emissions, and are adopted in this work. *Scope 1* emissions refer to direct emissions that result from the running of production and recycling processes, such as the GHGs produced in an incineration process without carbon capture. Scope 1 emissions would, therefore, generally originate from the foreground system. *Scope 2* emissions refer to the indirect emissions resulting from the production of any purchased and consumed electricity, steam, heat, or cooling (Greenhouse Gas Protocol 2015). *Scope 3* emissions are the upstream and downstream indirect emissions associated with acquired materials, and product distribution, storage, use, and EoL (Greenhouse Gas Protocol 2011). Scope 2 and 3 emissions, therefore, originate from the background system and is usually quantified by utilising an LCI database. However, if any of the aforementioned life cycle stages like EoL, for example, are included in the foreground system model, its emissions fall under Scope 1.

2.3 Techno-economic performance of PV systems

Both TEA and LCC offer tools for systematic assessment of the economic viability and/or performance of projects and product life cycles. Whereas TEA usually reflects the perspective of a specific stakeholder in the life cycle, LCC includes all costs linked to the life cycle, irrespective of which actors are paying for them (Moreau and Weidema 2015) and includes less tangible, hidden costs, and economic benefits of pollution controls (Klöpffer 2003). LCC is compatible with the standardised LCA structure and several authors stress the importance of using consistent functional units and system boundaries when conducted in combination with LCA (Klöpffer 2003, 2008; Swarr et al. 2011). While the PV-specific indicators selected for this dissertation (LCOE and MSP, described below) are considered techno-economic indicators, they are calculated from an LCC perspective and include costs associated with EoL circularity.

The initial inputs into a rigorous TEA would entail mass and energy balance data such as that generated using process simulation (described in Section 2.1.2). In industrial process design and cost estimation settings it would be the absolute minimum level of detail required. The maturity of a technology, commonly expressed in terms of its technology readiness level (TRL), typically determines how much data is available and at what level of detail (Buchner et al. 2018). By the time higher TRLs are reached, a significant amount of information is generally available, but technologies are challenging and expensive to modify; at the lower TRLs, less data are available, but modifications would be relatively easier (Thomassen et al. 2019). Therefore, the ability to predict mass and energy inventory data for emerging technologies is advantageous.

2.3.1 Levelized Cost of Electricity

The levelized cost of electricity (LCOE), also referred to as the levelized cost of energy in more general terms, is a useful indicator of the economic performance of power generation technologies, and a decision support tool when comparing technologies or comparing variations of the same technology. It is, therefore, also commonly used for the technoeconomic assessment of PV systems, for comparison of PV systems with other renewable and non-renewable energy sources, and for comparison of distinct types of PV, different material choices, manufacturing efficiencies and technologies, PCEs, and numerous other variables. The LCOE represents the minimum average price of electricity necessary to compensate for the cost of the system and is calculated as the ratio of the total investment in such a technology over its lifetime and the total amount of energy delivered by that system over its lifetime, as shown in Equation 1 (Sofia et al. 2019).

$$LCOE = \frac{I_{system} + \sum_1^n \frac{O\&M}{(1+r)^n}}{\sum_1^n \frac{E(1-d)^n}{(1+r)^n}} \tag{1}$$

where I_{system} is the initial investment for PV system installation (including modules, balance-of-system components, labour, permitting, and others), *O&M* the annual operation and maintenance cost, n the system lifetime, E the energy yield in the first year, r the nominal discount rate, and d the annual degradation rate. In this study, the initial investment (except for module cost) and *O&M* costs were estimated using recently published breakdowns of typical area- and power-related costs (Zafoschnig et al. 2020) in Europe. The energy yield (E) depends, among others, on the solar insolation at the location of the system and its performance ratio (PR), which refers to the ratio of actual energy output and its nominal output (i.e. design output). It takes into consideration losses that occur due to, e.g. temperature, peripheral equipment inefficiencies, shade, soiling, snow, and others.

2.3.2 Minimum sustainable price

To calculate the full initial investment for an installed PV system, a module price is needed. Powell and colleagues recommend using the minimum sustainable price (MSP) for this purpose given its financial sustainability perspective (Powell et al. 2013). In this work, it is calculated using bottom-up cost models that consider the capital and operational expenditures (capex and opex, respectively) of module manufacture. The discounted cash flow (DCF) and net present value (NPV) approaches are used to estimate the MSP, which is the minimum module price at which the manufacturer remains profitable and generates expected investment returns. The DCF considers the time value of money, in other words, the fact that the buying power of a given amount of money decreases over time. To calculate DCF, estimated annual cash in- and outflows are projected over the life of a project, after which all the future cash flows are converted to their present-day equivalents using the weighted average cost of capital (WACC) because it takes return expectations into account (Graham et al. 2014). Recently published WACC values by industry for the period 2020/2021 based on 332 companies in Germany, Switzerland, and Austria include, e.g. 4.9% for Energy and Natural Resources, 7.5% for Industrial Manufacturing, and 6.6% overall (KPMG 2021). The NPV is found by subtracting the initial investment from the DCF. These steps are represented by Equation 2 (Zweifel et al. 2017).

$$NPV = -I_{p+r} + \sum_{n=1}^{n} \frac{CF_n}{(1 + WACC)^n} \tag{2}$$

where CF_n is the estimated net cash flow in year n and I_{p+r} is the initial investment. A project is usually considered feasible when the NPV is equal to or greater than zero. When it is zero, the present value of all future cash flows breaks even with the initial investment, implicitly accounting for return requirements—the internal rate of return then equals the WACC. The module price at which this occurs is, therefore, the minimum price that sustains the manufacturer as a going concern while providing investors with their expected return. The standard methods used for preliminary estimation of capital and operating expenditures are described in more detail in Bartie et al. (2023) (Annexure D).

2.4 Integration of environmental and techno-economic assessments, and the effects of circularity

Environmental and techno-economic performance assessments are generally carried out and reported separately (Wunderlich et al. 2020; Mahmud et al. 2021). With the increased focus on overall sustainability and consequently, its quantification, economic viability cannot be the only criteria when designing or comparing technology options. Rather than considering the individual sustainability dimensions in isolation, integrated assessments of environmental and techno-economic performance could provide a more comprehensive picture of a life cycle system, in particular where environmental-economic trade-offs are likely to be present. From their review of the topic, Wunderlich et al. (2020) found that numerous methodological approaches with varying complexities and requirements are applied in practice to combine LCA and TEA, typically aimed at either identifying process hotspots, assessing the performance of alternative process designs, feedstocks, or product applications, or identifying fit-for-purpose technologies between alternatives. The authors presented a framework from which case-appropriate integration types can be selected based on the purpose of the integration, potential limitations due to the TRL, and the resources available for the task (Ibid.). In addition to potential TRL-related data availability challenges, specific methodological challenges remain for fully-aligned integration and are often related to the inconsistent selection of functional units and system boundaries, and discrepancies between the assumptions made for standalone LCAs and TEAs that are to be combined (Mahmud et al. 2021). There is a research gap with respect to tools that simultaneously perform LCA and TEA, while allowing for the influence of changes in process parameters to be investigated (Ibid.).

Integrated assessments of PV systems to identify sustainability trade-offs are practically non-existent (see Heath et al. (2022), for a recent review of CE for PV). Zhang et al. (2022) compare three perovskite technologies in terms of environmental impact and cost to identify material and manufacturing method combinations that could deliver the best environmental performance, and separately, the best economic performance. The authors identify trade-offs within the two sustainability dimensions with respect to material and manufacturing method selection, but highlight that additional methods are needed to quantify trade-offs between the dimensions.

Assessments of how circularity might simultaneously affect environmental and techno-economic life cycle performance, that is, the relationship between circularity and indicators from the two dimensions of sustainability considered in this dissertation, have not been found in the extant literature.

2.5 Surrogate models

Computational time and intensity can become problematic with large and complex process simulation models (Fröhling and Rentz 2010; Fröhling et al. 2012). One approach to deal with the complexity and associated computational intensity of large integrated system simulations is to use surrogate models (also called emulators, response surface models or meta-models). One of the purposes of a surrogate model is to represent the outcomes of a complex model that needs to be run over ranges of inputs – the surrogate model fits the available simulation data and can then be used to predict the results of the original simulation over predefined input ranges without having to run the computation-intensive simulation itself (Forrester et al. 2008). A surrogate model is a statistical black-box representation of a system's behaviour that is created by mapping the original input-output data to combinations of simple functions (Ferreira et al. 2019). It needs to deliver significantly improved computational efficiency and be usefully accurate (Forrester et al. 2008). Davis and colleagues compared the performance of several approaches for constructing surrogate models and found that, of the eight approaches tested, artificial neural networks (NN) delivered among the best estimations with the smallest errors (Davis et al. 2017, 2018). NNs conveniently enable generalized non-linear process modelling of complex systems without prior definition of regression equations (Reuter et al. 1992). NNs consist of layers of nodes that aim to emulate how neurons fire to transfer information in the brain. Each node is a computational unit with a selected number of inputs. It transforms a weighted sum of these inputs into an output using an activation function, for which the non-linear sigmoid function is typically used (Kubat 2017). The MATLAB (mathworks.com) programming environment is used in this work to create NNs using its built-in user interface, *nftool*. This tool uses the *tansig* transfer function, which is shown in Equation 3 below (Mathworks 2023), where Σw represents the weighted sum of the inputs into the node.

$$f(Sw) = \frac{2}{1 + e^{-2Sw}} - 1 \tag{3}$$

The number of assigned weights depends on the number of nodes defined and are the NN's degrees-of-freedom. Figure 2 shows a schematic representation of three-layer perceptron NN and how it is linked to the process simulation model. The simulation model and the NN receive the same inputs, and the NN is trained to replicate the outputs of the simulation as closely as possible.

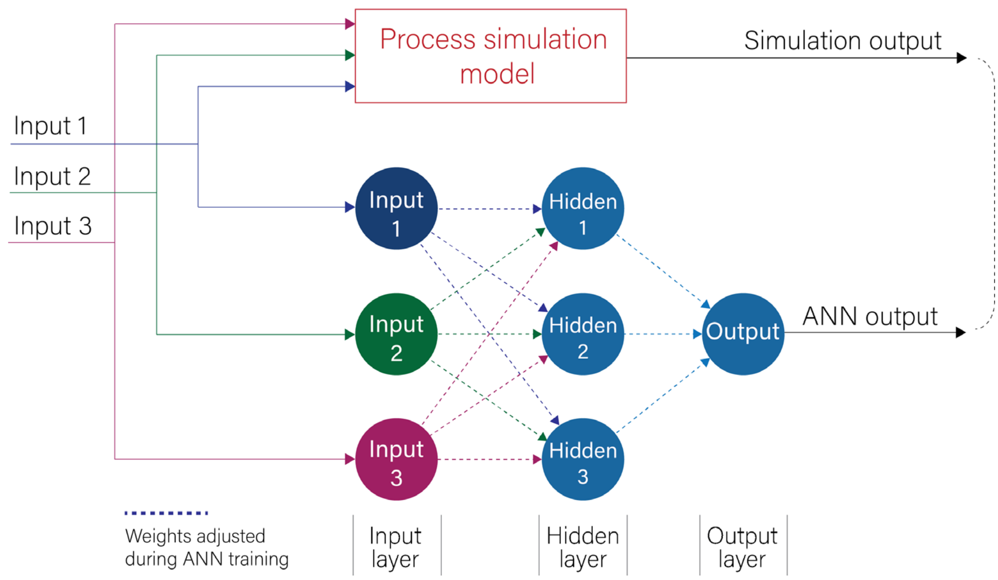

Figure 2. NN structure with its inputs and outputs linked to those of the process simulation model (note that each hidden and output layer has an additional 'bias' node, which is not shown) (adapted from Bartie et al. 2021a)

To generate the data needed to train, validate, and test the NN, the simulation model is run with a large number of random combinations of the independent variables of interest, in this case sampled from uniform distributions, and the corresponding system responses calculated. The resulting dataset is transferred from the HSC Chemistry simulation platform (described in Section 2.1.2) into the MATLAB (mathworks.com) programming environment where the networks are created, validated, and tested (Bartie et al. 2021a). For the work presented in this dissertation, several *three-layer perceptron* NNs (e.g. Reuter 1992), each with two to three inputs (i.e. independent variables) and one output (i.e. dependent variable), are employed to emulate input-output relationships of interest. More detailed descriptions of the procedure followed to create the surrogate functions and how errors are automatically handled in MATLAB are given in Bartie et al. (2021a).

2.6 Methods applied in the accompanying articles

In summary, the methods and indicators used are the following:

- Process simulation modelling, in which the laws of mass and energy are implicit to generate mass and energy inventory data (PS),
- Material resource efficiency represented by material recoveries (RE-M),
- Combined material and energy resource efficiency using exergy efficiency as proxy (RE-Ex),
- Combined material and energy resource consumption using exergy cost as proxy (RC-Ex),
- Energy return on investment using primary energy equivalents ($EROI_{PE\text{-}eq}$),
- LCA for potential environmental impacts, including carbon footprint,
- NPV, which includes the preliminary estimation of operating and capital expenditures for recycling to calculate MSP,
- LCOE as the main techno-economic performance indicator for the PV systems, and
- NN-based surrogate functions to represent simulation model outputs.

Table 2 provides a summary of methods applied in each of the included publications, also showing the elaboration of the overall approach during the course of this work.

Table 2: Systems analysed, and methods applied in each publication

Article no.	Title and detail covered		RQs addressed 1	RQs addressed 2	RQs addressed 3
1	<u>The simulation-based analysis of the resource efficiency of the circular economy – the enabling role of metallurgical infrastructure.</u>		●		
	Systems included in analysis	Cu, Te, Zn, Pb, and Cd production. CdTe PV module production and recycling (closed-loop recycling of Cd and Te).			
	Methods used	PS, RE-M, RE-Ex, RC-Ex, LCA			
	Analyses performed	Detailed flow analyses for Te and Cd, overall material recoveries, detailed exergy flow and efficiency analyses, exergy cost analysis, acidification potential, global warming potential, effect of electricity grid location on overall global warming potential.			
2	<u>The resources, exergetic and environmental footprint of the silicon photovoltaic circular economy: Assessment and opportunities.</u>		●	●	○
	Systems included in analysis	MG-Si production, SG-Si production, monocrystalline Si crystallization and wafering, Si (PERC) PV cell and module production, module recycling (closed-loop recycling of Si).			
	Methods used	PS, RE-M, RE-Ex, RC-Ex, LCA, NN			
	Analyses performed	Overall material recoveries, effects of circularity (EoL and kerf) on: nominal PV power generation potential, power consumption, carbon footprint; effect of SG-Si production technology substitution on carbon footprint, including response to changes in circularity; RE-Ex contribution analysis by production process, RC-Ex contribution analysis by module layer; effect of electricity grid location on carbon footprint and its response to circularity (EoL and kerf).			

Article no.	Title and detail covered		RQs addressed		
			1	2	3
3	Metallurgical infrastructure and technology criticality: the link between photovoltaics, sustainability, and the metals industry.		●	●	○
	Systems included in analysis	Consolidation of Articles 1 and 2 with additional analyses and comparison.			
	Methods used	PS, RE-M, RE-Ex, RC-Ex, LCA, NPV, NN			
	Analyses performed	MSP, comparison of RE-Ex and RC-Ex for CdTe and Si PV; effect of material intensity (via Si wafer thickness) on carbon footprint and its response to circularity (EoL and kerf); effect of carbon taxation on MSP and its response to EoL circularity.			
4	Cost versus environment? Combined life cycle, techno-economic, and circularity assessment of silicon- and perovskite-based photovoltaic systems.		●	●	●
	Systems included in analysis	In addition to Si system model from Article 2: perovskite precursor and cell production, perovskite module production and recycling, perovskite-Si tandem module production and recycling. Closed-loop recycling of Si in the Si and tandem PV systems.			
	Methods used	PS, RE-M, RE-Ex, RC-Ex, $EROI_{PE\text{-}eq}$, LCA, NPV, NN			
	Analyses performed	MSP, LCOE, variation of $EROI_{PE\text{-}eq}$, carbon footprint, and LCOE with EoL circularity; variation of carbon footprint and LCOE with PCE and PV system lifetime; effect of carbon taxation on MSP and its response to EoL circularity; effect of supply chain location on carbon footprint and MSP; qualitative analysis of environmental/techno-economic trade-off with a variation in circularity.			

○ Partial

The analyses listed in Table 2 for each of the articles are elaborated on in detail in Chapters 3, 4, 5, and 6.

3 Article 1[7]: The simulation-based analysis of the resource efficiency of the circular economy – the enabling role of metallurgical infrastructure

This Chapter provides a summary of the first package of work conducted and published in Bartie et al. (2020) towards answering the research questions, hereafter referred to as Article 1. It deals with the methodological research gap pertaining to the need to use more sophisticated tools to analyse the material, energy and exergy flows in complex technology life cycles, and addresses the first part of RQ 1: *"Which methods can be applied and combined to analyse the resource efficiency, environmental impacts, and techno-economic performance of complex technology life cycles, taking into account the laws of conservation and the SLT to produce physics-based mass and energy balances?"*. Techno-economic performance is addressed in Chapters 5 and 6.

To create digital representations of, and predict the technical performance of the interconnected production and recycling systems that constitute the life cycles of complex products, the tools used for their digitalization need to be able to track both the quantities and qualities of elements, compounds, and energy streams, i.e. beyond solely mass balancing. The CdTe PV system represents the intersection of several base- and carrier-metal systems. As was shown in Figure 1, Te and Cd are co-products of Cu and Zn production, respectively. These systems are brought together to produce the CdTe absorber used in the PV cells. Furthermore, the Pb production system is relevant due to often being integrated with Zn production and being a good carrier for many of the minor metals that need to be recovered. Both Cd and Te can be recovered in either of their primary production systems, namely the Pb/Zn and the Cu/Ni carrier metal systems.

Considering this metallurgical knowledge, a large process simulation model that simulates the integrated production of Cu, Zn and Pb as well as several co-products including Cd, Te, Co, Ag, Au and others, and the transformation of several hazardous residues into inert materials for discard or further processing. Furthermore, CdTe PV manufacturing and recycling processes are simulated so that Cd and Te can be returned to the life cycle for re-use, while accounting for the losses that occur along the way. Further integration of the production systems is achieved through the exchange of compatible by-products and residues such as slags and dusts between the processes. This large, integrated model allows for the system-wide effects of the exchanges to be evaluated to reduce dissipative losses

[7] Bartie, N. J., Abadías Llamas, A., Heibeck, M., Fröhling, M., Volkova, O., & Reuter, M. A. (2020). The simulation-based analysis of the resource efficiency of the circular economy – the enabling role of metallurgical infrastructure. *Mineral Processing and Extractive Metallurgy*, *129*(2), 229–249. https://doi.org/10.1080/25726641.2019.1685243.
The published article can be found in Annexure A.

and the degradation of material and energy quality. It consists of 223 interconnected unit operations, 869 process flows, and 30 elements with several of their compounds. A simplified block flow diagram of the entire system is shown in Figure 3 and further details can be found in Bartie et al. (2020).

Because of the elevated level of detail used throughout the integrated model, mass balances are generated at a detail level that allows for elements, compounds, and ionic species to be tracked through the entire system. At the same time it is taken into consideration that, until separated in the appropriate process, elements do not move through the system in pure form but are rather tied to one another in various types of mixtures.

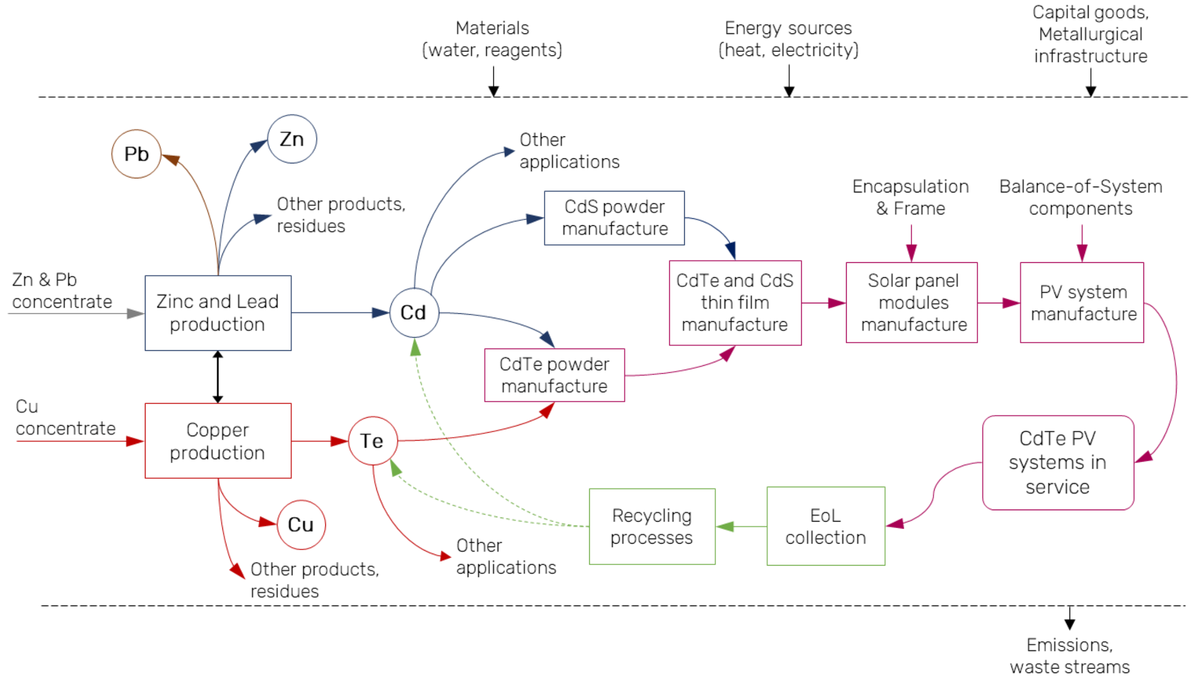

Figure 3. Simplified block flow diagram of the integrated production systems included in the simulation model. The process starts with the processing of Zn, Pb, and Cu concentrates to produce those base metals. Cd and Te are produced as co-products in these systems, i.e. without producing Cu, Pb, and Zn first, Cd and Te cannot be produced. Progressing to the right-hand side, Cd and Te are processed into the compounds needed to subsequently produce CdTe solar PV modules. After the use phase, Cd and Te are recovered via their respective recycling processes and are, in the closed-loop case, returned to the main production chain. This block flow diagram represents a simulation model consisting of 244 unit processes, more than 800 streams, and over 200 chemical species (adapted from Bartie et al. 2020 – Article 1, Annexure A)

The flows of Cd and Te are analysed as examples, demonstrating opportunities for closed-loop recycling, and showing the numerous locations Cd and Te are dissipatively lost from the system (shown in Figure 4). The quantification of these losses facilitates the rigorous calculation of material recoveries for these and several valuable metals.

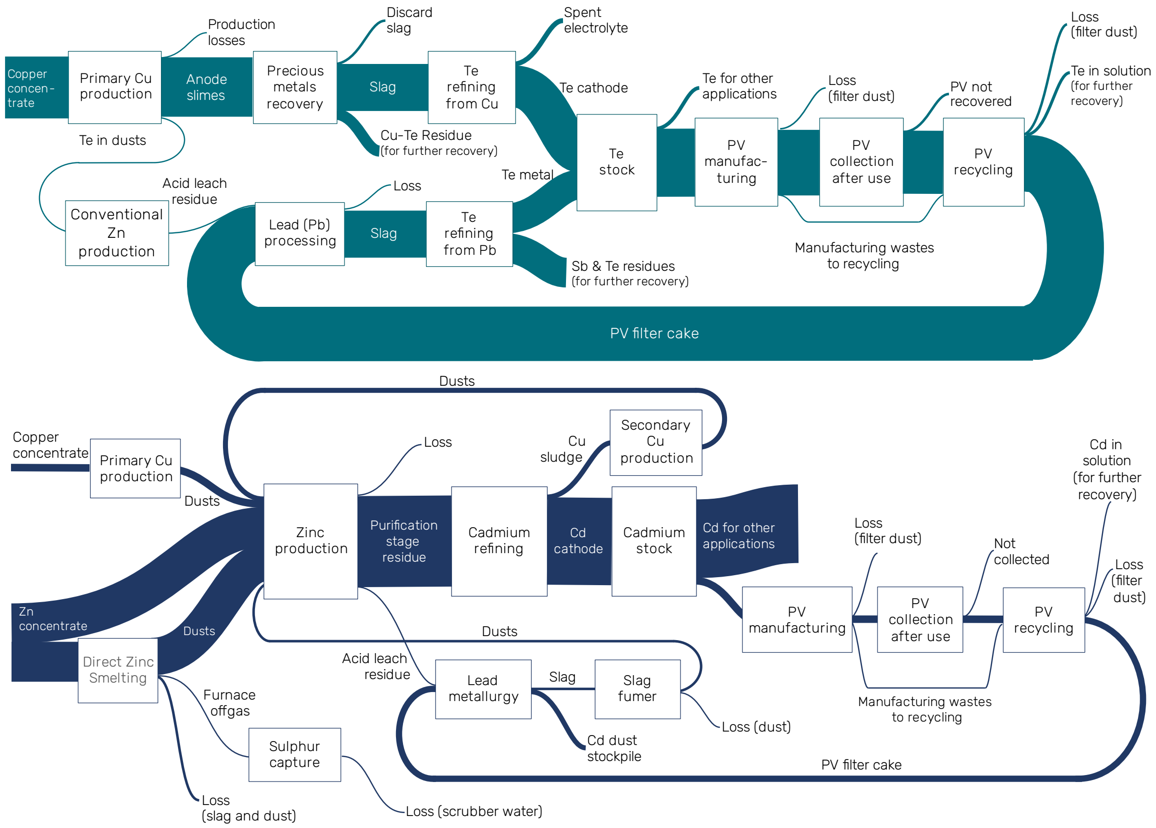

Figure 4. Sankey diagrams depicting the flows of Cd and Te through the entire life cycle, also showing where they are lost and where recovered Cd and Te may re-enter the life cycle via the Pb production system. It should be noted that, while these diagrams seem fairly simple, they represent the tracking of Cd and Te through the 244 unit processes the simulation model consists of. Furthermore, even though expressed as elements in the Sankey diagrams, Cd and Te are tracked in the actual elemental, compound, or ionic forms they are present as (adapted from Bartie et al., 2020 – Article 1, Annexure A).

The analyses shown in Figure 4 revealed that approximately 7% and 40% of Cd and Te, respectively, leave the life as waste or residue streams. In addition to calculating the material efficiencies of several valuable metals, the SLT is applied to determine gate-to-gate and the system's overall resource efficiencies. The selected figure of merit, exergy efficiency, is used because the exergy quantity simultaneously considers material and energy streams, all expressed in units of energy. The results are shown in Figure 5.

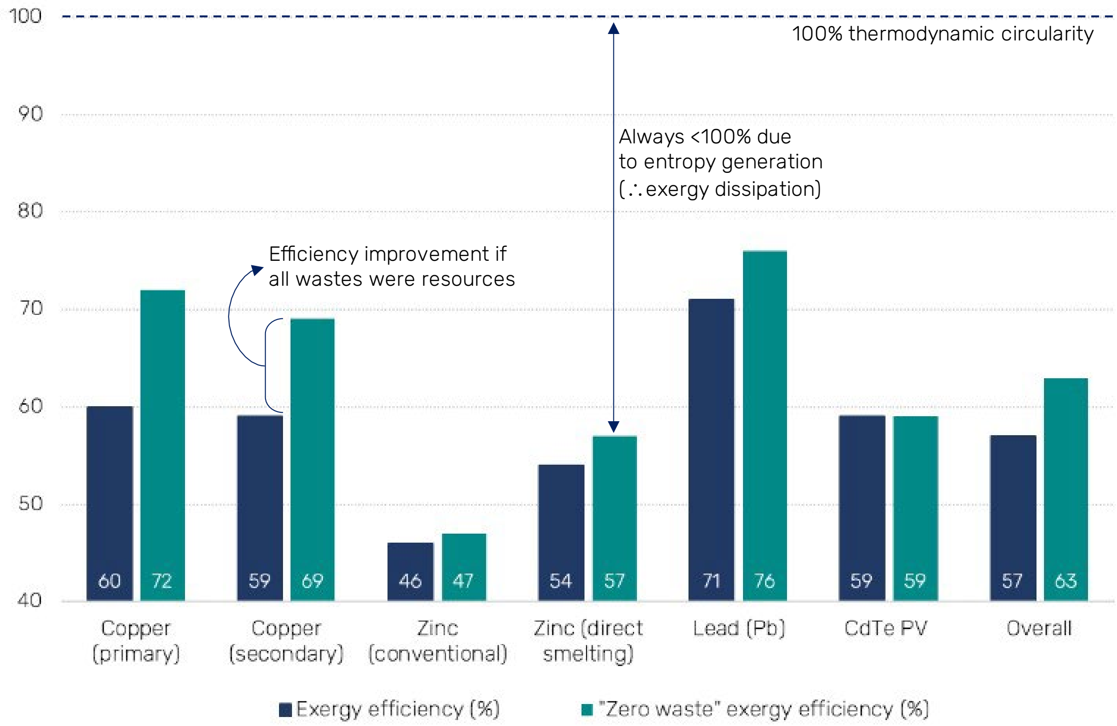

Figure 5. Exergy efficiency as a proxy for overall material and energy resource efficiency, shown for the main production processes in the CdTe PV life cycle. The blue bars on the left represent the efficiencies calculated from simulation model results. The green bars on the right represent a hypothetical case in which there are no waste streams, i.e. all wastes have become resources, as is often touted by some CE proponents as achievable. It can be clearly seen that, even in such a case, resource efficiencies are nowhere near 100%, i.e. real-life cycle loops always remain partially open (adapted from Bartie et al., 2020 – Article 1, Annexure A).

Figure 5 highlights inefficiencies and opportunities for innovation towards increased sustainability and CE. It also highlights the physical limits of CE—it is shown that, even if all wastes are hypothetically transformed into resources, resource efficiencies are always below 100%. The relatively low resource efficiencies of individual processes in the system show that considerable work must still be done to improve their resource-related and technical performance, but the unavoidable generation of entropy will result in maximum efficiencies always being less than 100%. The mass and energy balances also serve as the inventory data needed to assess environmental impacts. Here, the inventory data was directly mapped to an environmental database to estimate the potential environmental impacts of individual processes and the complete integrated system. Power consumption was found to be a major contributor to overall global warming and acidification potential. Results for the system's overall global warming potential and acidification potential are presented in the article (Annexure A).

As a first step in the body of work presented in this dissertation, Article 1 shows the benefits of using a single simulation platform to map all the physical[8] flows in the system to evaluate its true[9] resource consumption, resource efficiency, and environmental footprint based on a consistent set of inventory data. In doing so, the inconvenient truth about not being able to "close the loop" for this life cycle is quantified. The process simulation approach gives the ability to rigorously establish the baseline resource and sustainability performance of product systems that contain multiple metals and other materials, and to predict the potential system-wide effects of variations, e.g. because of efficiency improvements, technology changes with the aim to impactfully drive sustainable development and the transition to CE. Furthermore, Article 1 highlights the vital role the metallurgical process industry has to play in minimising the dissipation of materials and exergy to maximise resource efficiency. One of the main focus areas of Article 1 is the analysis of thermodynamic irreversibility and limits. However, as stated in the paper, it is acknowledged that economic, social, and environmental factors also contribute to limits and irreversibilities in product life cycles (Bartie et al. 2020). From the application-oriented point of view, the modelling of closed-loop material flows through the entire value chain for both Cd and Te facilitates the assessment of CE scenarios for CdTe PV modules. These topics are discussed further in the Chapters that follow.

[8] The term *physical flows* refers to the flows of materials, energy, and exergy in this dissertation.
[9] In this dissertation, *true* resource efficiency refers to efficiency that also incorporates losses due to the generation of entropy.

4 Article 2[10]: The resources, exergetic and environmental footprint of the silicon photovoltaic circular economy: Assessment and opportunities

Key themes from the second package of work conducted and published in Bartie et al. (2021a), hereafter referred to as Article 2, are summarized in this Chapter. The published article can be found in Annexure B. Whereas Article 1 demonstrated the complexity of a PV life cycle that contains by-product metals and CRMs, the subject of Article 2 is the life cycle of Si-based PV technology, which is, and is set to remain the dominant technology for many years to come (VDMA 2021). While Si is not a by-product metal[11], it is a CRM (European Commission 2020b). The article builds on Article 1 and further addresses methodological gaps pertaining to the need to establish the relationships between circularity and sustainability, i.e. to analyse the effects of circular strategies on overall life cycle sustainability. It addresses RQ 1 and the first part of RQ 2: *"What are the combined quantitative effects of circularity on the resource, environmental, and techno-economic performance of established and emerging PV systems"*. The techno-economic aspects of both RQ 1 and RQ 2 are covered in Chapters 5 and 6.

Starting with the same approach as that for the CdTe system in Article 1, detailed simulation models are created for the full Si PV life cycle, including the production of metallurgical grade Si (MG-Si) from quartzite (which consists predominantly of SiO_2), the production of solar grade Si (SG-Si), monocrystalline Si ingots, wafers, PV cells, and modules, and EoL recycling. Because Si is a CRM, specific attention is given to two recycling loops in this system. The first is the recycling of the so-called *kerf*, which is a residue from the process of cutting thin wafers from monocrystalline Si ingots using diamond wire cutting. The thickness of Si wafers for PV have decreased to a current average (in 2021) of 170 μm, with a kerf thickness of 60 μm. If not recovered, this equates to a 26% loss of high-purity monocrystalline Si from the cutting step alone (VDMA 2021). Residues from other process steps throughout the life cycle further add to the amount of Si metal needed to produce a wafer. Kerf recycling is problematic due to, among others, contamination with cutting media and the risk of explosions during handling because of hydrogen formation (Halvorsen et al. 2017). However, its importance as a potential secondary resource has been recognised. Recyclate purities of 2N (99%) to 4N (99.99%)

[10] Bartie, N. J., Cobos-Becerra, Y. L., Fröhling, M., Schlatmann, R., & Reuter, M. A. (2021). The resources, exergetic and environmental footprint of the silicon photovoltaic circular economy: Assessment and opportunities. *Resources, Conservation and Recycling*, *169*, 105516. https://doi.org/10.1016/j.resconrec.2021.105516

[11] Silicon is generally referred to as metal. However, it is a metalloid or semi-metal which possesses properties of both metals and non-metals.

have been demonstrated at pilot scale (Ibid.) and commercial processes for its recycling are emerging (e.g. rosi-solar.com). The second loop is the closed-loop recycling of monocrystalline Si recovered from used wafers during EoL recycling. Although not yet commercialised, the recycling process selected for the simulation model (described in detail in Article 2) has been shown to recover up to 90% of the EoL Si at SG-Si purity. Because the two recyclates are of different purity, they are returned to distinct locations in the life cycle to close the respective material loops, keeping in mind that only partial closure is possible due to dissipative losses and entropy creation throughout the life cycle. The production processes that constitute the life cycle and the two loops described here are shown in Figure 6.

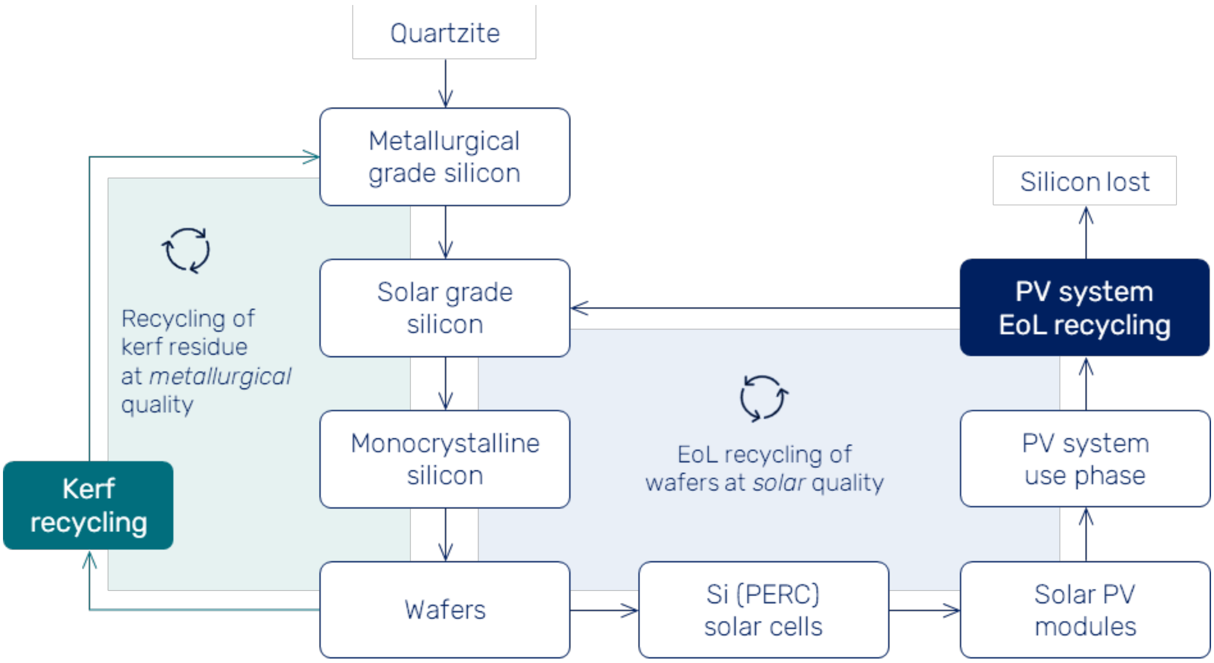

Figure 6. The flows Si-containing streams through the Si PV life cycle with the EoL and kerf recycling loops highlighted. Each of the boxes represents the production process for the named product. The acronym *PERC* refers to the *passivated emitter, rear contact* cell architecture modelled in Article 2. Note that, while *silicon lost* is only shown to exit the system during PV system EoL recycling, Si losses also occur during all other processes, without exception. These are not shown for the sake of readability but are highlighted in Bartie et al. (2021a) – Article 2, Annexure B.

As shown in Figure 6, the higher purity of the EoL stream allows for it to re-enter the life cycle after the energy-intensive SG-Si production process, so also excluding the environmental impacts associated with that energy consumption. As was the case for the CdTe system in Article 1, the elevated level of detail behind every process block shown in Figure 6 in the simulation models allows for all flows, including losses, to be quantified at the compound/element/ion level and for the subsequent physics-based calculation of material and energy/exergy efficiencies. These are first determined for three fixed operating cases. In the reference case, the recycling loops are not considered. In the second case, it is assumed that a 95% EoL recycling rate[12] of SG-Si is achieved. In the third, it is assumed that half of the recovered kerf residue can additionally be recycled at MG-Si grade to be reused in the PV life cycle.

Article 2 advances the approaches in previous studies that integrate process simulation with ecological and techno-economic assessments in the process industries, for example, to develop an operations planning decision support system for the blending of ferrous residues reused in the iron/steel industry (Fröhling and Rentz 2010), to improve resource efficiency in geographically distributed recycling networks (Fröhling et al. 2012), and the approach for simulation-based design-for-recycling in metal production and recycling systems demonstrated by Reuter et al. (2015). A significant elaboration of the methodology presented in Article 1 is the use of NNs as surrogate representations of the simulation model to quantify the system-wide effects one or several parameters in the system. As stated in Section 2.5, the use of NNs negates the need to define regression equations beforehand, which is especially advantageous for large systems. The combined use of the simulation model and its NN-based surrogate functions allows for the simultaneous effects of the two recycling loops on, among others, system-wide power consumption, nominal PV power generation capacity, and carbon footprint to be quantified. In other words, the effects of manufacturing and EoL circularity are incorporated into the approach to enable analysis of the relationships between circularity and the resource and environmental performance of the life cycle. As an example, Figure 7 shows the life cycle carbon footprint for the three cases superimposed on the systems carbon footprint response to change in the two recycling rates. The difference between the footprints of the reference case (146 $kgCO_2e/m^2$) and Case 1 (125 $kgCO_2e/m^2$) demonstrates the significant benefit of recycling EoL wafers as SG-Si to bypass the energy-intensive MG-Si and SG-Si production processes. The difference between Cases 1 and 2 shows that much smaller effect of kerf recycling. It is emphasised, however, that this analysis shows carbon footprint only, and that larger effects may be present in other impact categories. These findings are described in more detail in Article 2.

[12] Per definition, EoL recycling refers to the collection and physical recycling of the actual materials in products that have reached end of life after use. In other words, it refers to materials placed on the market likely several years ago (typically 25-30 years for PV systems). This recycling rate is also used as an indicator of circularity. See, for example, Hagelüken and Goldmann (2022).

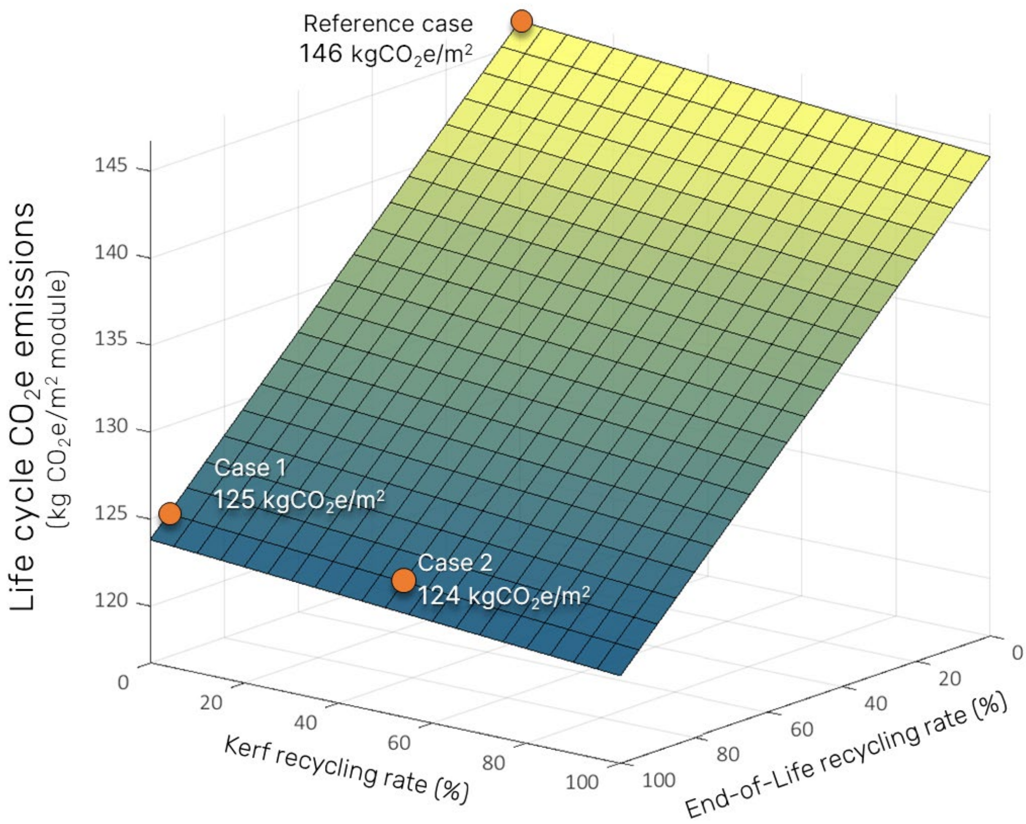

Figure 7. Carbon footprint of the complete Si PV life cycle for the three predefined operating points as well as the system's carbon footprint response over the full ranges of closed-loop kerf and EoL Si recycling generated using NN-based surrogate functions to represent the process simulation model. Adapted from Bartie et al. (2021a) – Article 2, Annexure B.

This is also done for an alternative SG-Si production technology, the so-called silane fluidised bed reactor (FBR) process, which consumes significantly less power and hence, exhibits reduced electricity-consumption-related carbon emissions. This case shows how the choice of technology can change the driver of carbon emissions with changes in the degree of circularity and quantifies the effects of such a change on absolute emissions. The FBR process clearly demonstrates significant carbon footprint benefits even if full circularity were to be achieved in the life cycle using the Siemens process. In the Siemens-based life cycle, a hypothetical full Si circularity would be needed to achieve a carbon footprint similar to that of a FBR-based life cycle with no Si circularity. It is clear that both kerf and EoL recycling reduce carbon footprint, but that EoL recycling is more effective at doing so. These findings are illustrated in Figure 8.

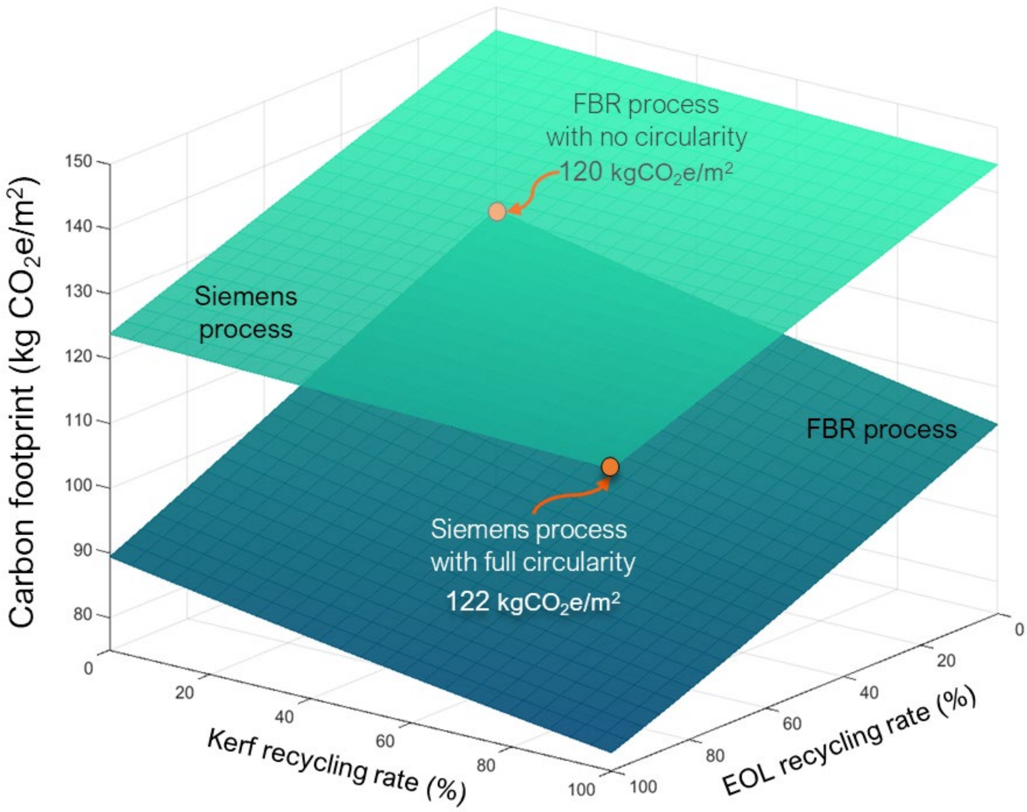

Figure 8. Si PV life cycle carbon footprint comparison for the production of SG-Si via the Siemens and FBR processes, showing the system's response over the full ranges of closed-loop kerf and EoL Si recycling generated using NN-based surrogate functions. With *full* Si circularity in the Siemens-based system, a carbon footprint of 122 kgCO_2e/m^2 modules produced is achieved, while in the FBR-based system with *no* Si circularity, the footprint is 120 kgCO_2e/m^2. Adapted from Bartie et al. (2021a) – Article 2, Annexure B.

While technoeconomic performance is not evaluated in Article 2, it is covered in expansions of this study discussed in Bartie et al. (2021b) and Article 3 (Bartie et al. 2022), as well as in Article 4 (Bartie et al. 2023). Articles 3 and 4 are covered in Chapters 5 and 6, respectively.

Article 2 presents high-detail analyses of the relationships between the selected resource and sustainability indicators, and two strategies that aim to increase circularity—the recycling of kerf residue and EoL Si wafers. By quantifying the links between CE strategies and sustainability, this research gap, also highlighted by Korhonen et al. (2018) and Roos Lindgreen et al. (2020), is addressed for the Si PV life cycle system. The results show both the directions and the relative magnitudes of change in the system's carbon footprint as a function of the degree to which the two strategies are implemented. The degree of implementation depends on a range of factors like the availability of recycling technologies and market conditions around secondary raw materials, among several others. Therefore, it is useful to be able to look at the indicator of interest (carbon footprint shown here) over the full ranges of recycling rate (i.e. circularity) as opposed to considering only specific operating points without knowing in which direction it would change, and by how much, if a change in circularity were to occur due to other factors, whether quantitative or qualitative.

The processes and circular strategies selected in Article 2 are in line with current developments in the PV industry, including the ongoing research and innovation for processes that enable high-quality kerf and Si wafer recycling. Furthermore, they are in line with expert recommendations for the Si PV industry, which include focusing on developing recycling processes that deliver high-grade products, reducing material intensity, and reducing manufacturing losses (Heath et al. 2020). While the FBR process for SG-Si production had a market share of only 5% in 2020, current expectations are for it to reach approximately 12% by 2030 (VDMA 2021). The quantification of the differences in carbon footprint between the FBR and the market-dominating Siemens process, combined with the potential effects of transitioning to CE (as shown in Figure 9), provides the Si PV industry with valuable information to guide the development and innovation of production and recycling technologies, also taking carbon footprint into consideration. It is emphasised that the findings presented here might differ for impact categories other than the carbon footprint and in the other dimensions (economy and society) of sustainability. This should be investigated further in future studies. To the best of the author's knowledge, however, this is the first study to simulate an entire PV life cycle at this elevated level of detail, also including the simultaneous recycling of internal and EoL residues at independent qualities and rates to evaluate the system's resource and environmental response to changes in circularity. As highlighted earlier, methodological advancement is demonstrated through the development of NN-based surrogate functions to closely mimic the simulation model with only a fraction of the computational intensity. It allows for the effects of circularity, whether internal to the life cycle or via EoL recycling, on the resource and environmental performance of the life cycle to be incorporated into the approach. Readers are referred to Article 2 (Annexure B) for significantly more detail on these and other findings.

5 Article 3[13]: Metallurgical infrastructure and technology criticality: the link between photovoltaics, sustainability, and the metals industry

Article 3—Bartie et al. (2022) in Annexure C— expands on Article 2 with the addition of a techno-economic assessment for the Si PV system, which also allows for the influence of carbon taxation to be analysed. Additionally, it serves as a consolidation and comparison of selected results obtained so far. A comparison of overall resource efficiencies for the production of CdTe and Si PV modules, using exergy to simultaneously account for material and energy flows, is depicted in Figure 9.

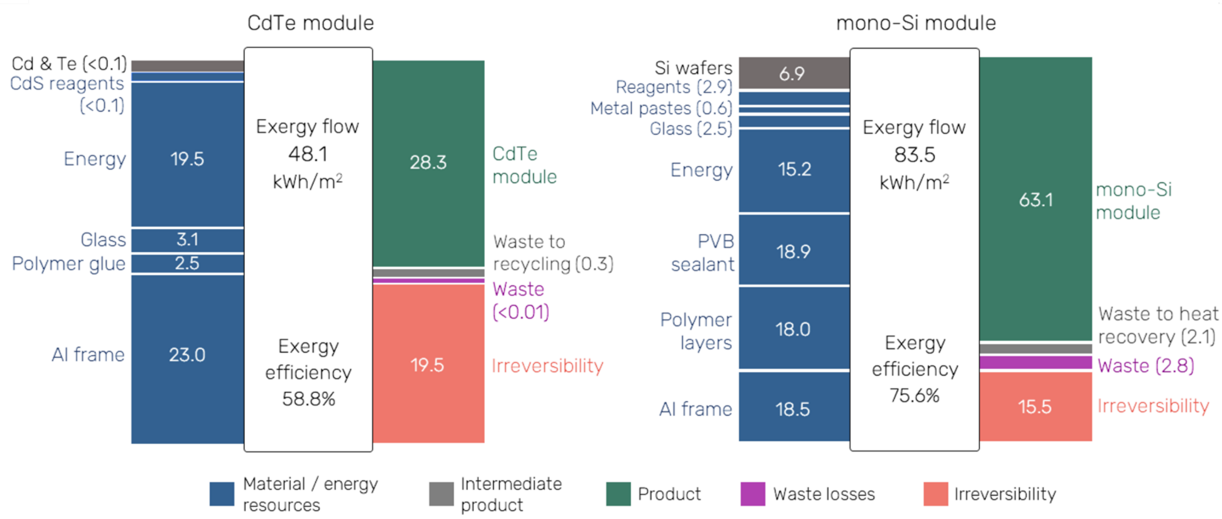

Figure 9. Comparison of exergy flows and efficiencies of CdTe PV (left) and Si PV (right) module production. Total exergy flow per square meter of PV module produced are 48.1 kWh/m^2 for CdTe PV and 83.5 kWh/m^2 for Si PV, with exergy efficiencies of 58.8% and 75.6%, respectively. The term *mono-Si* refers to monocrystalline Si. Adapted from Bartie et al. (2022) – Article 3, Annexure C.

Figure 9 shows irreversibilities—the amount of exergy dissipated during the module production process—of 19.5 and 15.5 kWh/m^2 for the CdTe and Si PV cases, respectively. In other words, more entropy is generated per unit area of CdTe PV modules produced compared to Si PV modules. The largest exergy flows in the CdTe system originate from the consumption of energy and aluminium (Al) frames, while in the Si system, the consumption of energy, polymer sealants and foils, and Al contribute similarly to the total exergy flow. Reducing the amounts of Al used for module frames or opting for a glass-

[13] Bartie, N., Cobos-Becerra, L., Fröhling, M., Schlatmann, R., & Reuter, M. (2022). Metallurgical infrastructure and technology criticality: the link between photovoltaics, sustainability, and the metals industry. *Mineral Economics*, *35*(3-4), 503–519. https://doi.org/10.1007/s13563-022-00313-7

glass module configuration in which the polymer-based rear layers are replaced by a second glass layer, and reducing overall energy consumption would all result in decreased irreversibility. This SLT-based analysis, therefore, provides designers and manufacturers with valuable guidance about opportunities to increase overall resource efficiency, also taking the resource loss due to entropy generation into account. In a further resource efficiency analysis, a change in Si material intensity and its effects on the system's carbon footprint was quantified. A decrease in wafer thickness from 175 to 150 μm, as forecasted for the period 2020 to 2031 (VDMA, 2021), was found to reduce life cycle carbon emissions by up to 4.7% without EoL recycling, while EoL circularity alone could reduce emissions by up to 15.6%. The combined effects of changes in wafer thickness and circularity are shown in Figure 10a below. These findings highlight the need to innovate processes that recycle Si at high purity and show the potential environmental benefits quantitatively.

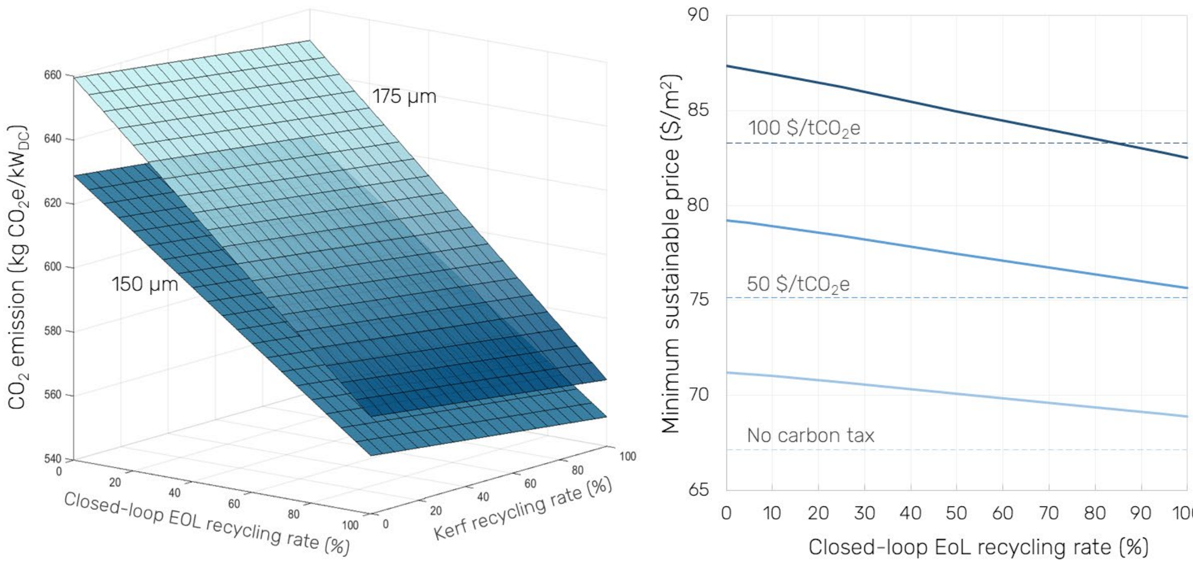

Figure 10. (a) The combined effects of changes in Si material intensity and circularity (kerf and EoL) on the system-wide carbon footprint, and (b) the effects of carbon taxation and EoL circularity on MSP in the Si PV system. Dashed horizontal lines represent the MSP without the costs of establishing and running the recycling process. Both figures adapted from Bartie et al. (2022) – Article 3, Annexure C.

In a significant expansion of the approach following Articles 1 and 2, a bottom-up cost model was developed for the Si PV system to evaluate the effects of Si circularity on the MSP. Furthermore, because the linked models allow for CO_2e emissions and costs to be linked, the effects of hypothetical carbon taxes were analysed. Results from this analysis (Figure 10b) quantify to what extent circular flows of Si recovered at EoL decrease MSP at different taxation rates. In the case where there is no tax, it is evident that circularity on its own cannot fully compensate for its costs. Consequently, other cost reduction measures would also have to be employed if the MSP is to be reduced to the original level. At higher tax rates, the beneficial effects of circularity on carbon footprint are more pronounced so that the MSP without recycling can again be achieved, as evidenced by the 100 $/t$CO_2$e line crossing the dashed line at an EoL recycling rate of approximately 85%. Tax rates that high are unlikely, though, again confirming the need to implement additional cost reduction approaches. These and other findings are discussed in more detail in Article 3. A similar analysis is also conducted for the perovskite-silicon tandem PV system in Article 4 in the next Chapter.

6 Article 4[14]: Cost versus environment? Combined life-cycle, techno-economic, and circularity assessment of silicon and perovskite based photovoltaic systems

Article 4—Bartie et al. (2023) in Annexure D—expands on the responses to RQs 1 and 2 up to this point, and also addresses RQ 3: "*Does EoL circularity contribute to the sustainability of established and emerging PV technologies?*". It expands on the work presented in Articles 2 and 3 in several ways. In addition to the Si PV system, two highly relevant emerging PV systems are modelled and analysed—the first is a thin-film perovskite-based system and the second, a tandem system in which the Si and perovskite cells are combined into one module (described in Section 1.1.6). Furthermore, techno-economic assessment is added to the resource and environmental performance assessment methodology presented in Article 2, so adding another dimension of sustainability to the approach for all three systems. The bottom-up techno-economic assessments are used to determine the MSP and subsequently, the LCOE for each of the three PV types for system lifetimes of between 5 and 40 years, and PCEs between 16% and 30%. The advantage of the methodology is that the carbon footprint can be determined over the same ranges to enable fully aligned assessments of economic and environmental performance at any combination of lifetime and PCE within these ranges. To illustrate, Figure 11 shows the simultaneous carbon footprint (left) and LCOE (right) results for the Si PV system to illustrate the above. Complete results and breakeven analyses for all three systems are presented in the article itself in Annexure D.

[14] Bartie, N., Cobos-Becerra, L., Mathies, F., Dagar, J., Unger, E., Fröhling, M., Reuter, M. A., & Schlatmann, R. (2023). Cost versus environment? Combined life cycle, techno-economic, and circularity assessment of silicon- and perovskite-based photovoltaic systems. Journal of Industrial Ecology, 1–15. https://doi.org/10.1111/jiec.13389

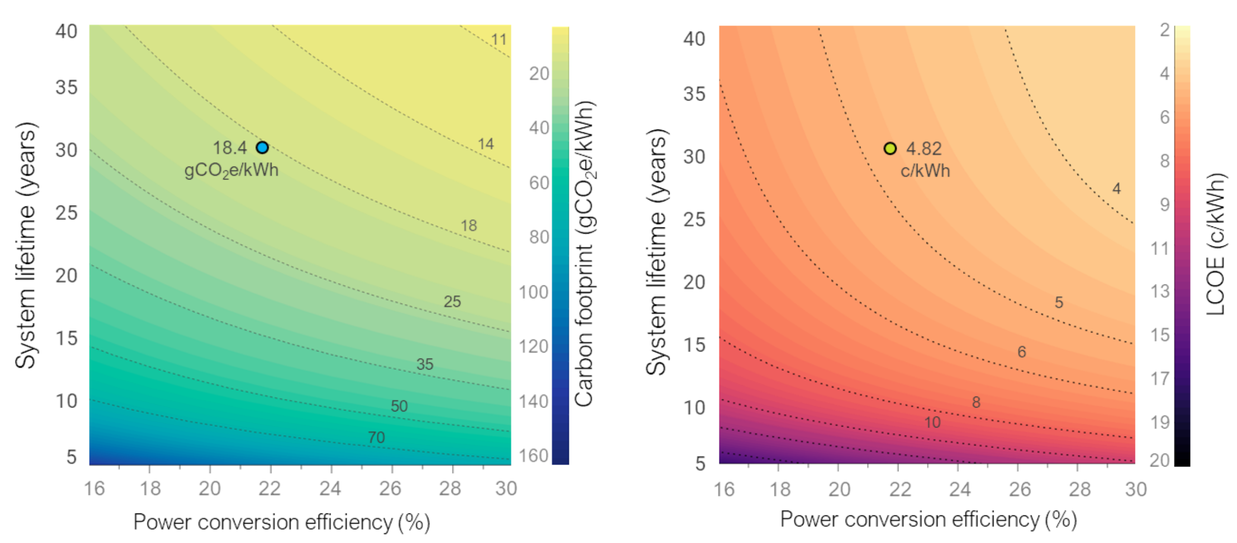

Figure 11. The variation carbon footprint (left) and LCOE (right) with system lifetime and PCE for the Si PV system. The datapoint shown on each of the maps represents the case for a system lifetime of 30 years and a PCE of 21.7%, which were selected to represent the current industrial reality, and for comparison against other studies. Results for the perovskite and tandem PV systems are presented in Bartie et al. (2023) – Article 4, Annexure D.

Additionally, material recoveries, the $EROI_{PE\text{-}eq}$, carbon footprint, and techno-economic indicators are determined at constant lifetime (30 years) and representative PCEs (18%, 21.7%, and 27.3% for the perovskite, Si, and tandem systems, respectively), while circularity is varied independently. This reveals the relationships between circularity, material and energy efficiency, and the indicators selected for the environmental and economic dimensions of sustainability. The results clearly show that, based on the assumptions and configurations of the simulation models, a trade-off exists between the environmental and economic dimensions in the tandem system. While an increase in circularity has a favourable effect on the system's carbon footprint, it also leads to an increase in LCOE. That is, a compromise would have to be found between minimising carbon footprint and minimising LCOE. Therefore, a key finding is that full circularity does *not* provide the most sustainable outcome in the tandem PV life cycle analysed in this work. In the perovskite and Si systems, on the other hand, this is not the case because their carbon footprints and LCOEs tend in the same direction as circularity changes. This finding is illustrated in Figure 12. For the sake of clarity and comparability of the three systems, the effects of kerf recycling are not presented in Article 4. This decision is considered valid because kerf is not present in the perovskite system and its impact on the results presented in Article 4 is small.

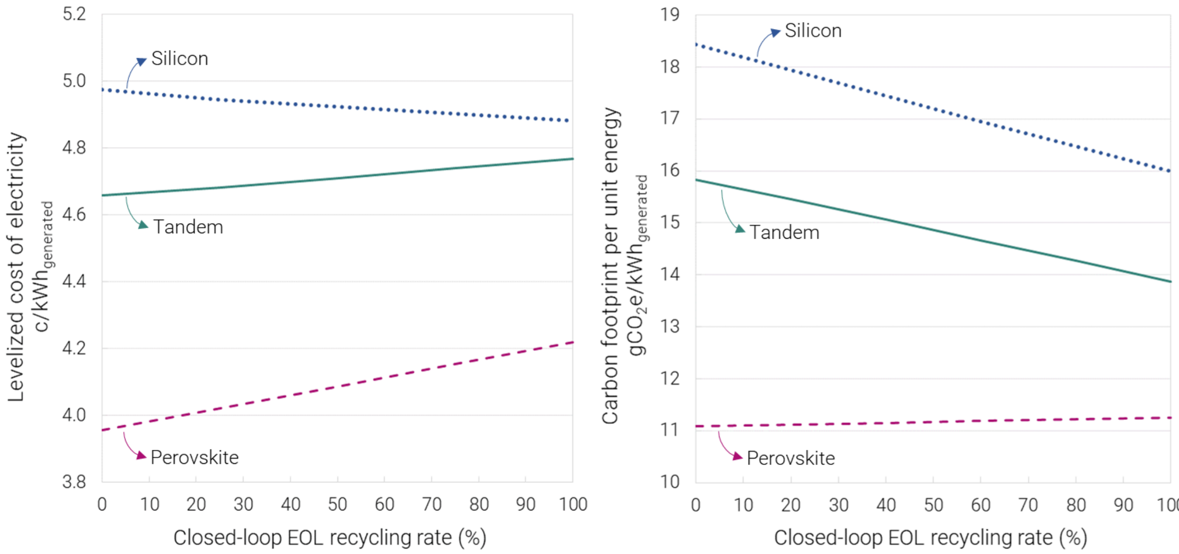

Figure 12. The variation of LCOE (left) and carbon footprint (right) with closed-loop EoL recycling rate in the perovskite, Si, and tandem PV systems, showing that LCOE and carbon footprint tend in opposite directions with a change in closed loop recycling rate (Bartie et al. 2023 – Article 4, Annexure D).

In essence, the answer to RQ 3—"*Does EoL circularity contribute to the sustainability of established and emerging PV technologies?*"—lies in this finding. From Figure 12 it can be seen that for the silicon system, the answer would be "yes". The assumption that circularity should be maximised to maximise overall sustainability does *not* hold in the perovskite and tandem systems. For the perovskite system, the answer would be "no". While maximum circularity would benefit environmental sustainability in the tandem system, it affects economic sustainability negatively. There is, therefore, a trade-off between the environmental and economic dimensions of sustainability with respect to EoL recycling as a circular strategy.

Every effort has been made to select parameters representative of the state of the art throughout the process and techno-economic models. However, it is highlighted here that perovskite-based PV (which applies to the perovskite and tandem systems in this dissertation) is under development and not a mature technology. Furthermore, there are numerous PV cell configuration options other than the single junction and four-terminal tandem variants selected for this work. It should be kept in mind, therefore, that the findings presented apply only to the selected systems. Significant changes in, for example, the materials used and their costs, recycling process efficiencies, impacts and costs, financial parameters like discount rates for techno-economic calculations, achieved PV PCEs and lifetimes, and the location of the electricity grid used for manufacturing, among others, could change the findings. Analyses of the sensitivity of LCOE to recycling cost and discount rate for the three PV systems indicate that the largest sensitivities are present in the Si PV system. Doubling the total recycling cost results in an 11% increase in LCOE, while halving it reduces LCOE by 5%. The sensitivity to discount rate is smaller, with a

5% increase and a 2% decrease in LCOE at double and half the discount rate, respectively. These sensitivities are smaller in both the perovskite and tandem systems.

Carbon taxation as a policy measure to curb global warming is also addressed in Article 4. Being able to simultaneously assess carbon footprint and LCOE enables the inclusion of costs that vary specifically with CO_2-equivalent emissions. The effects of a carbon tax on LCOE can therefore be quantified. Furthermore, because the responses of carbon footprint and LCOE to changes in circularity can be quantified, it is possible to evaluate how changes in circularity may exacerbate or diminish the effects of such a tax. It is shown that, from a cost perspective, circularity could counteract the effects of the tax on LCOE, but that the pre-tax LCOE would only be achieved when the tax is unrealistically high. The implication is that the best way to counteract the tax would be to reduce carbon emissions in the first place. The tax would, therefore, have the desired effect. The caveat is that high-emission processes are not moved to locations where carbon taxes are lower, in which case further regulation in the form of border tax adjustments would be required. The Council of the European Union has, in fact, recently reached agreement on the Carbon Border Adjustment Mechanism (CBAM) regulation that aims to prevent so-called 'carbon leakage' (European Commission 2021).

While Article 2 covered resources and carbon footprint for the Si system alone, Article 4 compares three PV systems in terms of energy consumed and produced, carbon footprint, and LCOE, for both open- and closed-loop scenarios. The three systems are compared from a CE and systems perspective in terms of physical flows, environmental impact, economic performance, and policy decisions, also considering interactions between them. Furthermore, this publication demonstrates the use of systems-based analytical tools with predictive capabilities to evaluate potential environmental-economic trade-offs that may exist, a research gap also highlighted by Heath et al. (2020).

7 Discussion

This Chapter summarises findings in the context of the posed RQs and highlights novel aspects and implications.

7.1 Addressing the Research Questions

7.1.1 Research Question 1

The first research question—*"Which methods can be applied to analyse the life cycle resource efficiency, environmental impacts, and techno-economic performance of a complex technology, taking into account the laws of conservation and the SLT?"*—is addressed in all the articles published during the course of this work and reflects the evolution of the research presented in this dissertation. Starting with Article 1 and the simulation of an integrated network of complex production and recycling processes and assessing the associated environmental impacts, the approach evolved in Article 2 to also include the effects of EoL circularity on resource efficiency and environmental performance, so beginning to link sustainability and CE indicators to address the second research question. This was achieved by uniquely creating NN-based surrogate functions to emulate the simulation model to increase computational efficiency. Large process simulations can be complex with many potential sources of non-linearity, like non-linear reaction kinetics and thermodynamic relationships that define the transformations that occur within process units, as well as non-linearities introduced by recycling loops and interactions of process units with others and with the environment. Unlike conventional linear approaches, NNs are capable of learning complex and non-linear patterns in data and can, therefore, capture non-linear input-output relationships (Reuter 1993). Therefore, the integration of process simulation and NN-based surrogate functions is a prudent approach. Additionally, if the process simulation approach to sustainability assessment is to be disseminated more widely, surrogate functions that simplify the generation of inventories would be highly beneficial.

In Article 4, the approach is taken another step further by incorporating bottom-up cost models to additionally assess techno-economic performance, also at varying degrees of circularity. In summary, the end-product is an approach that first creates a highly detailed, disaggregated process simulation model of the entire technology life cycle to quantify material and energy flows, as well as the enthalpy, entropy, and free energy of all elements, compounds, and mixtures to quantify exergy dissipation (due to entropy creation). The detailed mass and energy balances generated in this way then serve as the core source of physical flow data for assessments of resource, environmental and techno-economic life cycle performance. Material and energy efficiencies are expressed in terms of recovery rates and the $EROI_{PE\text{-}eq}$, respectively. The standardised LCA methodology is used to assess potential environmental impacts, and discounted cash flow methods are used to calculate

MSP and LCOE. The process simulation approach ensures that the laws of conservation and the SLT are adhered to at all times, so preventing overoptimistic assessments of resource efficiency and impacts.

7.1.2 Research Question 2

The second research question—*"What are the combined effects of circularity on the resource, environmental, and techno-economic performance of established and emerging PV systems"*—is addressed in Articles 2, 3, and 4. As already discussed, the use of NN-based surrogate functions allowed for the responses of all the abovementioned indicators to changes in the degree of EoL circularity to be assessed, so revealing potential interactions between circularity and different dimensions of sustainability. The effects of both kerf residue and EoL wafer Si circularity in the Si PV system are discussed in Article 2. The response of nominal PV power generation capacity quantifies the potential additional power that can be generated without increasing virgin raw material consumption for the production of Si when kerf residue is recycled and kept in the life cycle as MG-Si and Si recovered from EoL wafers as SG-Si. The responses of life cycle electricity consumption and CO_2e emissions reveal the considerable benefits of EoL SG-Si circularity and the relatively smaller benefit of kerf recycling. The carbon footprints of two technology options for the production of SG-Si, the Siemens and FBR processes, are also compared in Article 2 with respect to the kerf and EoL circularity options, highlighting the significant environmental benefits of the latter. It should be noted that no data are available for the FBR process in the popular inventory databases. However, the process simulation model could be adapted fairly easily to include it and to generate the required inventory data at the same high level of detail. The simultaneous effects of geographical location and circularity on carbon footprint are also analysed and quantified in Article 2 and the effects of Si wafer thickness as a measure of material intensity in Bartie et al. (2022).

In Article 4, further expansions to the approach are implemented to analyse the responses of energy efficiency (represented by $EROI_{PE\text{-}eq}$) and techno-economic performance (represented by LCOE) in addition to the carbon footprint over the full range of EoL circularity. These analyses are done for the established Si PV and the emerging perovskite and perovskite/Si tandem PV systems to enable their comparison and generate knowledge to guide the future development of these technologies that are currently receiving considerable research and development attention. Because of the relatively smaller contribution of kerf recycling, and for the sake of clarity, the kerf circularity option is not considered in Article 4. The approach is further applied to assess the potential effects of carbon taxation on LCOE in the tandem system and the potential for circularity to counteract the cost implications of the tax.

Among the key findings from Article 4 that answer the research question are that the perovskite system outperforms the Si and tandem systems in all dimensions—$EROI_{PE\text{-}eq}$, carbon footprint, and LCOE. However, the precondition for this finding to hold is that research and innovation activities find solutions for current challenges surrounding the long-term stability of perovskite absorbers. Circularity was found to be beneficial to the $EROI_{PE\text{-}eq}$ in all systems. It had little effect on carbon footprint in the perovskite system but contributed significantly to reducing CO_2e emissions in the Si and tandem systems. In the perovskite and tandem systems, increased circularity resulted in higher LCOE, while lowering LCOE in the Si system. With regard to carbon taxation, increased circularity was found to have a dampening effect on the tax-induced cost increase that becomes stronger as the tax rate increases. Above a certain threshold, in this case estimated at a high $210 per tonne of CO_2e emissions, increased circularity starts to reduce LCOE.

In summary, the findings from Articles 2, 3, and 4 answer the second research question successfully by linking resource, environmental, and techno-economic indicators to circularity. At the core of this outcome lies the consistent use of inventory data generated in the process simulation models across all other assessments.

7.1.3 Research Question 3

The final research question—*"Does EoL circularity contribute to the sustainability of established and emerging PV technologies?"*—is addressed in Article 4. While the effects of circularity on individual indicators were covered by RQ 2, RQ 3 refers to evaluating these effects together. In the perovskite and Si PV systems, carbon footprint and LCOE were found to change in the same direction with variations in EoL circularity—in the perovskite system, both carbon footprint and LCOE increase with increasing circularity, while in the Si system, both decrease. Therefore, it can be stated that circularity does contribute to sustainability in the Si system, while it does not contribute to sustainability in the perovskite system as defined in this work. In the tandem system, on the other hand, carbon footprint tends downwards and LCOE upwards with increasing circularity, indicating that a trade-off exists between these two dimensions with respect to circularity. In other words, complete EoL circularity cannot be assumed to deliver the most sustainable solution in both the environmental and techno-economic dimensions. Instead, with all other things constant, a degree of circularity that simultaneously minimizes carbon footprint and LCOE exists in the tandem system. This finding directly addresses and validates the argument that the contribution of circularity to sustainability must be assessed, as circular strategies are not automatically guaranteed to deliver the most sustainable outcomes (e.g. Korhonen et al. 2018).

7.2 Novelty and Implications

By answering the research questions, the methodological and PV system assessment shortcomings or gaps identified in Sections 1.5.1 and 1.5.2, respectively, are also addressed. With regard to the methodological shortcomings, process simulation is presented as a highly granular alternative to the MFA method, which is rightly based on the concept of mass conservation, but does not account for the creation of entropy and the complexities surrounding the movement of small amounts of minor metals, their various compounds, and solutions through product life cycles. Process simulation itself is not new. What is novel, however, its application in the sustainability and CE arenas to simulate entire life cycles at the process level, and to simulate the life cycle system's response to closed-loop recycling at the same level of detail. Apart from the higher-than-usual inventory data quality, further novelty in this dissertation lies in the application of generated results post-simulation. The use of exergy efficiency and exergy cost as more comprehensive indicators of resource-related performance and the subsequent identification of resource efficiency hotspots (as is done in Article 2), for instance, has not been done for PV systems based on the extant literature. Such an SLT-based analysis is neither an automatically generated result, nor a requirement for a simulation model to work, but it cannot be done unless rigorous process simulation is done first and cannot be done at all using only MFA. This significantly extends the set of suitable indicators for sustainability assessment in the CE context.

Significant novelty is introduced with the combined use of NN-based surrogate functions to facilitate efficient and simultaneous calculation of the selected resource efficiency and sustainability indicators over complete ranges of two circular strategies. The degree to which circular strategies are or will be implemented depends on various factors, for example, the availability of technologies that produce high-quality recyclates, attitudes towards using secondary materials, and associated raw material market conditions, among several others. Therefore, it is useful to evaluate the indicator of interest over ranges of circularity, as opposed to only at a specific operating point. The latter does not provide information about the direction in which an indicator would change, or by how much, if a change in circularity were to occur. Leveraging the benefits of using NN-based surrogate functions, Articles 2, 3, and 4 show both the direction and the relative magnitude of changes in production capacity due to the increased availability of secondary resources, power consumption, nominal PV power generation capacity, $EROI_{PE\text{-}eq}$, carbon footprint, MSP, and LCOE. By examining sustainability indicator trends over ranges of circularity, the contribution of circularity to specific aspects of sustainability, and the emergence of trade-offs can be evaluated in a straightforward manner, while the inventory data for these assessments always remain aligned.

Whereas assessments of environmental and techno-economic performance are generally done in isolation, the inventory data is considered the foundation of all other assessments in this work to ensure consistency—assessments of resource consumption, resource efficiency, environmental impacts, and techno-economic performance are all based on the physics-based mass and energy balances calculated in the simulation model for each system. Furthermore, the closed-loop recycling that facilitates circularity is modelled in the foreground system, reducing the number somewhat arbitrary methodological choices that would have to have been made if typical approaches had been adopted. The approach developed in this work allowed for the resource and sustainability indicators to be linked to circularity via the detailed inventory data, which facilitated identification of the environmental/techno-economic trade-off with respect to circularity in the tandem PV system. In the process, the PV system assessment shortcomings are also addressed automatically. Furthermore, the PV systems selected for analysis and comparison are highly relevant technologies receiving considerable academic and industry attention at present. As the perovskite-based systems are emerging, the findings presented in this dissertation are expected to contribute useful insights to these technology development communities about perovskite-containing systems' sustainability performance so that life cycle sustainability can be built into their design.

In the European Green Deal communication of 2019, the stable supply of CRMs from both primary and secondary resources is considered a strategic security issue to achieve its 2050 climate neutrality goals (European Commission 2019). CRMs and other minor elements that are crucial for the functioning of sustainability-driving technologies can only be traced through life cycles using methods that consider thermodynamics and solution chemistry in addition to mass and energy conservation. This is especially relevant in dynamic arenas like the development of renewable energy generation and storage technologies that are constantly evolving—the resource flows through these life cycles and the resulting sustainability impacts need to be predicted, as historical data do not exist. Methods that only consider mass balances based on historical data cannot provide a true representation of the resources needed to re-extract these materials for reuse in a circular economy and the subsequent sustainability implications. The approach developed in this dissertation does that and is, therefore, ideally placed to support decisions pertaining to decarbonisation and sustainability, the transition from linear to circular economies, and to achieving the objectives of the European Green Deal. Furthermore, the approach can be used, for example, to directly link the physical flows of CRMs to the outcomes of specific policy measures. The linking of resource flows, their resulting environmental impacts, and associated cost impacts, all as functions of circularity, allows for the effects policy measures like carbon taxation to be evaluated directly.

8 Conclusion and Outlook

The transition to sustainable and renewable energy supply is crucial for achieving the SDGs and climate goals. The development and deployment of renewable energy technology need to accelerate to meet carbon emission reduction targets and solar PV has a critical role to play. Recent forecasts indicate that annual PV deployment needs to increase considerably from current levels (Haegel et al. 2023). PV systems combine two key sustainability-related themes: on the one hand, they deliver clean energy during their use phase and on the other, many of the chemical elements needed to produce PV systems are CRMs, making their mindful consumption and EoL management imperative. The required PV industry growth will result in a proportional increase in material and energy demand for production, as well as an increase in waste when these systems reach the point where recycling can no longer be postponed through, e.g. reuse or refurbishment. The implementation of EoL CE strategies could help to displace at least some of the primary extraction and production of materials needed to realise this growth sustainably. A precondition is that circularity-enabling recycling processes, infrastructure and business models are developed to recover materials from EoL modules at purities that permit their reuse in PV life cycles rather than being downcycled into lower-value applications. In doing so, recovered materials remain in the economy at their highest value. Additionally, the design and optimisation of processes to run at their thermodynamic limits would maximise resource efficiency. Before any such measures can be implemented, their current and/or potential future performance need to be assessed or predicted from a holistic sustainability perspective to ensure that the desired outcomes would be achieved. To measure and predict their effectiveness and efficiency, rigorous methods must be used to quantify physical flows and the resulting environmental, economic, and social impacts.

The work presented in this dissertation has two dimensions. The first is the development of an approach that allows for the rigorous type of sustainability and circularity assessments mentioned above. The second is the application of the developed approach to assess the performance of contemporary and emerging PV systems. The dissertation draws upon concepts from several disciplines—process engineering and design, industrial ecology, environmental economics, business administration, policy, and circular economy—to address the overall objective to explore and quantify the links between circularity and sustainability for complex technologies. The objective is achieved by addressing the RQs, which has resulted in the quantification of resource consumption (in terms of exergy cost) and efficiency (in terms of exergy efficiency and $EROI_{PE\text{-}eq}$), carbon footprint, and techno-economic performance (in terms of MSP and LCOE), all aligned with one physics-based inventory dataset for each of the PV systems investigated. Furthermore, the responses of these indicators to changes in circularity, primarily that of Si recovered at EoL, is revealed.

As discussed in relation to RQ 3 (Section 7.1.3), whether circular strategies increase sustainability is not a given, and depends on the characteristics of the specific life cycle system under investigation. In this dissertation, three different answers were obtained for three different PV systems. Examples of such analyses and conclusions have not been found in the published literature. The author concurs with the assertion that it should not be assumed that all circular strategies automatically enhance sustainability—such statements need to be tested comprehensively, especially for complex product systems that require sophisticated EoL management processes to recover often small quantities of valuable and critical secondary resources at high purities. Circular approaches should not be 'advertised' as being able to achieve the unachievable (i.e. complete circularity), as this would lead to an underestimation of the amount of effort needed to achieve sustainable development into the future, potentially leading to mediocre progress.

As with any simulation model, those developed in this work remain abstractions that cannot fully represent real-world complexity. Furthermore, because emerging technologies are, per definition, not fully developed, operational data for model validation are not available. However, the fundamental basis from which the simulation models have been developed alleviates some of the uncertainty associated with predicting the performance of emerging processes and products—not unlike the standard practice of using process simulation as one of the first steps in designing industrial production and refining facilities in engineering design consultancies. Nonetheless, models such as those presented in this dissertation need to be validated with operating data when available and updated accordingly, if required.

It should be noted that the results obtained for the specific PV systems analysed in this work cannot be directly applied to other PV systems due to the numerous types of cells and modules that exist (e.g. in terms of the materials used, manufacturing methods, and assumptions around electricity mixes, solar insolation, and PCE, among many others). A simulation model needs to be customised or developed anew for the specific system being analysed. Furthermore, although considered realistic based on the current state of the art, changes in some of the assumed parameter values could result in different conclusions from this work. Local sensitivity analyses have been done for important economic parameters (see Chapter 6 and Annexure D), and in essence, the majority of results presented represent the sensitivities of selected resource and sustainability indicators to circularity. However, due to the substantial number of parameters in each of the simulation models, it was not possible to conduct sensitivity analyses for all. This would be a data-intensive exercise requiring significant computer processing power but should be addressed in more detail in future work.

While the term sustainability is referred to throughout this dissertation, only environmental and techno-economic life cycle performance were assessed. To gain a truly comprehensive understanding of the potential for specific life cycles to contribute to overall sustainability, future work should expand the approach to also include the assessment of social impacts. With regard to environmental impacts, only the global warming potential and acidification potential midpoint impacts were assessed in Article 1, and only carbon footprint was considered in Articles 2 and 4. The analysis of carbon footprint only was considered valid as previous studies have shown that other indicators relevant to PV system assessments generally tend in the same direction as carbon footprint (see Article 4, Annexure D). However, the risk of shifting environmental impacts from one impact to another, or from one life cycle stage to another is still present. Future work should consider this aspect of environmental impact assessment more comprehensively by including additional impact categories.

While the trade-off identified in Article 4 in the tandem PV system is only described qualitatively, a future research avenue might be the further development and application of the NN functions in other approaches that aim to find optimal solutions. One such example is reinforcement learning (RL), in which a system uses a reward-driven trial-and-error process for a so-called agent to learn to interact with a complex environment to find the most rewarding solutions (Aggarwal 2018). NNs are often used as function approximators to facilitate decision making in RL (Ibid.). Here, the complex environment would be the simulation model's solution space, which, as is shown in this work, can be represented by NNs. In this way, the combined use of the NN-based surrogate functions and reinforcement learning could be used to quantify optimal levels of circularity and other parameter values that maximise overall sustainability. The trial-and-error process usually takes place in a live or online environment where the agent can interact with the environment directly. However, it would have to use a static information database in this case, which is also referred to as batch RL, or off-policy RL. While this approach presents additional challenges, considerable progress has already been made towards successful implementation using new classes of algorithms (Fujimoto et al. 2019).

This dissertation has contributed to sustainability and CE research by developing a high-resolution, physics-based approach for analysing complex product systems and their resource, environmental, and techno-economic performance at the process level. At the same time, the responses of these to the introduction and variation of circularity are analysed. Exergy efficiency and exergy cost have also been proposed as additional resource hotspot and sustainability indicators for the CE. Furthermore, it has provided PV-specific insights that researchers, industry, and policymakers can use to guide their decisions. In the Si PV case, for instance, results have quantified and highlighted the importance of recycling

Si at high grade in order to avoid the parts of the value chain with the highest energy consumptions and hence, the highest environmental impacts. To recycle and recover maximum quantities of materials at qualities high enough to keep them in the life cycle requires research and innovation, and the infrastructure needed to run the developed processes. These activities need the commensurate funding and investment. Results can thus be used to direct research efforts to design materials and products for circularity and sustainability, to develop sophisticated recycling processes, and to guide funding and investment decisions, in the process contributing impactfully to achieving the SDGs. Additional applications of the approach discussed in the preceding text and publications, e.g. on the effects of material intensity, the use of alternative production technologies, electricity grid location, supply chain location, and carbon taxation similarly provide useful quantitative information that can be used to guide policy and industry decision making. With emerging PV technologies, for instance, the simultaneous environmental and techno-economic effects of semiconductors and/or solvent substitution could guide decisions about which technologies to combine in tandem PV systems to avoid trade-offs such as that identified in Article 4, again contributing to achieving the SDGs.

The dissemination of this work to date has already generated considerable interest, both academically and in industry, with further discussions and work underway. In the bigger, longer-term picture, continued dissemination, adoption and further development of the approach can enhance the quality of sustainability and circularity assessments, particularly in the context of global climate change goals and rapidly developing renewable energy generation and storage technologies for which no operational data are available yet. The approach is, however, in no way limited to the PV systems analysed in this dissertation. It can be applied to any complex product system and calls for transdisciplinary collaboration, as such investigations combine knowledge and experience from several fields to create a shared understanding of complex systems and problems.

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Annexure A: Article 1

The simulation-based analysis of the resource efficiency of the circular economy – the enabling role of metallurgical infrastructure

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RESEARCH ARTICLE

The simulation-based analysis of the resource efficiency of the circular economy – the enabling role of metallurgical infrastructure

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ABSTRACT
Process metallurgy is a key enabler and the heart of the Circular Economy (CE). This paper shows the state-of-the-art approach to understanding the resource efficiency of very large-scale CE systems. Process simulation permits system-wide exergy analysis also linked to environmental footprinting. It is shown that digital twins of large CE systems can be created and their resource efficiencies quantified. This approach provides the basis for detailed estimation of financial expenditures as well as high-impact CE system innovation. The cadmium telluride (CdTe) photovoltaic technology life cycle, which brings several metal infrastructures into play, is studied. The results show that considerable work remains to optimise the CdTe system. Low exergy efficiencies resulting specifically from energy-intensive processes highlight areas with the greatest renewables-based improvement potential. This detail sheds light on the true performance of the CE and the inconvenient truth that it cannot be fully realised but only driven to its thermodynamic limits.

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Circular economy; exergy; life cycle assessment; metallurgy; photovoltaics; resource efficiency; sustainability; digital twin

Nomenclature

*a*LCA	Attributional life cycle assessment	GWP	Global warming potential
AP	Acidification potential	IRR	Internal rate of return
BF	Blast furnace	LCA	Life cycle assessment
BOF	Basic oxygen furnace	LCC	Life cycle costing
BOS	Balance-of-system	LCI	Life cycle inventory
CAPEX	Capital expenditure	LCIA	Life cycle impact assessment
CdS	Cadmium sulphide	LCT	Life cycle thinking
CdTe	Cadmium telluride	NPV	Net present value
CE	Circular economy	OPEX	Operating expenditure
CED	Cumulative energy demand	PV	Photovoltaic
CFD	Computational fluid dynamics	RC	Resource consumption
CIGS	Copper-indium-gallium-diselenide	RE	Resource efficiency
DfR	Design for recycling	REE	Rare earth element
DZS	Direct zinc smelting	RLE	Roast-leach-electrowinning
EAF	Electric arc furnace	SD	Sustainable development
eLCC	Environmental life cycle costing	S-	LCA
Social life	cycle assessment		
EoL	End-of-life	SLT	Second law of thermodynamics
EP	Eutrophication potential	SWB	Solid waste burden
GHG	Greenhouse gas	TPS	Thermodynamic process simulation

Introduction

Circular Economy (CE) promotes the use of waste materials as resources instead, transforming our economies from linear to circular models. At its core lies the responsible use of all resources – human, natural, and economic. Sustainable development (SD), which encompasses the social, environmental and economic dimensions (Beaulieu et al. 2015), and life cycle thinking (LCT) – which aims to increase resource efficiency, decrease environmental impact, and improve the social and socio-economic performance of products (or services) over their entire life cycles (Life Cycle Initiative 2017) – go hand in hand with the CE concept (Beaulieu et al. 2015). It aims to gain control of and limit our negative impacts on the planet and to ensure the welfare of future generations. Whilst the first priority is generally to extend product life cycles by as much as possible through waste hierarchies following various *R* frameworks such as reduce, re-use, refurbish or remanufacture, the ultimate fate of any product is end-of-life (EoL) recycling, which aims to close material loops to maximise resource efficiency. The less frequently discussed bad news is that no such loop can be closed completely, as any real transformation process will always be subject to inevitable losses and inefficiencies. Our aim should be to identify and minimise these along entire product life cycles (Reuter et al. 2019). This will be elaborated upon later.

CE is studied by many disciplines (Lieder and Rashid 2016), and many approaches exist to evaluate different CE-related issues at varying levels of detail (Finnveden and Moberg 2005; Jeswani et al. 2010;

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Pihkola et al. 2017). Some of these have been integrated for more holistic assessment, but examples for products that contain complex material combinations are limited. This limitation will be addressed in this paper.

Minerals and metals are vital to modern society, equipping most technologies – including those that harness renewable energy such as photovoltaics, wind turbines and energy storage devices – with structure and functionality. Consequently, the innovation necessary to drive SD would not be possible without them. In addition to the typically considered bulk base metals, increasingly complex combinations of valuable precious and special metals – most of which are produced as by-products of other metals (Bleiwas 2010) – impart very specific functionalities to complex products (UNEP 2013). Some are considered *critical* in terms of supply risk and increasing cost of extraction from ores whose metal concentrations are on the decrease (Frenzel et al. 2017). The metallurgical infrastructure and knowledge necessary for the sustainable extraction and recycling of these metals such as iron (Fe) and steel, aluminium (Al), copper (Cu), lead (Pb), zinc (Zn), tin (Sn) and their associated minor elements are, therefore, all central to and critical enablers of the CE (Reuter 2016).

The interconnectedness of metals is often overlooked in analyses of material flows. Analogous to physical and chemical interactions in geological ores and minerals (Schouwstra et al. 2000; Frenzel et al. 2017), complex metal and other material combinations exist in man-made urban minerals. Some of these can contain in excess of forty different metals, none present in pure form (e.g. Cucchiella et al. 2015; Ballester et al. 2017). Accounting for any of the metals or compounds in isolation underestimates resource requirements, losses and impacts associated with the life cycle in which they participate. It should be avoided by adopting a mineral- or product-centric approach (Reuter and Kojo 2012), meaning that a product is treated as a whole – not merely as the combination of various pure elements that are analysed one by one. This is particularly important for complex products that contain one or more by-product metals.

The typical life cycle of a generic metal-containing product is shown in Figure 1 with the metallurgical processing value chain represented by the top half in blue. Apart from primary extraction, Figure 1 shows an apparently closed loop. It should be remembered, however, that leakage of materials and useful energy occur at every step along the way. The re-use and re-manufacturing loops extend the life cycle until recycling can no longer be avoided. The design for recycling (DfR) loop provides recycling efficiency feedback to product designers so that designs can be enhanced to maximise product sustainability. For the adoption and performance measurement of CE, participants in the life cycle need to collaborate. In addition to creating desired products, outputs include social, economic, and environmental impacts, and processes that aim to mitigate impacts and losses are subject to resource inputs, losses and impacts themselves. Human behaviour has a significant influence on circularity – consumer/user decisions determine whether sustainable products are purchased, when the EoL stage will be entered and the amounts of EoL products collected and recycled to (almost) 'close the loop' (Cleveland et al. 2005; Young et al. 2009).

In Figure 1, coloured blocks represent gatekeepers that strongly influence progression from one life cycle stage to the next (see legend). For example, consumer decisions (blue) affect progression from *Use* to *Collection* – if the decision is not made to recycle an EoL product meaningfully, the CE loop for that product opens up and ceases to exist. Decision making is affected by complex interactions of personality, motivation, demographics, and many other factors (Kalliath et al. 2014), which cannot be modelled without significant assumptions. Market forces (green) act between *Collection* and *Recycling* – not everything collected for recycling will in fact be recycled, as this depends on, amongst others, the market dynamics surrounding particular metals at the time. Therefore, it is highly unlikely that recycling replaces primary extraction on a one-to-one basis (Geyer et al. 2015) – there needs to be an economic incentive. Politics, policy, and regulation all have strong influences on the running of any CE.

The power generation sector has been identified as having the highest potential for cutting, and potentially eliminating, its greenhouse gas (GHG) emissions by 2050 (EC 2015). With electric vehicles, wind power and concentrated solar power generation, solar photovoltaic (PV) technologies are set to play a significant role in achieving climate change mitigation targets. Thin-film PVs require specialty metals like cadmium (Cd), gallium (Ga), indium (In), selenium (Se) and tellurium (Te) for their functionality. These include e.g. CdTe, amorphous silicon and CIGS (copper-indium-gallium-diselenide) thin-film solar cells (Elshkaki and Graedel 2013). Emerging third-generation technologies include e.g. perovskite solar cells, which utilise lead (Pb) in the organic/inorganic perovskite molecular structure. As an example of one of these complex products, the life cycle of a CdTe PV cell is shown in Figure 2. It consists of several distinct production and manufacturing processes with clearly defined system boundaries. In CdTe cells, the second semiconductor is a layer of cadmium sulphide (CdS). These gate-to-gate processes – all geographically dispersed – are linked to one another by global supply chains. The situation would be even more complex in the case of e.g. CIGS PV cells, as primary Ga is largely a by-product of Al production, which would then additionally introduce the Al production system into Figure 2. In the case of

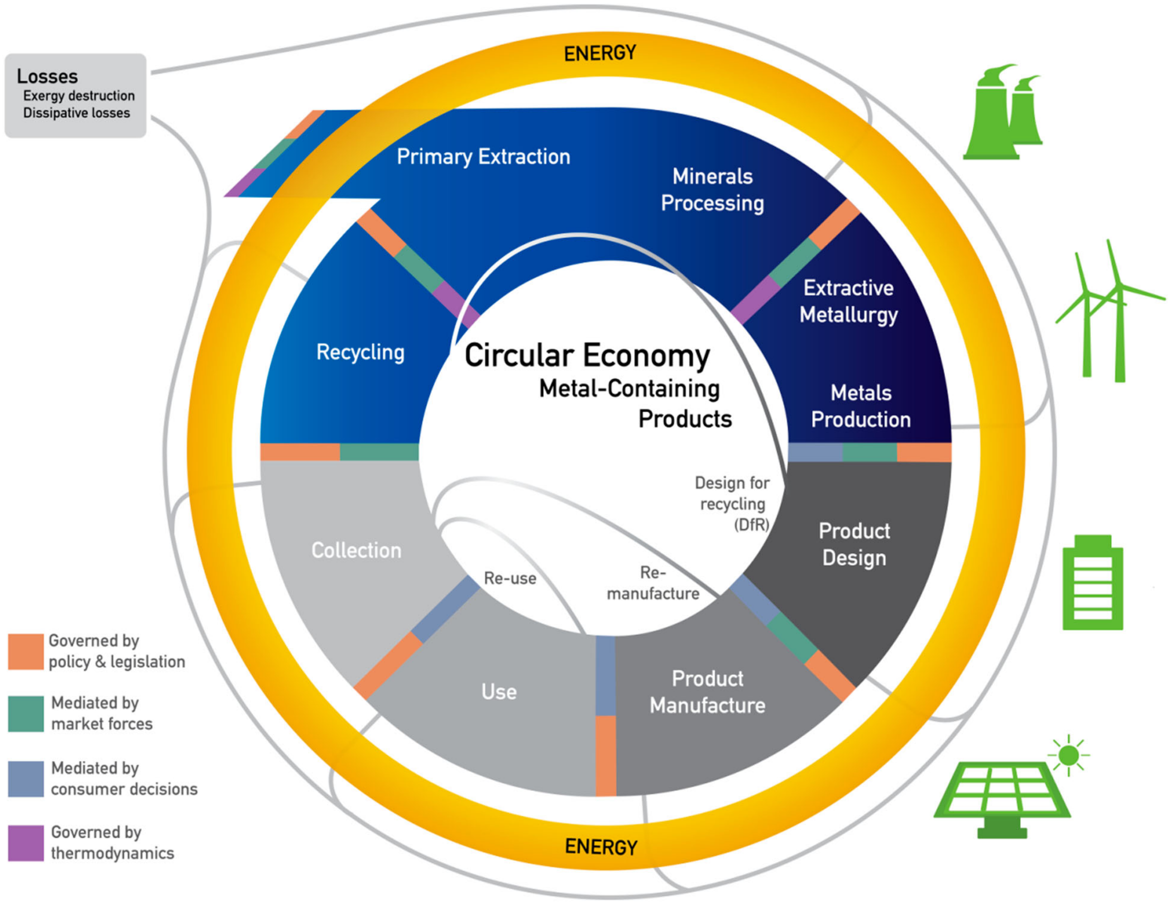

Figure 1. CE loop of a generic metal-containing product including the key mediators between the different stakeholders.

wind power, one would have to include systems for the production and recycling of REEs such as neodymium (Nd), which is used in permanent magnets.

With respect to interactions *between production systems*, Figure 2 demonstrates that – to produce the semiconductor layers for this technology – the Cu, Zn and Pb production value chains are all needed, the reason being that Cd is primarily a by-product of Zn and Pb production (usually integrated) and Te a by-product of Cu as a consequence of usually being associated in ore minerals. The same applies to secondary production (recycling) of these metals. If metals collected for recycling are diverted to an incompatible production system (e.g. Fe/steel in the case of Cd and Te) they would most likely be lost (Verhoef et al. 2004). The absence or removal of any of the primary production systems would collapse or limit the CdTe PV circular production capacity. The limiting factors would be the quantities and qualities of metals that could still be economically extracted from primary and secondary resources using only the production infrastructures that remain. With respect to interactions *within products*, Figure 2 shows that once pure Cd and Te have been joined – through powder production, smelting and atomisation processes (Fthenakis 2004) – to produce the CdTe semiconductor layer, their flows remain linked until such time that they are physically separated into their pure forms or into compounds in other by-products. Ignoring these interactions in sustainability assessments results in the omission of resource requirements for the separation and reuse of the metals, as well as the associated environmental, economic, and social impacts, in the process overestimating efficiency and underestimating impacts. This highlights the importance of product-centric approaches, and the inferiority of material-centric ones when it comes to complex products.

The infrastructure shown in Figure 2 would, of course, not exist without construction materials like steel. Although not shown in detail in Figure 2, a brief description of steel production and recycling is given here. Iron and steelmaking processes are very energy intensive and emit considerable amounts of carbon dioxide (CO_2). Crude steel is, in principle, produced via two routes: the blast furnace-basic oxygen furnace (BF-BOF), and electric arc furnace (EAF) routes. Whereas basic oxygen furnaces (BOF) in the BF-BOF route can be charged with up to 30% steel scrap, EAFs can treat inputs consisting entirely of

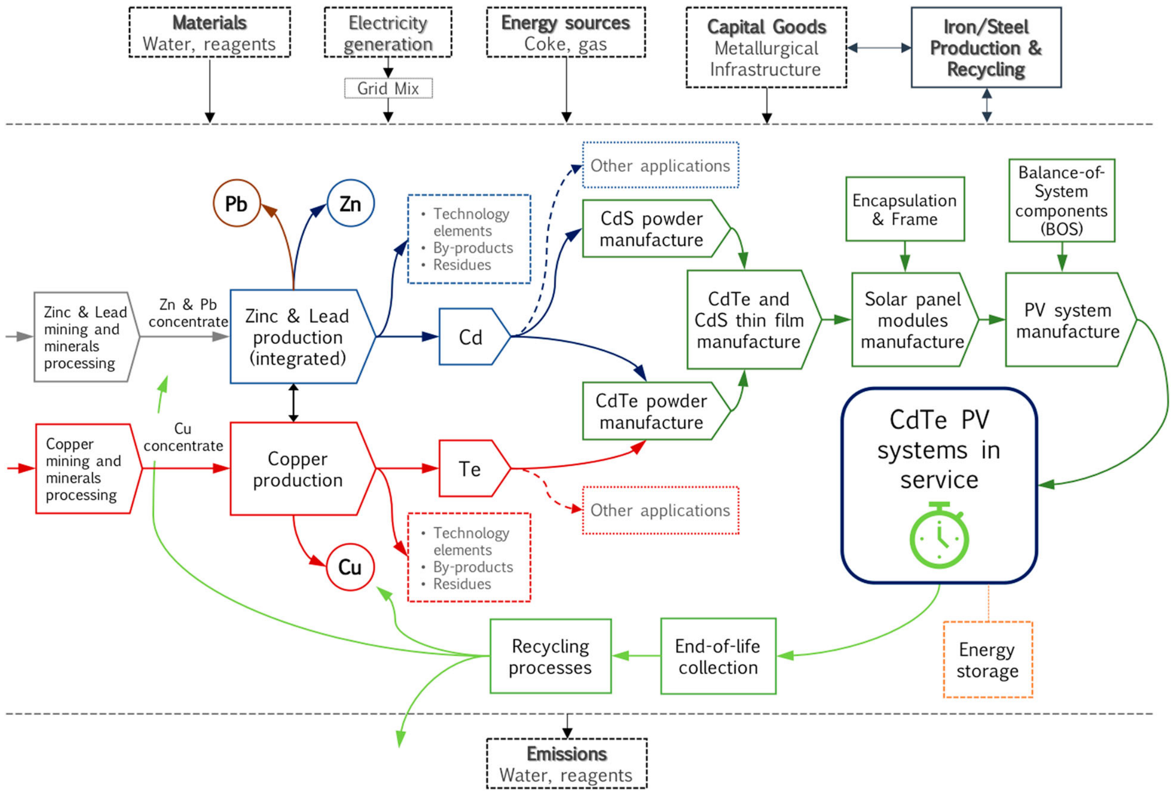

Figure 2. Full life cycle of CdTe PV systems including base and by-product metals production infrastructure and end-of-life processing.

steel scrap. Even though steel is a material that can be completely recycled, however, only 28% of crude steel is produced via the EAF route worldwide (worldsteel 2019). Constraints on the use of EAFs for production, and for the replacement of BOFs, are higher production prices, a lack of reliable power supply and steel scrap, and the impossibility of producing certain steel grades in an EAF. For example, the production of flat products in EAFs is hardly possible today (Grummes 2019). On average, 780 kg of coal is used today to produce one tonne of crude steel using the BF-BOF route (worldsteel 2019). Of course, many current research projects such as SALCOS (Hille and Redenius 2018) focus on replacing carbon with hydrogen. Unfortunately, it is not technologically possible to produce steel completely carbon-free. Carbon is used in the EAF mixing process for CO-bubble formation, for EAF slag foaming with the aim of reducing energy consumption and refractory wear, for crude steel deoxidation and as an alloying element. In 2017, on average, 1.83 tonnes of CO_2 were emitted per tonne of steel produced (worldsteel 2019).

Steel is not the only useful steelmaking product. By-products include slag, dust, exhaust gas, steam, and waste heat. In 2018, for example, 14.2 million tonnes of slag were produced in Germany, of which 38% originated from steel plants (Merkel 2019). The slag was used in the cement and construction industry, as fertiliser and as slagging agent during steelmaking. Only 11% went into intermediate storage or ended up in slag heaps in that year (Merkel 2019). Dust is briquetted and used as Fe carriers. Slag and dust contain valuable elements such as chromium (Cr), vanadium (V), molybdenum (Mo), phosphorous (P), and rare earth elements (REE), which are very often lost. Due to poor sorting of scrap, the contents of tramp elements such as Sn and Cu in steel increase over time, worsening the mechanical properties of the finished steel product. These losses and downcycling effects highlight that the possibility might exist to establish symbioses with other systems similar to that shown in Figure 2 for Cu, Pb, Sn and Zn to optimise overall sustainability (see Metal Wheel, Reuter et al. 2019). The useful waste heat potential in German integrated iron and steel plants was estimated to amount of 0,322 GJ per tonne of solid crude steel (Sprecher et al. 2019), offering further opportunities for maximising sustainability.

To assess the impacts of such exceptionally large systems in all three dimensions of sustainability, a robust digital foundation that captures the quantities and qualities of all relevant stocks and flows in the life cycle is required. The utilisation of consistent input data, generated in the physical flows layer (shown in Figure 3), across the environmental, economic, and social dimensions – as opposed to disjointed analyses with potentially incompatible system boundaries – facilitates robust sustainability assessment. As alluded to in a previous paper (Reuter et al. 2019), digitalisation – starting with physical quantities and qualities – is key to such assessments.

Using the approaches outlined in the remainder of this paper, resource consumption, resource efficiency and environmental impacts are analysed for the extensive metallurgical infrastructures that need to be in place to drive the CdTe PV life cycle – as an example of a complex technology – at the process level of detail. It is shown in this paper how a simulation-based approach using metallurgical process simulation tools are used to analyse the CE system. This paper will use CdTe PV technology to show the state-of-the-art metallurgical simulation tools used to analyse the CE, considering both steel and non-ferrous metals production.

Simulation tools and digital platforms

Figure 4 shows the modelling and simulation platforms that are required to analyse large-scale systems. The various aspects reflected include:.

Physical Quantity & Exergetic Quality: The various tools that are applied to define the quality in each stream are, amongst others, thermodynamic simulation using FactSage (FactSage 2019), computational fluid dynamic (CFD) modelling, process simulation with tools such as HSC Sim for exergy analysis and energy balance models, data analysis including artificial intelligence (AI) and big data methods. Not to be neglected is industrial experience.

Environmental Impact: Life cycle assessment (LCA) is widely applied for the assessment of potential environmental impacts. The software tools used to conduct LCA are GaBi (thinkstep 2019) and openLCA (GreenDelta 2019). Relevant data are exported from process simulations into these software tools to conduct LCAs that are compliant with the ISO standards (ISO 2006a, 2006b) and in line with guidelines for PV systems (Stolz et al. 2017) and the mining and metals industries (Santero and Hendry 2016).

Economic Impact: Economic impacts are assessed using techno-economic methods that consider e.g. capital and operating expenditure (CAPEX and OPEX), amongst others, life cycle costing (LCC) and environmental life cycle costing (eLCC). Thermoeconomics provides a link between resource consumption (via exergy analysis) and economics. While outside the scope of this paper, the necessary tools for economic impact assessment on the simulation platform are available.

Social Impact: The systematic assessment of social impacts is considered to still be in the developmental phase and therefore, few relevant case studies have been published. The UNEP guideline for social life cycle assessment (S-LCA) is a good starting point for developing approaches to assess social and socio-economic effects. Acknowledging its importance, further work is required to incorporate social impact assessment into the simulation framework.

The above points will be discussed in more detail below with reference to the simulated case study for CdTe PV modules within a CE paradigm. The exergy basis permits the linkage of the energy production system and its exergy destruction with that of the metals production system.

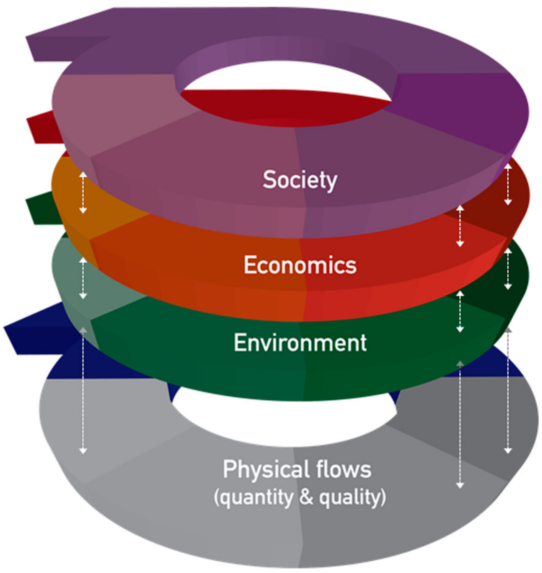

Figure 3. Input data for sustainability assessments originating from the physical quantity & quality layer, scaled and simplified into a form that society understands, and policy can convert into useful strategy.

Modelling and simulation of resource consumption and efficiency of the CE

In this Section, the analysis and assessment of physical flows – the bottom layer in Figure 3 and left-hand circle in Figure 4 – are discussed.

The laws of conservation state that mass and energy *quantities* are always conserved. Their *qualities*, however, are not. In this paper, resource *consumption* (RC) refers to the transformation – in terms of quantity and/or quality – of flows within a system. Resource *use*, on the other hand, refers to throughput without transformation. It is worth pointing out, then, that methods based solely on the laws of conservation (e.g. material and energy flow analysis) analyse resource use rather than consumption, evaluating a system's interaction with its environment in terms of throughput without considering changes in resource qualities resulting from transformations within the system (Gößling-Reisemann 2008).

The second law of thermodynamics (SLT) states that energy quality – in both material and energy streams – undergoes irreversible degradation in all

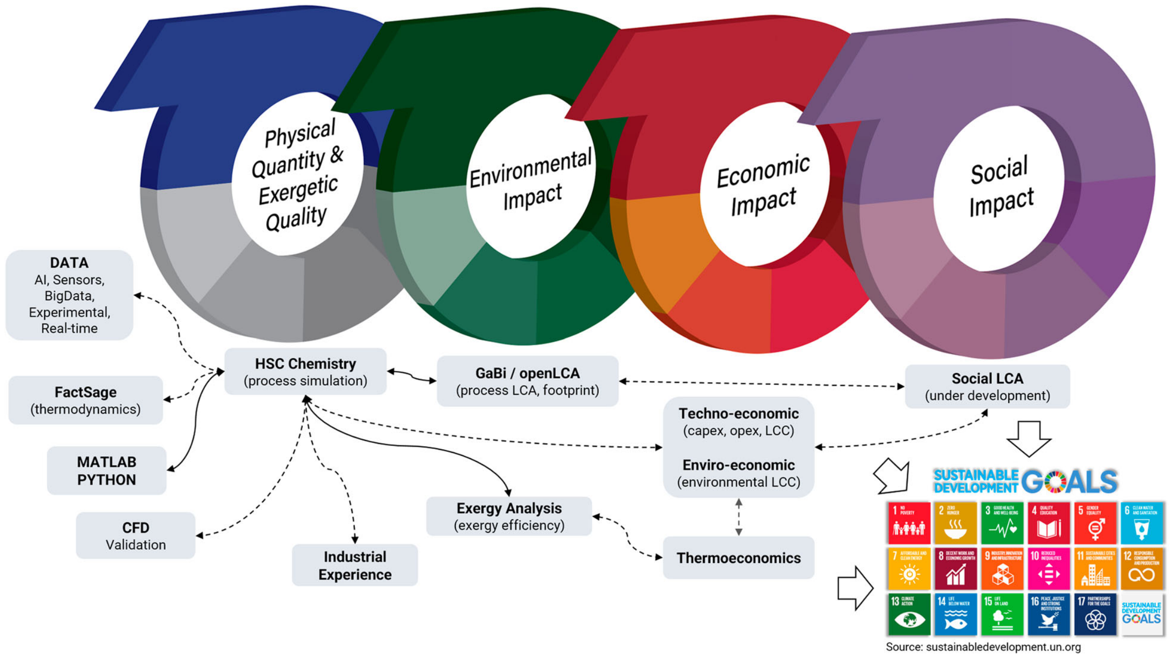

Figure 4. The various tools and methods that have to be integrated to analyse the various cycles of CE and progress towards achieving sustainable development goals i.e. the physical description, environmental, economic and social impacts (solid lines represent direct software connections, while dotted lines represent connections that, at this stage, require manual intervention). These methods are discussed in various forms in this special edition.

real-world transformation processes due to inevitable increases in entropy (Dincer and Cengel 2001). The law manifests through the loss and destruction of useful energy, and the dissolution of metals and other compounds into one another – in the direction of increasing entropy – to the detriment of product quality. To counter the resultant build-up of contaminants in desired product streams, dilution with virgin resources is needed – *in sharp contrast to the concept of closing a material loop*. The degradation of quality can act as a proxy for RC and the extent of degradation determines whether – and at what resource cost – EoL products could theoretically be valorised. This way, the circularity potential and the technical limits of CE systems – the degree to which degradation and losses cannot be avoided – can be determined.

Thermodynamic process simulation (TPS) provides the granularity that is missing from 'black box' modelling approaches (Jacquemin et al. 2012; Reuter 2016) by incorporating the physical and thermodynamic properties of minerals, metals and other compounds, and implicitly also the transfer and fluid dynamics processes that are used to calibrate process units in industrial reactors. Such platforms aid the development of representative, bottom-up models that counter the oversimplification of transformation processes as generally applied in the CE discussions. TPS captures the non-linear effects of dynamic material combinations and compositions on downstream processing and recovery efficiency (Reuter and van Schaik 2015) as well as the reality of industrial operations. These are functions of product design and the chosen processing route, and directly impact the recovery of materials and energy (Van Schaik and Reuter 2010; Bollinger et al. 2012; Reuter and van Schaik 2015). Furthermore, TPS provides a scientific basis for the allocation of RC and emissions to outputs, to identify inefficiency hotspots and to maximise RE throughout the system. In summary, the creation of these large-scale models requires a detailed understanding of industrial practice and systems and at the same time a fundamental understanding of each reactor such as described by Obiso et al. (2019) in terms of CFD.

Exergy analysis encompasses various methods that apply both the first and second laws of thermodynamics to show how effectively resources are being utilised, where losses occur, where there is potential for technological and operational improvements (Ness et al. 2007), and quantify flow degradation along product life cycles. It introduces a single unit of measure for the assessment and direct comparison of all inputs and outputs, both material and energetic (Ayres et al. 1998). The *exergy cost* accounting method, also referred to as cumulative exergy consumption (CExC) or demand (CExD), allows for calculation of a single total exergetic cost for each product by starting with raw materials and accounting for all feedbacks, wastes and recycles along the value chain (Dewulf et al. 2007). It allows for the objective comparison of total RC for many different product types. As with other life cycle approaches, proper allocation is important in multi-product systems. This requires the definition of clear system boundaries, the productive structure of the system, interactions between components, and the production purpose for each component (Valero 2006), all of which are implicit in TPS. It does not involve any monetary calculations (Dewulf et al. 2007). Exergy *efficiency* is useful as an indicator for the theoretical improvement potential of a process (Ayres et al. 1998), to compare process configuration options, and as an optimisation parameter in system improvement problems (Ignatenko et al. 2007). Because it accounts for both material and energy flows, exergy efficiency also represents resource efficiency (RE) for our purposes. Hepbasli (2008) mentions two types commonly used: *brute force*, which refers to the ratio of total exergy outputs and total exergy inputs, and *functional*, which refers to the ratio of the exergy associated with the desired output to the input exergy required to achieve that output. For the purpose of this paper, we define the numerator in the efficiency ratio for a process as the total exergy output excluding irreversibility[1] and the exergy contents of waste streams, therefore assuming that all other output streams are useful, or would be processed further. Our definition, therefore, lies between the *brute force* and *functional* efficiencies. It is worth highlighting that, whilst higher efficiencies are always desired, it is also possible to consume vast amounts of resources very efficiently. For this reason, we focus on RC, as discussed earlier, in addition to RE.

Building upon a recently conducted exergy cost and environmental impact analysis for a detailed mine-to-metal Cu production system (Abadías Llamas et al. 2019b), a detailed steady-state TPS model was developed for the entire CE system shown in Figure 2 using HSC Sim (Outotec 2019). This unique simulation model consists of 223 interconnected unit operations, 869 flows and around 30 different elements and their compounds. It simulates the integrated production of Cu, Zn and Pb as well as several co- and by-products including Cd, Te, cobalt (Co), silver (Ag), gold (Au) and others, and the transformation of several hazardous residues into inert materials for discard or further processing. Furthermore, CdTe PV cell manufacturing and recycling processes are simulated. The completion of models for steel production and recycling are currently underway. The individual metal production, PV manufacturing and recycling flowsheets are based on published best available processing routes and technologies (Sinclair 2009; Schlesinger et al. 2011; Wade 2013), as well as the authors' own industry knowledge and experience. Integration is achieved by creating Cu-Zn-Pb metallurgical symbioses through intra- and inter-process exchanges of compatible by-products and residues, some of which via an added intermediate slag fuming furnace. The main connections between the metal production chains are shown in Figure 5. The model is based on the amount of metals required to manufacture 2,000 m^2 of PV cells. The validation and calibration of this model is achieved through a mix of industrial experience, thermodynamic modelling (FACT Sage, Van Schalkwyk et al. 2018), CFD modelling (e.g. Obiso et al. 2019), process simulation experience (HSC Sim) and others as summarised by the mix of skills shown in Figure 4. There could still be inaccuracies, no doubt, but this paper shows that it is possible to bring together large systems for analysis.

The integrated model allows for the system-wide effects of various recycling and residue exchange scenarios – aimed at countering overall resource quality degradation and reducing dissipative losses – to be evaluated. The advantage of using HSC Sim as the digitalisation platform – in addition to being able to simulate non-linear material and energy transformations properly – is that it offers a built-in exergy analysis function. Furthermore, process simulation data can be mapped and exported directly for the assessment of life cycle environmental impacts – the subject of Section 2.2 below. Figure 6 shows the primary Cu smelting flowsheet, one of nineteen flowsheets that describe the system depicted in Figure 2. These are (from left to right): (i) Primary Cu smelting, (ii) gas cleaning, (iii) Cu reduction furnace, (iv) Cu/Co-Ni solvent extraction, (v) Co/Ni solvent extraction, (vi) precious metals recovery, (vii) Te production, (viii) electricity and heat production, (ix) sulphur capture, (x) oxygen production, (xi) Cu electrolyte cleaning, (xii) secondary Cu processing, (xiii) Zn roasting and leaching (RLE), (xiv) jarosite precipitation and Cd production, (xv) direct Zn smelting (DZS), (xvi) Pb production, (xvii) slag fuming, (xviii) PV manufacturing, use and collection and (xix) PV recycling. Selected flowsheets from the Zn and Pb production systems are shown in Figure 7.

Modelling and simulation of environmental impact – linking process simulation and LCA

In this Section, the analysis and assessment of environmental impacts – the green layer in Figure 3 and second from left circle in Figure 4 – are discussed.

Life Cycle Assessment (LCA) is useful and widely applied for the assessment of potential environmental and human health impacts of a process or product over its entire life cycle (ISO 2006a). It is used to assist in identifying improvement opportunities, to inform decisions at various levels of society, and to select environmental impact indicators, and it provides a means to communicate environmental impact information (ISO 2006a). It is also used for the identification

[1]The amount of exergy dissipated in a transformation process is referred to as the irreversibility of that process.

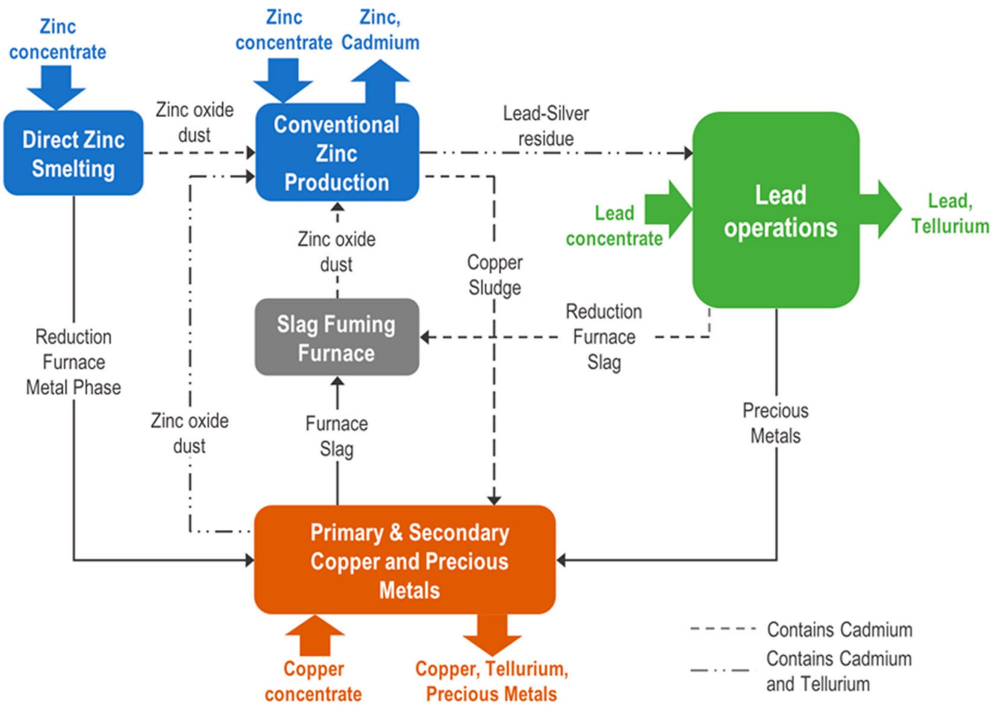

Figure 5. Metallurgical processing symbiosis of Cu, Zn and Pb with an added intermediate slag fuming furnace within the bigger flowsheet depicted by Figure 2.

and avoidance of environmental problem shifting between life cycle stages, between regions or from one environmental problem to another (Finnveden et al. 2009). Distinction is made between two modes of assessment – attributional and consequential LCA. The former focuses on the analysis of relevant environmental flows to and from the life cycle under investigation, and the latter on the wider effects of changes in these flows as a result of decisions at hand (Finnveden et al. 2009).

The LCA standard (ISO 2006a) defines Goal and Scope Definition, Inventory Analysis (LCI), Impact Assessment (LCIA), and Interpretation as the four stages in which LCA is performed – the latter occurring

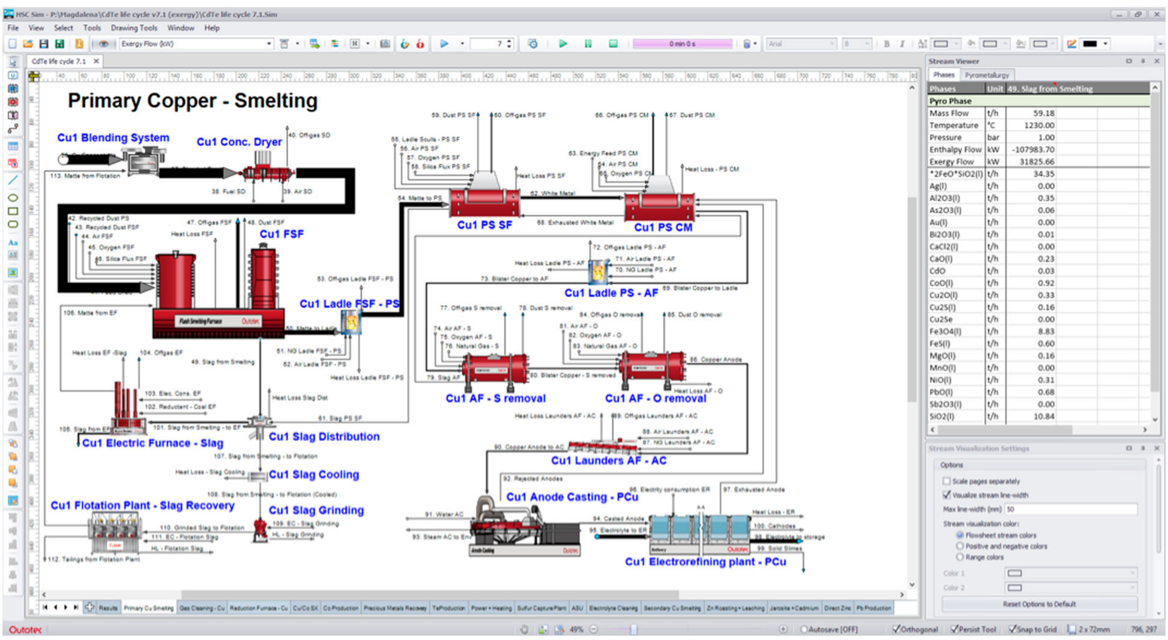

Figure 6. The Cu processing flowsheet of the complete simulation for Figure 2 with 16 of 19 flowsheet tabs shown at the bottom of the simulation pane – tabs 17–19 provide the CdTe module production and recycling.

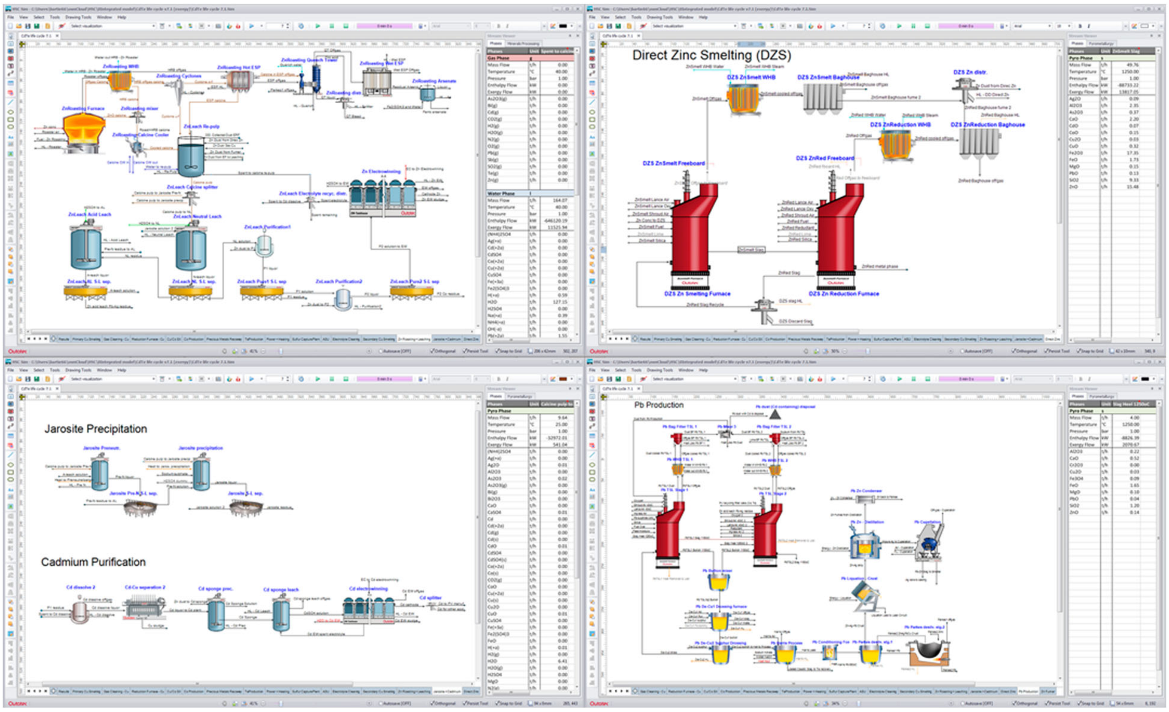

Figure 7. The detail of zinc processing and linked flowsheets (tabs 13–16 on the simulation pane) is shown with clockwise from top left (i) conventional Zn roast-leach-electrowinning, (ii) direct zinc smelting, (iii) jarosite precipitation and Cd purification, and (iv) Pb production.

alongside the three others. Allocation is required to proportion overall burdens between individual contributors in multi-input/output processes but is to be avoided as far as possible by expanding the system and disaggregating processes into more detailed sub-processes such that the part to be analysed can be isolated (Azapagic and Clift 1999). This is not always possible because some co-products cannot be produced in isolation (Klöpffer 2003). When unavoidable, allocation should be based on actual causal relationships between inputs and outputs in the first instance, and on economic value as a last resort (Ardente and Cellura 2012). Allocation procedures have been questioned, however (e.g. Ekvall and Finnveden 2001), and is one of the most debated issues in LCA (Finnveden et al. 2009). In the LCI phase – usually the most data and resource intensive phase of LCA – in- and outflows between the system and the environment are determined.

Numerous LCA studies have been conducted in the mining, minerals, and metals industries. They are typically used on their own to identify impact hotspots in existing processes, to assess and compare scenarios such as different technology options, or in combination with techno-economic assessments. The environmental impacts of primary extraction and production processes are usually assessed using a cradle-to-gate approach. This does not provide complete information for whole CE systems but makes sense for studies focusing on multi-metal production impacts. It also generates useful baseline data for primary production. A few relevant examples are mentioned here. Norgate et al. (2007) assessed and compared global warming potential (GWP), acidification potential (AP), solid waste burden (SWB), and cumulative energy demand (CED) for pyro- and hydrometallurgical Cu, Ni, Al, Pb, Zn, Steel and titanium (Ti) primary production processes. Khoo et al. (2017) compared the environmental impacts of three Ni laterite processing technologies for the input of Ni into stainless steel production using various impact assessment methods. Weng et al. (2016) conducted LCIA for the primary production of REE in terms of GWP and CED. Duan et al. (2018) compared the energy consumption, GWP, AP, eutrophication potential (EP) and other environmental impacts of new methods for the recovery of waste heat from blast furnace slags with those of existing approaches in the Fe and steel industry and Cherubini et al. (2008) reported GWP and AP for global magnesium (Mg) production. Van Genderen et al. (2016) conducted a global LCA for primary Zn production, in line with methodological guidance developed through a collaborative effort to harmonise LCA methodologies in the minerals and metals industry (PE International 2014). Abadías Llamas et al. 2019b conducted a mine-to-metal study for primary Cu production and compared GWP and RE for the system with and without waste treatment. These assessments all adopt a cradle-to-gate perspective. Burchart-Korol (2013) assessed the gate-to-gate impacts of primary

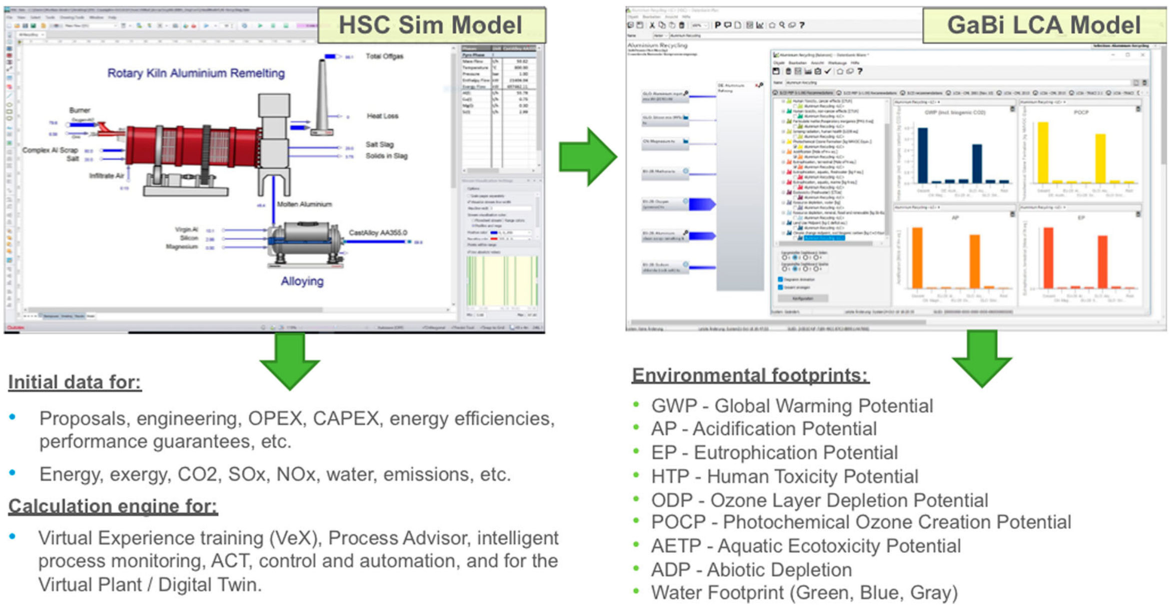

Figure 8. Linking HSC Sim (Outotec 2019) – which provides the usual engineering design data as well as process control basis – and environmental impact assessment tools such as GaBi (thinkstep 2019) or openLCA to produce the environmental indicators for the specific flowsheet, here shown for aluminium recycling (Reuter et al. 2015).

Fe and steel production and compared the effects of different fuel sources on GWP, CED, human health and RC.

Studies of metal and other material recoveries through recycling typically adopt a gate-to-gate approach to assess and compare the environmental burdens of, inter alia, secondary production options, specific EoL products or waste management technologies. Examples of such studies include the assessment of impacts for the recovery of multiple base and precious metals from e-waste (Van Schaik and Reuter 2010; Bigum et al. 2012; Stamp et al. 2013; Reuter et al. 2015; Van Eygen et al. 2016), the recycling of passenger vehicles (Amini et al. 2007; Reuter and van Schaik 2015), lithium ion batteries (Dewulf et al. 2010), and solar energy technologies (e.g. Fthenakis 2004; Gong and Wall 2014). Some of these studies also incorporate exergy analysis, either separately (e.g. Amini et al. 2007; Abadías Llamas et al. 2019b) or as part of the LCA itself (e.g. Dewulf et al. 2010).

Reuter et al. (2015) developed an approach for linking HSC Sim with openLCA and GaBi, making it possible to map and export detailed inventory data from the simulator into the LCA software, as depicted in Figure 8.

Using this approach, an attributional LCA was performed for the entire system shown in Figure 2 with the production of 2,000 m^2 of CdTe solar cells as the functional unit. Detailed inventory data were imported into GaBi from the TPS model in HSC Sim in the *ecoSpold1* (ecoinvent 2019) data format after mapping process streams to selected impact categories. The focus was on the impacts of the system as a whole, however, when required to analyse specific aspects of subsystems such as unit processes or specific products, allocation was based on physical/chemical relationships – based on mass, exergy content and exergy cost – in line with the ISO standard and made relatively easy by the fact that it was possible to capture these non-linear relationships in significant detail in the TPS model. Allocation based on economic value was not considered for this paper. Impacts were assessed in terms of GWP and AP.

Modelling and simulation of economic and social impact

In this Section, the analysis and assessment of economic and social impacts – the red and purple layers in Figure 3 and the two right-hand circles in Figure 4 – are discussed.

Economics

The economic performance and impacts of systems are assessed in various contexts and for different purposes. At the process level, *techno-economic* assessments are most common and evaluate the potential viability of projects or products using indicators such as capital and operational expenditure (CAPEX and OPEX), net present value (NPV) and internal rate of return (IRR) to name but a few. These assessments consider environmental impacts explicitly to a limited extent. Techno-economic assessment is standard practice for any engineering design office conducting bankable feasibility studies.

Environmental-economic assessments include, among others, life cycle costing (LCC) and environmental life cycle costing (eLCC). LCC is the economic counterpart of LCA and addresses the economic pillar of sustainability. Financial LCC (closely related to Total Cost of Ownership) answers questions from one stakeholder's perspective (Norris 2001), while eLCC considers the costs of all activities linked to a life cycle, regardless of which actor bears the cost (Moreau and Weidema 2015). It enhances financial LCC by including less tangible costs and benefits of pollution controls (Klöpffer 2008). In LCC, it is important to use the same functional unit and equivalent system boundaries when conducted in combination with LCA (Ciroth and Franze 2009, Ekener-Petersen and Finnveden 2013; Klöpffer 2003, 2008; Swarr et al. 2011) to ensure compatibility and consistency.

Thermoeconomics provides a link between exergy analysis and economic assessment. It links component prices to operating parameters and exergetic efficiencies, pricing the specific exergy content of a stream rather than the unit mass. Due to non-linearities and multi-component in- and outputs, proper allocation is required (Sciubba and Wall 2007), as is the case with LCA. Valero et al. (2006) mention that with system design or to maximise system benefits, thermoeconomic cost (which includes capital) should be used, while exergy cost (which excludes capital) would be more appropriate when the focus is on inefficiencies.

Although not conducted within the scope of this paper, the use of a common simulation platform such as that shown in Figure 4 simplifies economic assessment problems significantly. Process simulation results combined with cost estimate data serve as inputs for techno-economic evaluations. Further combination with the LCA structure and results provides the inputs for LCC and eLCC, and the addition of exergy analysis facilitates thermoeconomic analysis. These process level economic assessments can, therefore, all be done consistently with the same physical data foundation.

Social impact

Social LCA (S-LCA) is a method that assesses the social and socio-economic impacts of a product's complete life cycle (Benoît and Mazijn 2009). It follows the standard LCA framework, also specifying goal and scope definition, LCI, LCIA and interpretation as the four phases comprising an assessment. Relevant stakeholder categories include, for example, workers, local community, society, consumers, and value chain actors. Impact categories include, for example, human rights, working conditions, health and safety, cultural heritage, governance, and socio-economic repercussions (Benoît and Mazijn 2009). Attributional S-LCA collects static social performance data from all organisations in the life cycle, which can be used to generate some aggregated score to track social performance over time (Macombe 2019).

As systematic social impact assessment has only recently started to gain more traction, it is still subject to several teething problems. Weidema (2018) argues that the practical application of social footprinting is limited by excessive data requirements, limited understanding of the cause–effect relationships, and insufficient focus on the materiality of impacts. Petti et al. (2018) argue that more clarity is needed around the methodology itself e.g. specifying specific tools for the collection and processing of qualitative data, and improved capture of positive impacts. For these reasons, few relevant case studies have been published to date. Nonetheless, a few examples include a social hotspot analysis for laptop computers (Ekener-Petersen and Finnveden 2013), the evaluation of mobile phone life cycle options (Suckling and Lee 2017; Wilhelm et al. 2015), and the benefits and impacts of a solar PV installation (Yu and Halog 2015). Social impact assessment is beyond the scope of this paper and will be the subject of future work.

Resource efficiency of a large-scale CE system

This section will show various results to demonstrate the state-of-the-art that goes beyond 'simpler' sustainability impact assessment. This is achieved by combining all the tools linked to the simulation platform, as shown in Figure 4, to analyse the system shown in Figure 2. The rigour of simulation makes it possible to scale between the different levels shown in Figure 3, as it is always possible to use all the tools that are linked to the simulation platform. It should be noted that, in order to manufacture 2,000 m^2 of CdTe solar cells, the system has to also produce 108 t Cu, 71 t Zn, 37 t Pb, 290 kg Cd (for other applications), 640 kg Ag, 0.9 kg Au, 570 kg Co and other by-products like sulphuric acid. For this reason, the results reported in this section represent the entire integrated system and all of its products, prior to allocation of consumptions and impacts to specific primary and by/co-products.

Resource use, consumption and efficiency

Conservation law (mass and energy) analysis

For illustration, Sankey diagrams showing the closed mass balances for Cd and Te are shown in Figures 9 and 10, respectively, extracted from the complete flowsheet shown in Figures 6 and 7. Note that, while Cd and Te are shown separately in Figures 9 and 10, they are recycled as CdTe in the PV filter cake stream until they reach the Pb processing system, thus following a product/mineral-centric approach mentioned earlier.

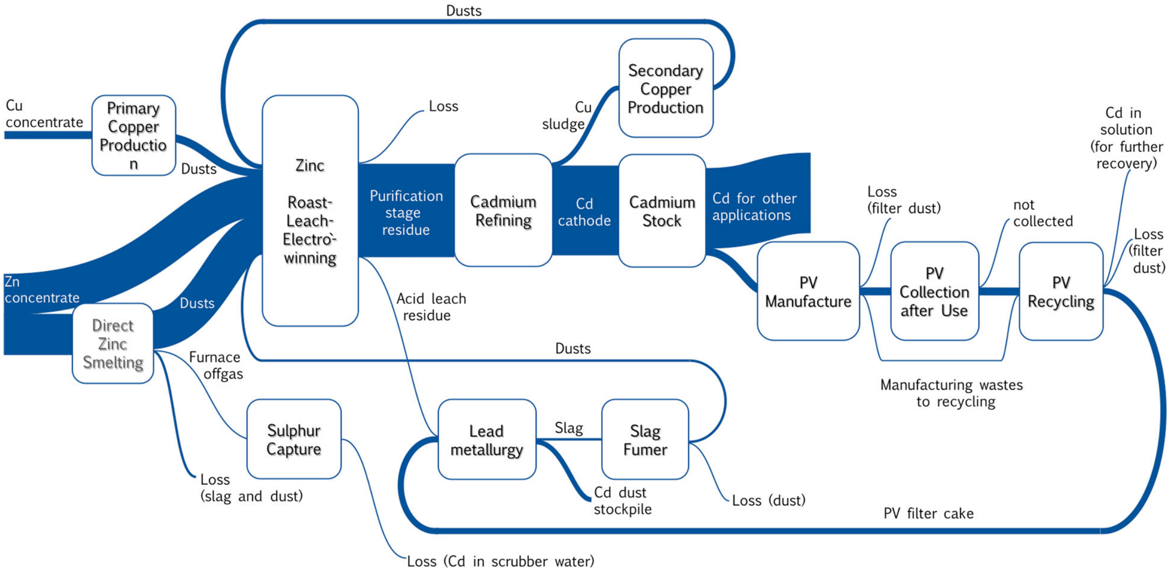

Figure 9. Mass flows of Cd through the integrated metallurgical production system, extracted from the flowsheets in Figures 6 and 7. While only a small portion of produced Cd goes to CdTe PV production, the whole infrastructure, which also produces a multitude of other linked co-product elements, is required.

In the Zn roast-leach-electrowinning (RLE) process, the leaching step is central to Cd recovery because Cd in dusts from Cu, Zn and Pb processing are treated there before being recovered. Most of the Cd losses from the system occur in the Direct Zn Smelting (DZS) process slag, PV manufacturing and recycling dusts, and scrubber water from the sulphuric acid plant. During recycling in the Pb circuit, most Cd reports to dust and is collected and stockpiled i.e. not released to the environment. Minor quantities are recycled to Zn RLE via the slag fuming furnace. Of the initial Te input in Cu concentrate, 3.4% is lost, primarily via dusts collected during PV manufacturing and recycling. Te entrained in intermediate products like slag from precious metals recovery, Cu-Te residue treatment, spent electrolytes, other process solutions and antimony-containing residues can be recovered and are not considered to be losses for the purposes of this study.

The mass balances clearly show the locations and relative quantities of metals within the system, where they leave and in what form, highlighting areas to focus on for the minimisation of losses and improvement of recoveries. Also evident is the contribution of connecting the individual metallurgical infrastructures towards closing material loops. Integration minimises the quantities of untreated residues and losses. In the case of Te, approximately 40% of refined metal originates from secondary production via the Pb circuit. The absence of Pb processing infrastructure would, therefore, be highly detrimental to the RE of the system. These analyses allow for the determination of, amongst others, mass-based RE in terms of metal recovery. Figure 11 shows the recoveries of selected base and minor metals, here defined as the recovery of elements in pure form unless indicated otherwise. More detailed descriptions of the Cu and Zn production system simulations and their associated

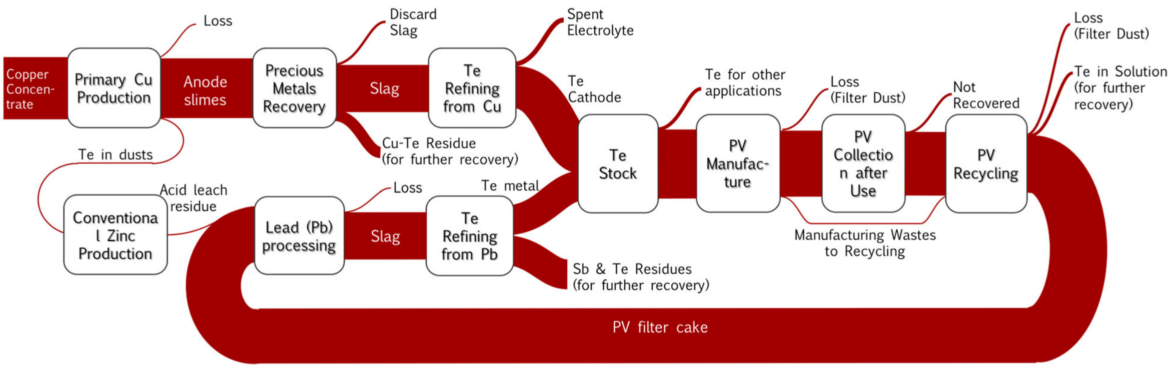

Figure 10. Mass flows of Te through the integrated metallurgical production system, extracted from the flowsheets in Figures 6 and 7. As mentioned for Cd in Figure 9, all the infrastructure shown is required to produce Te.

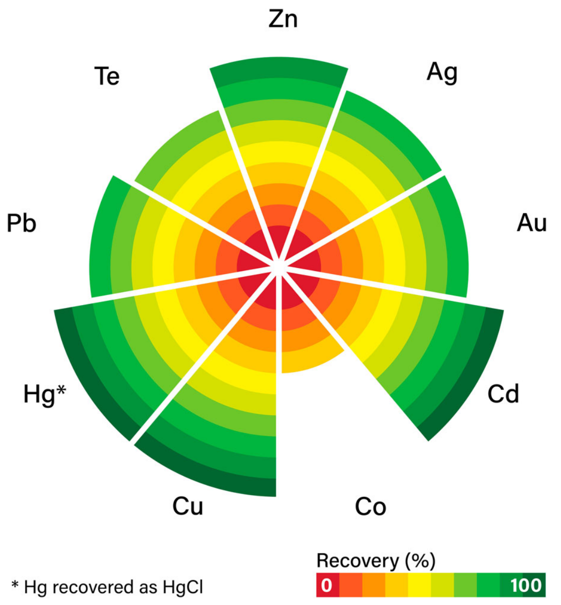

Figure 11. Metal recovery in the integrated metallurgical processing system, here representing a selection of the 30 elements modelled.

residues, losses and metal recoveries can be found in Abadías Llamas et al. (2019b, 2019a).

Second law (entropy/exergy) analysis

As discussed earlier, exergy analysis allows for consumption – rather than use – to be determined. To conduct exergy analysis, the inputs and outputs – material and energetic – to and from a system are classified as fuels[2], products, losses and irreversibilities. Losses include, for instance, metals entrained in slags or sulphur emitted to the atmosphere. A Sankey diagram of total exergy flows through the integrated metallurgical production system is shown in Figure 12. It shows the absolute amounts of exergy lost and dissipated in each of the subsystems for all of the primary and by/co-products that need to be produced in order to manufacture 2,000 m^2 of CdTe PV cells.

Important to note is the contribution of the added slag fuming furnace (bottom left) to the circularity of the system via slag valorisation. Conventional Pb and Cu production subsystems can produce slags with e.g. Pb or Zn contents high enough to prevent them from being suitable for other uses, requiring them to be landfilled as hazardous materials. The fuming process could be used to treat residues, recover the metals, and produce a clean slag suitable for use in construction applications. However, it would consume resources, and create waste streams and emissions itself. The net effect of being able to classify the Pb and Cu slags as by-products that will be processed further – rather than waste streams – however, would be an overall reduction in RC and impacts. This example demonstrates the benefit of digitalising the entire system – it facilitates evaluation of the systemic effects of various process configurations, which can then be used to optimise the system.

As two more detailed examples, the subsystems for primary Cu production and CdTe PV manufacturing are shown in Figures 13 and 14, respectively.

With primary Cu production, the loss of input exergy through waste streams and irreversible destruction results in an exergy efficiency of 60%. If all waste streams could be eliminated – the popular definition of '*closing the loop*' – the maximum achievable RE would be 72% (not 100%), which represents the true technical RE limit for this subsystem *in its current configuration*. It is the unavoidable irreversible losses that reduce the maximum possible efficiency from 100% to 72%. For the CdTe PV manufacturing subsystem, the actual and maximum achievable exergy efficiencies are practically the same at 59% due to the relatively small exergy loss via waste streams. This indicates that the subsystem, as configured within the current model, is operating close to its technical RE limit. It is important to note that the RE limits mentioned here are not set in stone – they could be improved by optimising process configurations and operating parameters.

The actual and maximum REs for other subsystems in their current configurations are shown in Figure 15. The overall RE equates to 57%. The lower exergy efficiency for conventional Zn production is because of the large amount of electricity consumed by the electrowinning process. The difference between the actual and maximum efficiencies represents the maximum theoretical potential for RE to be improved through the elimination of losses or using *all* wastes as resources. The total elimination of losses is not possible, of course, but the theoretical value gives an upper constraint, which is useful in optimisation problems. Again, it should be noted that the maximum values shown in Figure 15 are valid only for the model and process configurations in their current states, and could be changed through system optimisation and/or the use of more efficient technologies.

Whereas exergy *efficiency* gives snapshot values of the RE, exergy *cost* is the cumulative destruction of exergy along the value chain of a commodity or product and can therefore be used as a proxy for RC. For the production of refined Cd and Te, exergy costs were determined to be approximately 1.7 MWh/t Cd and 1.4 MWh/t Te, respectively. Figure 16 shows the contribution of relevant subsystems to these total Cd and Te production exergy costs.

[2]In exergy analysis, *fuels* do not only refer to conventional fuels, but to all sources of exergy entering the system—as mass and energy.

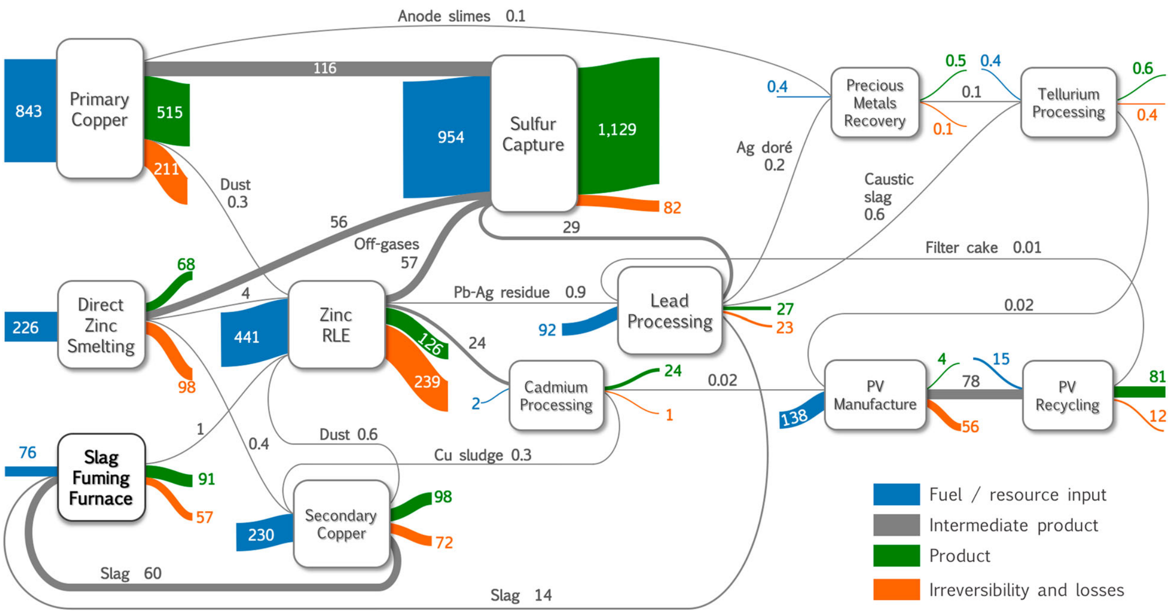

Figure 12. Sankey diagram of exergy flows (in megawatt) for the integrated metallurgical production system, extracted from the results of the complete flowsheet representing all subprocesses.

Figure 16 facilitates quick identification of the best- and worst-performing parts of the system in terms of RC. Zn electrowinning is the largest contributor to the exergy cost of Cd production because it supplies the spent electrolyte used as the leaching agent during Cd purification. Zn leaching is the second-largest contributor as all process streams containing Cd for recovery – including dusts from primary and secondary Cu production, slag fuming and DZS – are treated there. As mentioned, primary and secondary Te are produced via the Cu (60%) and Pb (40%) production systems, respectively. The Te electrowinning process is responsible for half of the exergy cost, followed by precious metals recovery, and recovery from Pb slag. Contributions from primary Cu processing are mainly due to Cu electrorefining. Utilising the exergy costs above, total RC – in terms of exergy destruction – is 1.55 kWh/m^2 CdTe PV cell manufactured. As rightly stated by Brunner and Rechberger (2017), energy streams can dominate in exergy analyses combining material and energy flows. While the type of energy resource alone would not change gate-to-gate efficiencies and costs such as those shown in Figures 15 and 16, cradle-to-gate and cradle-to-cradle perspectives would capture the exergy destruction history behind those resources, revealing improvement focus areas, especially in the context of energy grid mixes and the transition to renewable resources. In combination with environmental impact assessment, this becomes even clearer as will be shown below.

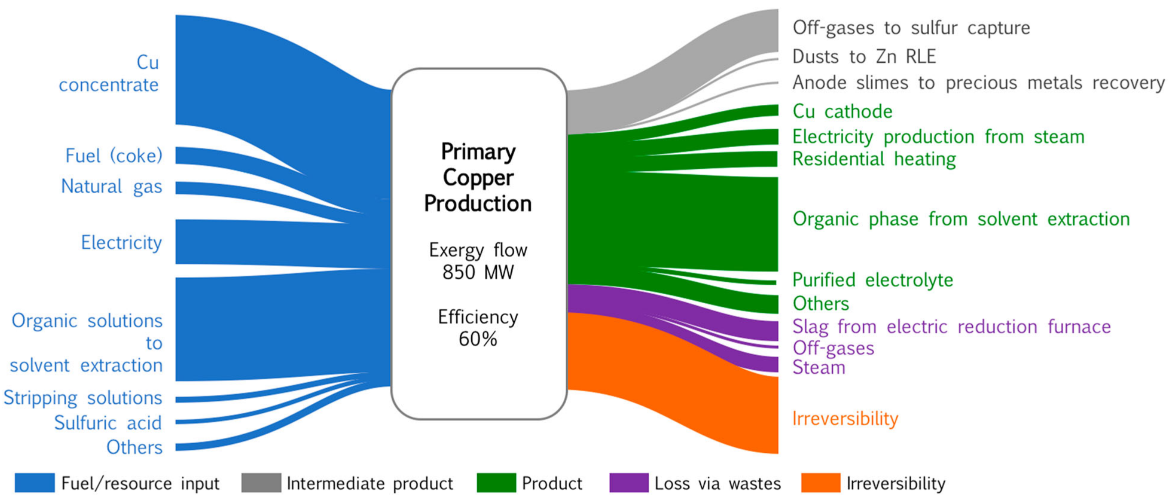

Figure 13. Exergy flows for the primary Cu production subsystem, showing a rather low exergetic efficiency of 60%.

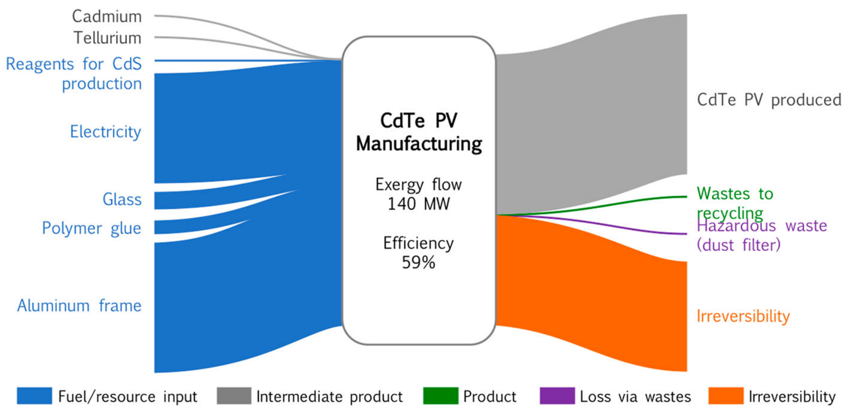

Figure 14. Exergy flows for the CdTe PV manufacturing subsystem, also showing a low exergetic efficiency of 59%.

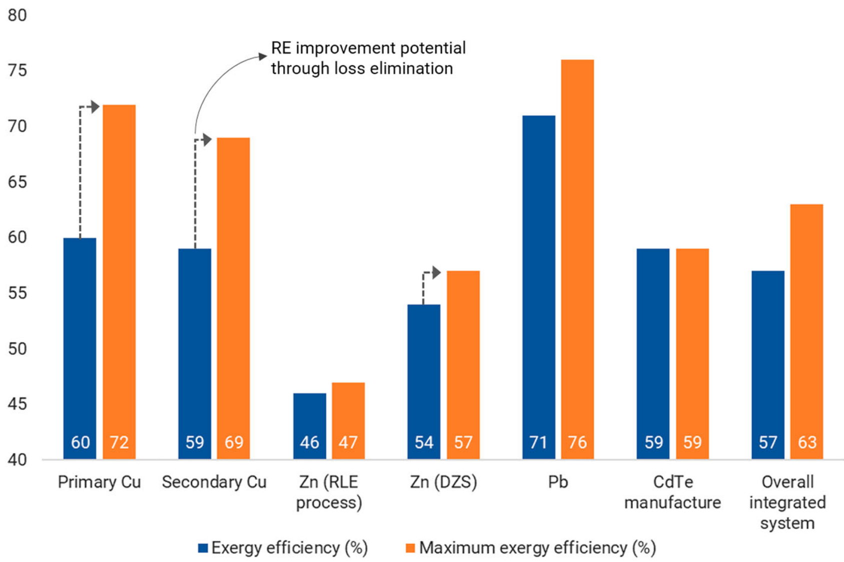

Figure 15. Exergy efficiency and technical RE improvement potential for the system and its components depicted by Figure 2, reflecting a low overall efficiency but also showing the relatively high efficiency of lead production.

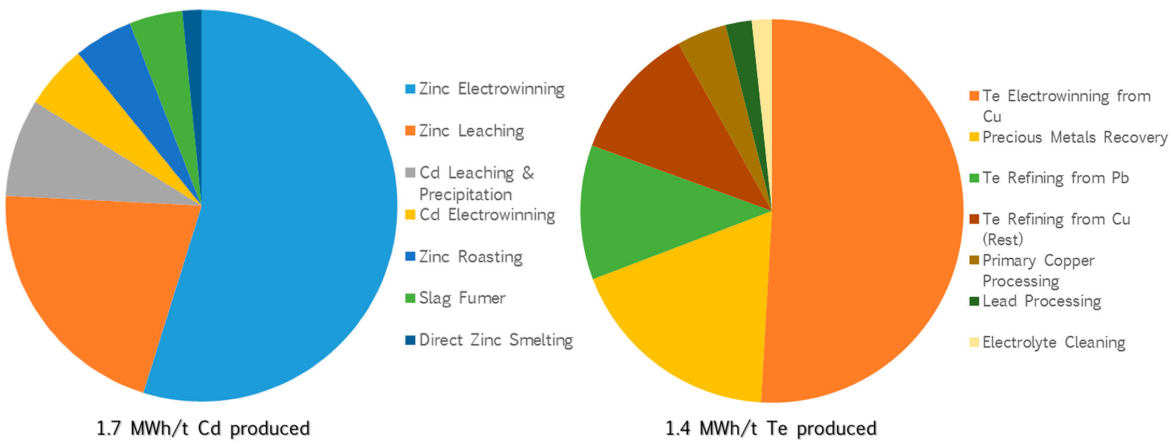

Figure 16. The contribution of individual subsystems to the overall exergy cost for Cd (left) and Te (right) production.

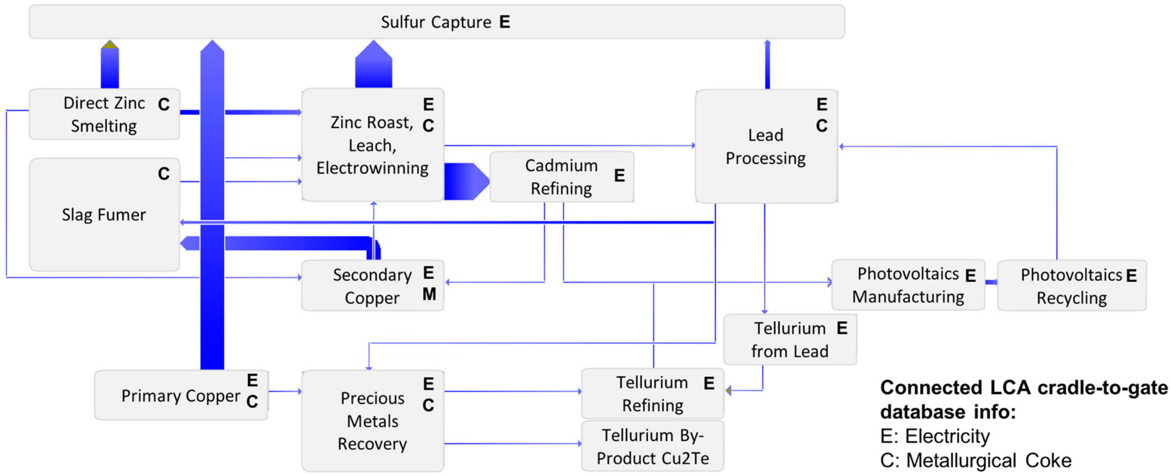

Figure 17. LCA model in GaBi for the system depicted in Figure 2 used to determine the environmental footprints of the system as depicted by Figures 18 and 19.

Environmental impact of the integrated metallurgical production system

As mentioned earlier, an *a*LCA was performed for the entire system shown in Figure 2, and the results shown here represent the total environmental impacts of the entire integrated system and all its products, prior to allocation of impacts to specific primary and by/co-products. The production of the metallurgical infrastructure (capital goods), manufacture of complete installed CdTe PV systems, the associated balance-of-system (BOS) components, and the *Use* phase of the life cycle were excluded from the environmental impact assessment in this study. The equivalent system in GaBi is shown in Figure 17.

Figure 18 shows the LCA results for system-wide GWP, disaggregated to show the relative contributions of individual production subsystems. As mentioned earlier, the World Steel Association (worldsteel) estimates that on average, 1.83 tonnes of CO_2 were emitted per tonne of steel produced in 2017 (worldsteel 2019).

Pyrometallurgical processes like DZS, primary Cu production and the Zn fuming furnace contribute to climate change because of the considerable amounts of metallurgical coke combusted as fuel in these processes. The off-gases from many subsystems are collected in the sulphur capture plant and therefore, its apparent greenhouse gas (GHG) emissions actually derive from other subsystems. The generation of electricity required to run the integrated system is, however, the largest contributor to climate change. Therefore, the total impact depends strongly on the electricity grid mix at the location the electricity is generated. To illustrate this, Figure 19 shows scenarios for situating the entire integrated system represented by the simulation model in various locations globally, evaluating the climate change impact based on location-specific energy mixes. Countries with different grid mix compositions (based on 2014 data from the GaBi LCI database) such as India (16% lignite/60% hard coal/10% hydropower) and Norway (96% hydropower), as well as countries like Chile, which actually host most metallurgical infrastructures, were chosen.

The impact of electricity generation is dominant when more than 50% of it is generated from fossil fuels like hard coal, lignite, or natural gas. In countries generating electricity mostly through hydro, nuclear and wind power, emissions from the metallurgical infrastructure itself make the largest contribution to climate change. Figure 19 highlights the importance of the balance between electricity requirements and the infrastructure footprint itself and shows clearly where the largest improvement potential in terms of GWP lies.

LCA results for AP are shown in Figure 20. Sulphur dioxide (SO_2) emissions occur primarily during Pb processing, and through the release of cleaned off-gases from the sulphuric acid plant. Even with highly efficient sulphur removal, the acid plant itself remains a significant contributor to the acidification potential – it treats all captured sulphur-containing off-gases in the system, but can never achieve a 100% conversion rate (cf. Abadías Llamas et al. 2019a). The pyrometallurgical units are the greatest contributors to this impact category due to the substantial amounts of sulphur-containing gases generated during the roasting and smelting of sulphide concentrates as well as the combustion of fossil fuels. To further reduce SO_2 emissions and AP, sulphur fixation technologies with higher efficiencies would be required.

While only GWP and AP are discussed in this paper, the inventory data for other impact categories are implicit in the process model data including, for instance, the use and quality degradation of natural resources such as water.

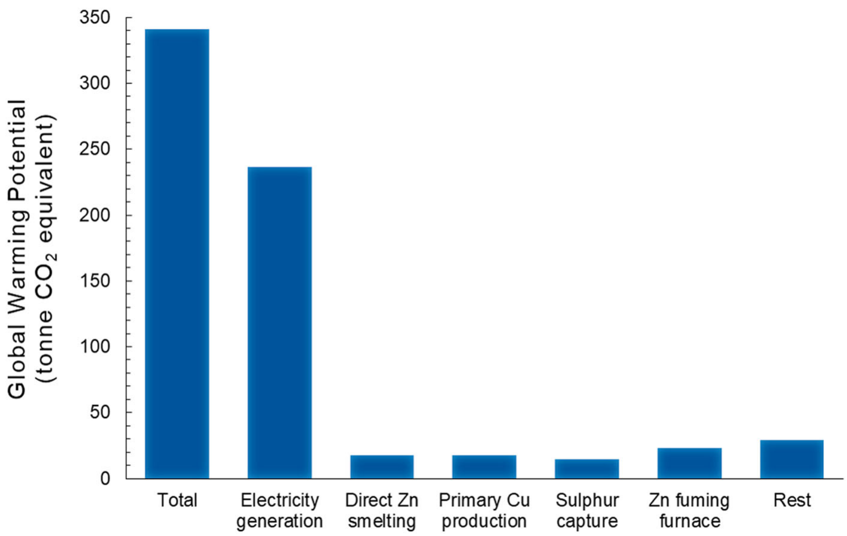

Figure 18. Global warming potential for the integrated production system prior to allocation of impacts to specific primary and by/co-products. Electricity generation is the largest contributor to this impact category.

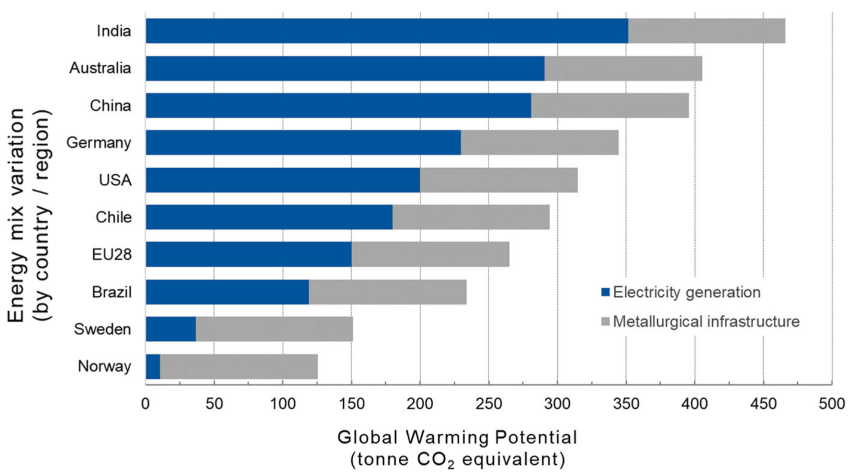

Figure 19. The effect of energy mix variation (by country or region) on the relative contribution of electricity generation to GWP. The electricity production bar in Figure 18 is represented here by the horizontal (blue) bar for Germany, as the German energy mix was used for this study.

Discussion

This paper shows how to integrate analyses of energy, exergy, material flows and environmental impacts at a high level of detail. It demonstrates the benefits of using a single simulation platform to map all the physical flows in the system and to subsequently evaluate its true RC and RE as well as its environmental footprint based on the same consistent inventory data. The focus of the paper is on thermodynamic irreversibility and metallurgical processing limits and opportunities as well as the application of complex, large flowsheets. It is acknowledged, however, that other factors – economic, social, and environmental – also contribute to irreversibility (Gößling-Reisemann 2008). The following have not been modelled yet:

- Financial life cycle costing (LCC) and environmental life cycle costing (eLCC).
- The market dynamics for the produced metals. For this reason, one-to-one displacement of primary production by metals recovered through recycling processes – thus closed loop recycling without market mediation – is assumed in this paper.
- Potential social benefits and impacts.

While power can dominate absolute exergy efficiency, a different picture emerges when renewable energy infrastructure starts to dominate the energy mix. In the renewable and circular economy paradigm, metals become closed-loop ‘fuels’ as these, on the one hand, create the energy but can also be recycled to an extent. In the linear economy paradigm fuels are

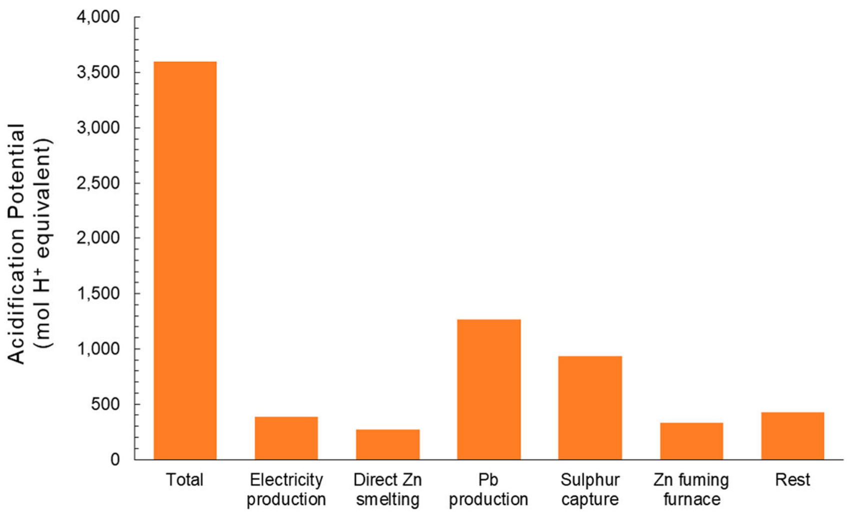

Figure 20. Acidification potential for the integrated production system prior to allocation of impacts to specific primary and by/co-products. Lead processing contributes most to this impact category.

obviously open loop i.e. exergy is dissipated when combusting them in power stations.

The large-scale simulation model represents a system of linked micro-level production processes. In the current model, all the metal production infrastructures are assumed to be in the same location. Individual sections can be located anywhere, however, and the associated supply chains incorporated, providing an even more detailed analysis. This demonstrates the benefits and flexibility of the approach. For instance, the use of a single sulphuric acid plant to treat all sulphur-containing off-gases is not realistic as these gases obviously cannot be transferred around the globe into a single plant. However, sulphuric acid plants or other sulphur fixation facilities can easily be duplicated or added in the simulation platform. At this stage none of the transport supply chains between the subsystems are included and therefore, the associated RCs (exergy costs) and environmental impacts are not accounted for. However, transport can be included in the simulation platform easily as processes that consume fuels and produce off-gases and other residues – transport simply becomes a ‘reactor’ and the transport length/distance the duration of the ‘batch process’.

In addition to addressing the items above that have not yet been modelled, the platform depicted by Figures 3 and 4 is presently being expanded from an integrated metallurgical process simulation and LCA platform to include material databases from the product design side, and hence all material properties that are of interest not only for physical separation processes but also the compositional design detail of any arbitrary part of a consumer product. As mentioned in the Introduction, the utilisation of consistent physical quantity and quality data for RC and RE determination, and across the environmental, economic, and social dimensions on a common digital platform facilitates robust sustainability assessment, in contrast to disjointed analyses with potentially incompatible system boundaries. Furthermore, integrating the tools depicted by Figure 4 and linking the simulation model to multi-criteria constrained optimisation tools permit exploration of the complete simulated model space and finding the constrained optima for the system reflected by Figure 2.

Conclusion

In this paper we demonstrate that it is possible to simultaneously assess the RC, RE and environmental impacts of very large symbiotic metallurgical production and recycling systems for specific products. In other words, a digital twin for the complete CE system is created. This is achieved through the process simulation of all relevant transformation processes at the unit operation level of detail, which then becomes the foundation from which consistent inventory data (i.e. flows of elements, compounds, energy etc.) Is generated and made available for assessments across all dimensions of sustainability.

This foundation is used to conduct exergy analysis to determine the RC and RE. Furthermore, on this simulation basis a LCA is conducted to determine potential environmental impacts (GWP and AP), using the CdTe PV cell life cycle as a contemporary example. The authors’ future work includes the incorporation of economic and social impact assessments. This approach provides the ability to rigorously establish baseline performance, and to predict the potential systemic effects of variations in the life cycles of complex, multi-metal containing products, the aim being to impactfully drive sustainable development and the transformation to a CE.

This work clearly also shows the central role the metallurgical process industry plays at the heart of the CE to help minimise the various dissipative (i.e. material and exergy) losses from the system. Through this the inconvenient truth of not being able to 'close' the loops is quantified. It evolves the CE discussion and also policy development to a fact-, economics- and physics-based discussion.

Disclosure statement

No potential conflict of interest was reported by the authors.

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Annexure B: Article 2

The resources, exergetic and environmental footprint of the silicon photovoltaic circular economy: Assessment and opportunities

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The resources, exergetic and environmental footprint of the silicon photovoltaic circular economy: Assessment and opportunities

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The resources, exergetic and environmental footprint of the silicon photovoltaic circular economy: Assessment and opportunities

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ABSTRACT

The photovoltaic industry has shown vigorous growth over the last decade and will continue on its trajectory to reach terawatt-level deployment by 2022–2023 and an estimated 4.5 TW by 2050. Presently, its elaboration is driven primarily by cost reduction. Growth will, however, be fuelled by the consumption of various resources, bringing with it unavoidable losses and environmental, economic, and societal impacts. Additionally, strong deployment growth will be trailed by waste growth, which needs to be managed, to support Sustainable Development and Circular Economy (CE). A rigorous approach to quantifying the resource efficiency, circularity and sustainability of complex PV life cycles, and exploring opportunities for partially sustaining industry growth through the recovery of high-quality secondary resources is needed.

We create a high-detail digital twin of a Silicon PV life cycle using process simulation. The scalable, predictive simulation model accounts for the system's non-linearities by incorporating the physical and thermochemical principles that govern processes down to the unit operation level. Neural network-based surrogate functions are subsequently used to analyse the system's response to variations in end-of-life and kerf recycling in terms of primary resource and power consumption, PV power generation capacity, and CO_2 emission. Applying the second law of thermodynamics, opportunities for improving the sustainability of unit operations, the larger processes they are the building blocks of, and the system as a whole are pinpointed, and the technical limits of circularity highlighted. We show the significant effects changes in technology can have on the conclusions drawn from such analyses.

1. Introduction

Solar photovoltaics (PV) is one of the electricity generation technologies set to play a key role in the transition to low-carbon energy systems. Over the last decade, the global solar PV industry has grown at a rate of more than 35% annually, reaching record levels and outpacing annual conventional power capacity additions in many regions. At the end of 2019, the world's cumulative installed PV capacity was 583.5 GW with an annual module production capacity of 143 GW (Fraunhofer ISE, 2020). This exponential growth can be largely attributed to dramatic cost reductions (VDMA, 2020), solar technology innovation, and specific support policies aimed at reducing the price gap between PV and conventional electricity sources (IEA PVPS, 2019). The further development and deployment of solar PV can result in new business models that stimulate industrial and employment growth (Michas et al., 2019). Industry forecasts project global installed PV power to reach the terawatt (TW) level by 2022–2023 (Haegel et al., 2019). The COVID-19 pandemic and associated market uncertainty and volatility has, however, exposed vulnerabilities such as the susceptibility of PV supply chains to shocks, and is highly likely to cause delays in several planned PV projects (IEA, 2020a; NREL, 2020). With a current share of 95% and its market dominance set to continue, crystalline Silicon (c-Si) PV is well positioned to increase the annual production of PV modules by 3–4 terawatts (TW) annually by 2040 (VDMA, 2020).

Several other costs, not just monetary but also in terms of resources, waste, and environmental, economic and societal impacts, are associated with industry growth. PV technologies rely on the availability of various materials, particularly Silicon metal in c-Si-based PV

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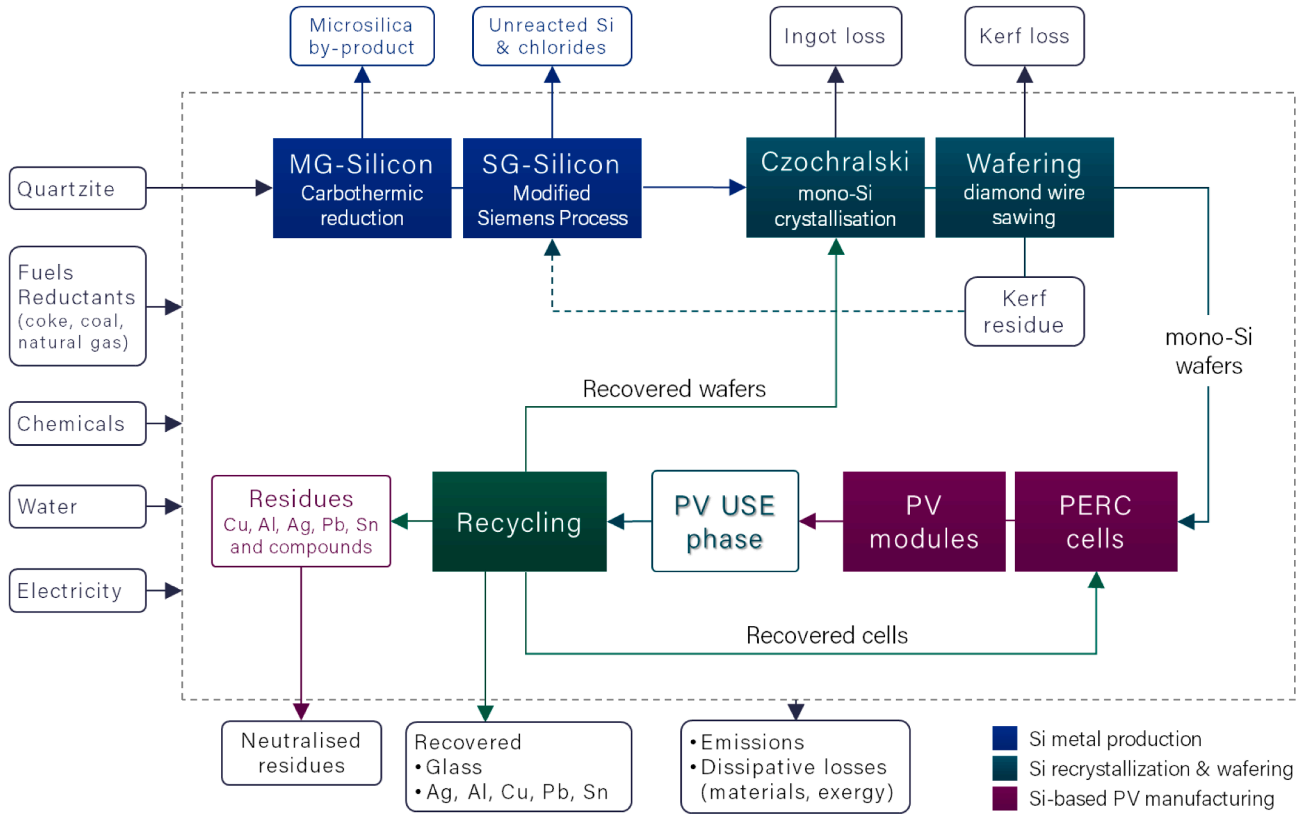

Fig. 1. mono-Si PERC PV life cycle system.

technologies, the high economic importance of which, together with its increased supply risk, justified its inclusion in the European critical raw materials (CRMs) list (European Union (EU), 2017a). Improvements in cell and module efficiencies and efforts to reduce material consumption have resulted in the amount of polysilicon needed per watt of power generated decreasing from 7.2 g/W in 2009 to 3.6 g/W in 2019 (Fraunhofer ISE, 2020; IEA PVPS, 2019). While this trend will cause a proportional decrease in Si demand, PV deployment will still cause a net increase—demand for the EU PV sector is expected to increase from 33 kt in 2015 to 235 kt in 2030 (European Commission, 2018). Furthermore, end-of-life (EOL) PV module quantities are expected to increase significantly between 2020 and 2030 because of modules commissioned over the last few decades now beginning to come out of service—the global waste-to-new installation ratio is expected to increase from 4 to 14% in 2030 to over 80% by 2050 (Sica et al., 2018). With cumulative global PV waste quantities forecast to reach 8 million tonnes by 2030 and ten times as much by 2050 (Heath et al., 2020), the need for increased focus on design-for-X (DfX, where X refers to e.g. circularity, sustainability, disassembly, or recycling), and for actions that foster the development of circular business models and sophisticated recycling processes capable of recovering high-purity Si and other materials, is clear.

It should be noted that projections of waste quantities are based on the assumption that PV modules reach EOL once their power generation efficiency has deteriorated to about 80% of the nameplate efficiency. This technical lifetime is typically 25 to 30 years; it does not mean that modules are no longer useful after this period. However, several factors come into play considering the trade-offs between recycling and re-use after the first lifetime. For utility-scale installations, Wade et al. (2017) found that high-value recycling provides an economic incentive as the revenue from recycled materials would exceed decommissioning costs. Furthermore, the probability of the levelized cost of electricity (LCOE) from a re-use system being lower than that from a new system at the same location is less than 10% (Wade et al., 2017). Aside from purely economic considerations, the export of modules for re-use to e.g. first world or developing countries risks, inter alia, the informal recycling of second-life EOL modules at standards below those prescribed by European health and safety regulations, potentially creating negative environmental and social impacts in communities already at a disadvantage relative to European living standards. On the other hand, such exports could provide these communities with access to electricity for the first time. *Re*-use also lowers environmental impact by increasing life cycle power generation without the additional materials and energy consumption associated with manufacturing (Heath et al., 2020). As stated by Tsanakas et al. (2020), research to date have been somewhat biased towards recycling, leaving many of these trade-offs as yet unexplored. Ongoing projects like CIRCUSOL (circusol.eu) aim to formalize PV industry value chains for repair, refurbishment and re-use, and to develop more circular PV business models (Tsanakas et al., 2020).

While recycling aims to close material loops to maximise resource efficiency (RE), the seldom-discussed quandary is that no such loop can be closed entirely—any real transformation process is always subject to material and energy losses because of inevitable inefficiencies and the creation of entropy. Therefore, the identification and minimisation of these along entire life cycles, not just at EOL, are key (Reuter et al., 2019). If not accounted for, losses cannot be 'designed out' because the need for innovation would not be identified in the first place. All of these effects, as well as environmental, economic and social impacts need to be quantified in the conceptual and early design phases, so that efforts typically focussed on PV-specific cost reductions and power conversion efficiency (PCE) improvements can be evaluated within the bigger picture of entire product life cycles, sustainability and CE. Without such comprehensive assessments, the true contribution of the PV industry to decarbonisation and achieving sustainable CE would be difficult to ascertain.

This paper provides insights on the resource and sustainability performance of the mono-Si PV module life cycle using the PERC cell architecture. We apply the methodology presented in a previous

Table 1
Cases mapped on the parameter space.

	Base case	Case 1	Case 2
Primary mineral resource			
Quartzite	Fixed*	Fixed*	Fixed*
Secondary resources			
EOL modules	Not recycled	95% recycled (as SG-Si)	95% recycled (as SG-Si)
Kerf residues	Not recycled	Not recycled	50% recycled (as MG-Si)

* Fixed at various levels, as specified in the text and in Figures.

publication, in which the resource consumption (RC), RE, and environmental impacts of the cadmium telluride (CdTe) PV module life cycle were assessed (Bartie et al., 2020), to create a similar digital twin for the c-Si PV life cycle. Detailed, highly disaggregated digital twins enable the quantification of resource requirements as well as potential sustainability impacts using up-to-date equipment and operational information, for both current technologies and those under development. In doing so, there is the potential to facilitate DfX by complementing design activities with resource and sustainability information. The simulation-based approach takes into consideration the many non-linear physical, chemical, and thermodynamic transformations that govern production and recycling processes, as opposed to somewhat over-simplifying approaches that sometimes apply outdated process data to current and developing technologies. Comprehensive inventory databases, albeit extremely useful sources of information, do not yet include data for e.g. mono-Si PERC cells and other newer technologies; a significant proportion of source data for Si-based modules date back to between 2011 and 2015 (e.g. Frischknecht et al., 2020). The application of older data to newer technologies, and the use of linear, mass-based material flow analyses, could lead to inaccuracies as well as the exclusion of thermodynamic processes and non-linearities from assessments, especially where recycling loops are present. For this paper, we exclude the use phase as it has virtually no emissions (Muteri et al., 2020), and the impacts of material transfers between life cycle steps as it is assumed that all value chain stakeholders are co-located. The focus is on the effects of recycling on the resource efficiency and carbon footprint of the system. The scenarios investigated, and the methods used are described in Section 2, and results are presented and discussed in Section 3. The paper is concluded and future work briefly discussed in Sections 4 and 5, respectively.

2. Methods

2.1. System and scenario definition

The life cycle system analysed in this paper is shown in Fig. 1 and consists of the following processes:

1 Metallurgical grade silicon (MG-Si) production through carbothermic reduction of silica (SiO_2) with ladle refining,
2 Solar grade silicon (SG-Si) production using the Siemens process,
3 Monocrystalline silicon (mono-Si) ingot crystallisation via the Czochralski (Cz) process, wafer cutting using diamond wire sawing (hereafter also referred to as *wafering*), and the recycling of kerf residue,
4 Production of PV cells of the PERC design,
5 PV module assembly, and
6 EOL recycling, consisting of thermal delamination and polymer combustion, followed by leaching/etching processes that aim to recover valuable and hazardous metals/compounds.

We cover the parameter space that includes the full ranges of EOL and kerf (a residue that forms during wafering) recycling rates, and map the cases shown in Table 1 onto this space. The base case, a linear production scenario in which no recycling takes place, serves as a reference. In Case 1, 95% of the produced PV modules are collected and all enter the EOL recycling process, while all kerf residue is lost. Case 2 builds on Case 1 by additionally recycling of 50% of the kerf residue. The Si recovered from EOL modules is recycled to the Cz process at SG-Si grade, while kerf residue is recycled to the Siemens process at MG-Si quality. The recycling of kerf residue at SG-Si quality is briefly explored (not shown in Fig. 1).

For comparison, the solution space for a scenario that represents a complete process change—replacing the Siemens process with a silane (SiH_4) fluidised bed reactor (FBR), and changing from mono-Si to multicrystalline silicon (mc-Si)—is presented in Section 3.4.

The methods employed are described in Section 2.2. Descriptions of the production and recycling processes and assumptions can be found in the Appendix.

2.2. Methods used

2.2.1. Process simulation

Understanding the mass and energy flows through production systems is essential for their design, simulation and optimisation, and to manage their complexity and interconnectedness (Fröhling et al., 2013; Klatt and Marquardt, 2009; Reuter, 1998, 2016). The laws of conservation dictate that mass and energy must balance over every piece of equipment, process chain and system, and the second law of thermodynamics (SLT) states that any real process can only occur in the direction of increasing entropy. At its core, process simulation (PS) is based on these principles—incorporating large databases of the physical, chemical and thermodynamic properties of tens of thousands of metals, minerals and other compounds, PS platforms enable detailed analysis, complying with the laws of conservation and the SLT at every step. Therefore, these constraints are implicit and ensure that the laws of physics are not violated (Diwekar and Small, 2002). Instead of assuming linearity over large process blocks, processes are disaggregated into their constituent unit operations and relevant thermochemical and physical transformation processes used to determine input-output relationships over each unit. The distribution of valuable and hazardous substances are calculated and predicted where they occur, making it possible to allocate emissions to the correct outputs, to identify consumption and pollution hotspots, to maximise recovery of materials and energy, and to minimise entropy creation. This is the minimum level of detail required to characterise process and recyclate flows, the extent of downcycling, and processes' contribution to sustainable development and CE (UNEP, 2013). Representative simulations rely on high-quality input data and in-depth knowledge of metallurgical and other processing options and their limits (Verhoef et al., 2004; Reuter, 1998, 2016), which requires industry buy-in and collaboration.

Applying the above principles, the HSC Chemistry platform, HSC Sim (Outotec, 2020) is used to create a high-resolution digital twin of the mono-Si PV life cycle system to assess its resource and environmental performance. For each process block shown in Fig. 1, the constituent unit operations are separately modelled and connected to create a simulation for that process. The processes are then connected to create a closed-loop industrial symbiosis system that represents the life cycle. The effects of solution chemistry and entropy creation are accounted for by means of Gibbs free energy minimisation in HSC Chemistry and FactSage (version 7.2) (GTT-Technologies, 2020), as well as phase diagrams, Pourbaix (E_h-pH) diagrams for aqueous solutions, and Ellingham diagrams to estimate product compositions and process requirements. The result is a deterministic simulation model of the entire life cycle, in this case comprising 75 unit operations, 334 streams and 163 species. It is parameterised by various physical relationships, chemical reactions, thermodynamics, and constants, and validated against known operating points and industrial reality. Using such a model, blanket assumptions of linearity for complex processes and systems are largely avoided. The simulation is used to predict the system's response to changes in the input variables—in this case EOL and kerf recycling rates at constant quartzite consumption—over the ranges defined in Table 1.

2.2.2. Exergy analysis

As mentioned, the SLT states that entropy must increase for any real-world process to take place. Exergy, which represents the *quality* of an energy quantity, is dissipated as entropy increases. Therefore, all real transformation processes occur with an unavoidable level of inefficiency. Contrary to mass and energy, therefore, exergy is not conserved and does not balance over real processes—the exergy dissipated i.e. the *imbalance* is referred to as the irreversibility of the process at hand. Dissipated exergy can only be restored through input from outside of the system (Dincer and Cengel, 2001), which, in turn, cannot happen without exergy dissipation in another system. In other words, we are caught in a spiral of ever-decreasing exergy as the system gradually approaches equilibrium with its surroundings. Complete equilibrium is not a desired outcome if products are to be kept in circulation for as long as possible—the dissipation of exergy should be counteracted or prevented from happening in the first place. Manifestations of the law can be observed, for example, when metals or other compounds are intentionally or unintentionally combined or dissolved into one another, at the outset likely to endow products with specific functionalities, but ultimately to the detriment of EOL treatment efficiency and recyclate purity. As compounds are exposed to contaminants during recycling, entropy increases further, again causing decreases in quality (Amini et al., 2007). This downcycling can only be countered by valorisation processes that themselves dissipate exergy. The material and energy resources required for these can only come into being if exergy is dissipated elsewhere. This highlights the critical importance of DfX in the early stages of sustainable production and supply chain design.

Exergy analysis provides a useful set of tools with which to keep track of the locations and magnitudes of these degradations, for material and energy simultaneously, highlighting opportunities for improvement. We use the *exergy efficiency* of processes and systems, and the *exergy cost* (Lozano and Valero, 1993) for intermediate and finished products as proxies for RE and RC, respectively. More detailed explanations of calculation methods can be found in previous publications (Bartie et al., 2020; Abadías Llamas et al., 2019).

2.2.3. Neural networks

For complex, non-linear life cycle simulations such as that presented in this paper, computational time and intensity can become problematic. Neural networks (NN) can be used to model input-output interrelationships in highly complex systems (Casalino et al., 2016), allowing for generalized non-linear process modelling without the need to predefine regression equations (Reuter et al., 1992). NNs are, therefore, useful tools with which to create surrogate functions for the input-output relationships of interest while considering the entire system's response. Aiming to emulate how neurons in the brain fire to transmit information, NNs consist of layers of neurons. Each neuron is a computational unit that transforms a weighted sum of its inputs into an output using an activation function, for which the non-linear sigmoid function is typically used (Kubat, 2017). The weights are the NN's degrees-of-freedom, the number of which depends on the number of neurons used. The NN learns via a training function that adjusts the weight of each input into each node iteratively until the overall input-output error is minimised. We implement a basic NN architecture, a multilayer perceptron, to emulate process simulation results.

The first step in creating a NN is to generate the dataset needed to train, validate, and test it. To this end, HSC's scenario editor is used as follows:

- Random combinations of the three independent variables (quartzite consumption, and EOL and kerf recycling rates) are generated by randomly sampling from a continuous uniform distribution over specified data ranges.
- The simulation is run with each set of inputs to calculate the system response. Here, each run requires 21 iterations to converge.
- Thirty-two dependant variables are read from the simulation into the dataset and a further 40 calculated.
- For the results presented in this paper, the exercise was repeated 3070 times i.e. performing 63,170 simulation iterations.

MATLAB's (MathWorks, 2020) NN user interface (*nftool*) is used to generate the code that initiates, trains, tests and validates the networks. The dataset is imported and randomly divided into three subsets such that 70% of the 3070 data combinations is used for network training, 15% for testing, and 15% for validation. During network training, the validation error decreases but could increase again if over-training occurs, the equivalent of the NN 'memorising' the dataset instead of learning to generalise. Using the MATLAB tool, network training stops once a validation error increase is detected over six consecutive iterations (MathWorks, 2020c). We use small, shallow perceptrons comprising three neuron layers to minimize the number of weights without restricting the NN's ability to learn the input-output relationships effectively (Reuter et al., 1992). A schematic representation of such a network and its relation to the process simulation is shown in Fig. A1.1.

A separate single-output NN (such as that shown in Fig. A1.1) is created for each dependant variable of interest. For the results presented in this paper, these include power consumption and CO_2 emissions during production and recycling, and nominal PV power generated. All these NNs have the same inputs i.e. the amount of quartz consumed for Si production, the module EOL recycling rate, and the kerf recycling rate. The number of hidden neurons is kept to a minimum to ensure that the ratio of samples to degrees-of-freedom is high, and is chosen as the smallest number that produces a stable NN i.e. one that produces the same result every time it is called. Stability is evaluated visually by running the NN repeatedly, and by comparing regression coefficients and validation errors obtained for different numbers of hidden neurons. We generally obtain the best results with a hidden layer width of three to four neurons using the *Bayesian regularization backpropagation* training function (*trainbr*), which *"updates the weight and bias values according to Levenberg-Marquardt optimization"* and *"minimizes a combination of squared errors and weights, and then determines the correct combination so as to produce a network that generalizes well"* (MathWorks, 2020b). In this case, redundancies vary between 146 and 192. This approach reduces the risk of overtraining a NN to the quirks of the specific dataset used for its training.

2.2.4. Environmental impact

Carbon footprints are evaluated for the cases in Table 1, focussing on direct (Scope 1) and electricity-related (Scope 2) emissions (Greenhouse Gas Protocol, 2011). To ensure consistency and compliance with the laws of physics, the system boundary is exactly that of the process simulation. In processes with multiple outputs, overall environmental impacts need to be distributed between products sensibly. We largely avoid allocation by disaggregating the life cycle into individual unit processes (Ekvall and Finnveden, 2001), allowing for actual emissions to be quantified where they are generated.

The simulation-based approach expands the foreground system, so avoiding the over-use of aggregated databases to populate background system inventory. It also allows for individual process steps and production routes to be optimised or updated to reflect current technologies and up-to-date operating parameters, and for emissions data to be updated accordingly. Furthermore, it allows for reactors and equipment, production routes and supply chains (potentially regionally distributed) to be configured for the study at hand. The databases remain valuable and essential tools, however.

CO_2-equivalent direct emissions (Scope 1) are obtained from chemical reactions defined for individual process steps in the process simulation. Scope 2 emissions are determined from the power requirements

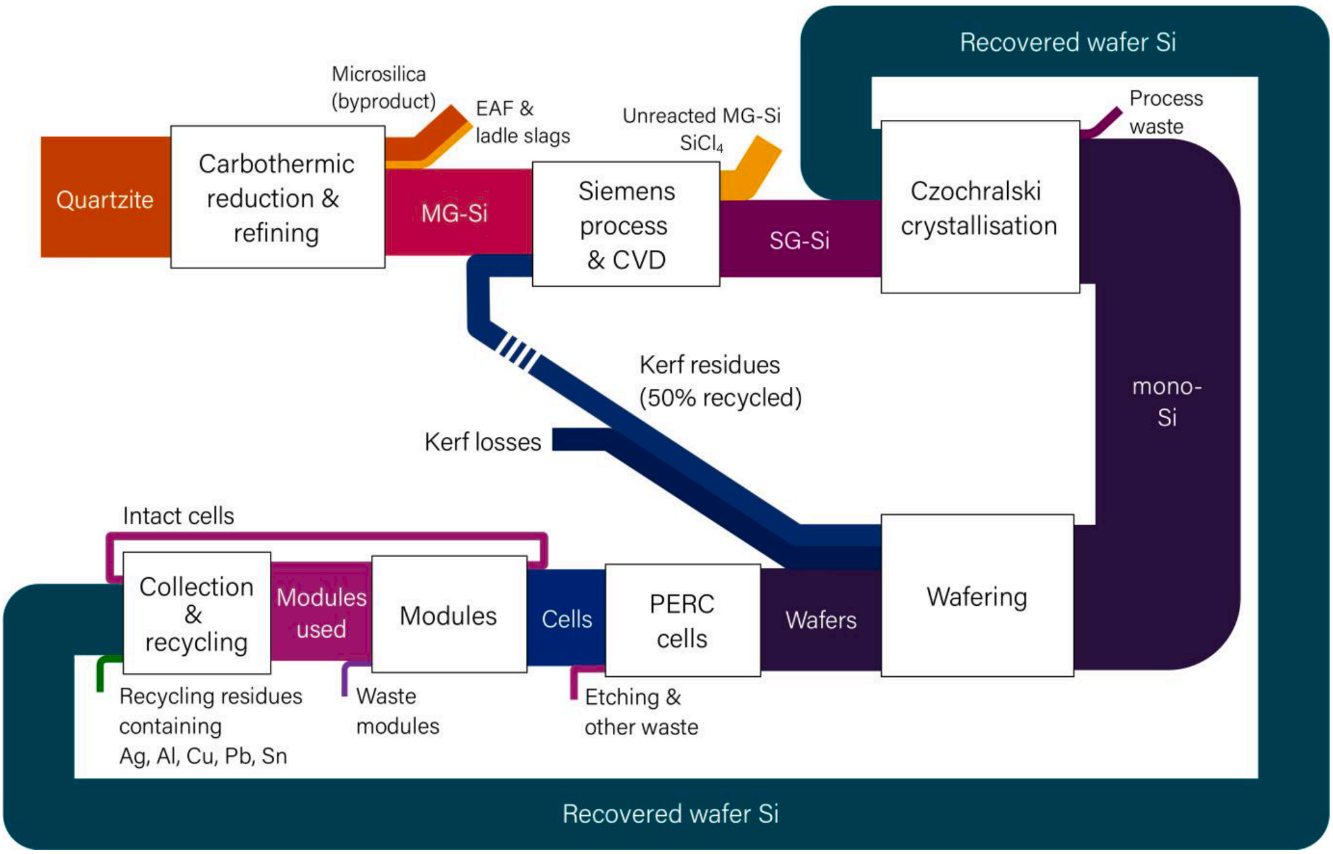

Fig. 2. Silicon balance for the life cycle with 95% EOL recycling and 50% kerf recycling (SiO_2 in glass excluded).

determined in the simulation and the emission factors for regional energy grid mixes from the GaBi database (thinkstep, 2018). Background process emissions (Scope 3) are only included for PV module glass, aluminium frames, mounting systems and cabling (De Wild-Scholten, 2013; Frischknecht et al., 2016; Stolz et al., 2017). Because of excluding other background emissions and simulating a closed-loop system, absolute CO_2-equivalent emissions presented here cannot be compared with other studies directly. As mentioned, we exclude the use phase as its emissions are virtually negligible, and the impacts of material transfers between life cycle steps, as it is assumed that all stakeholders are co-located.

3. Results and discussion

In this section, findings are reported for two key aspects of the life cycle: *material and energy resources* (consumption and efficiency) and *environmental impact* (in terms of CO_2 emission).

3.1. Conservation law analysis (based on balanced mass and energy flows)

The laws of conservation state that mass and energy must balance over all processes. The steady-state Si mass balance shown in Fig. 2 clearly shows the locations and relative magnitudes of Si-containing streams in the life cycle for Cases 1 and 2. Note that line thicknesses are to scale for total Si mass flow.

The *Recovered wafer Si* loop results from the 95% EOL recycling. Total kerf residue is shown as the two diagonal streams exiting wafering. With the progress already made, and with ongoing kerf recycling R&D, the assumption of 50% kerf recycling as MG-Si is believed to be conservative in terms of both quantity and quality. Based on the parameterisation of our simulation, considerable amounts of Si also leave the system as microsilica from MG-Si production and as unreacted MG-Si and $SiCl_4$ from the Siemens process.

3.1.1. Effects of recycling on nominal PV power production

As an example in the base case, 100 kt of quartzite consumption allows for a nominal PV power production of 10.4 GW_{peak}. In Case 1, this value increases by 86% (to 19.4 GW_{peak}) without additional quartzite consumption, and in Case 2 by 136% (to 24.5 GW_{peak}), the latter greater than 100% because, in this simulation, quartzite consumption is not displaced by the recycled Si. Without kerf recycling, primary quartzite consumption would have to increase to 126 kt to produce the same amount of modules, and to 237 kt if no recycling took place at all.

The combined effects of EOL and kerf recycling at fixed levels of quartzite consumption is shown in Fig. 3. Response surfaces show that the effect of EOL recycling on PV power production is non-linear, and that kerf recycling amplifies the benefits of EOL recycling i.e. the module production increase is stronger when kerf recycling complements EOL recycling. The three scenarios mentioned above are indicated as points on the response surface. Note that the non-linearity cannot be attributed to a single factor—it is rather a result of the recycling loops and various non-linear relationships that define the simulation.

Two additional data points are shown to put these numbers into perspective. In 2019, 4 GW_p's worth of PV modules were installed in Germany (Fraunhofer ISE, 2020), which, in our Case 1 simulation equates to a primary quartzite consumption of 20.5 kt. Without recycling, 38.6 kt of quartzite would be needed. Similarly, the EU-28′s 16.7 GW_p (Fraunhofer ISE, 2020) corresponds to the consumption of 85.5 kt quartzite with 95% EOL recycling and 161.0 kt without recycling. Fig. 3 quantifies the potential benefits of EOL, kerf and combined recycling at the RC/PV-power nexus, and highlights how system circularity can be

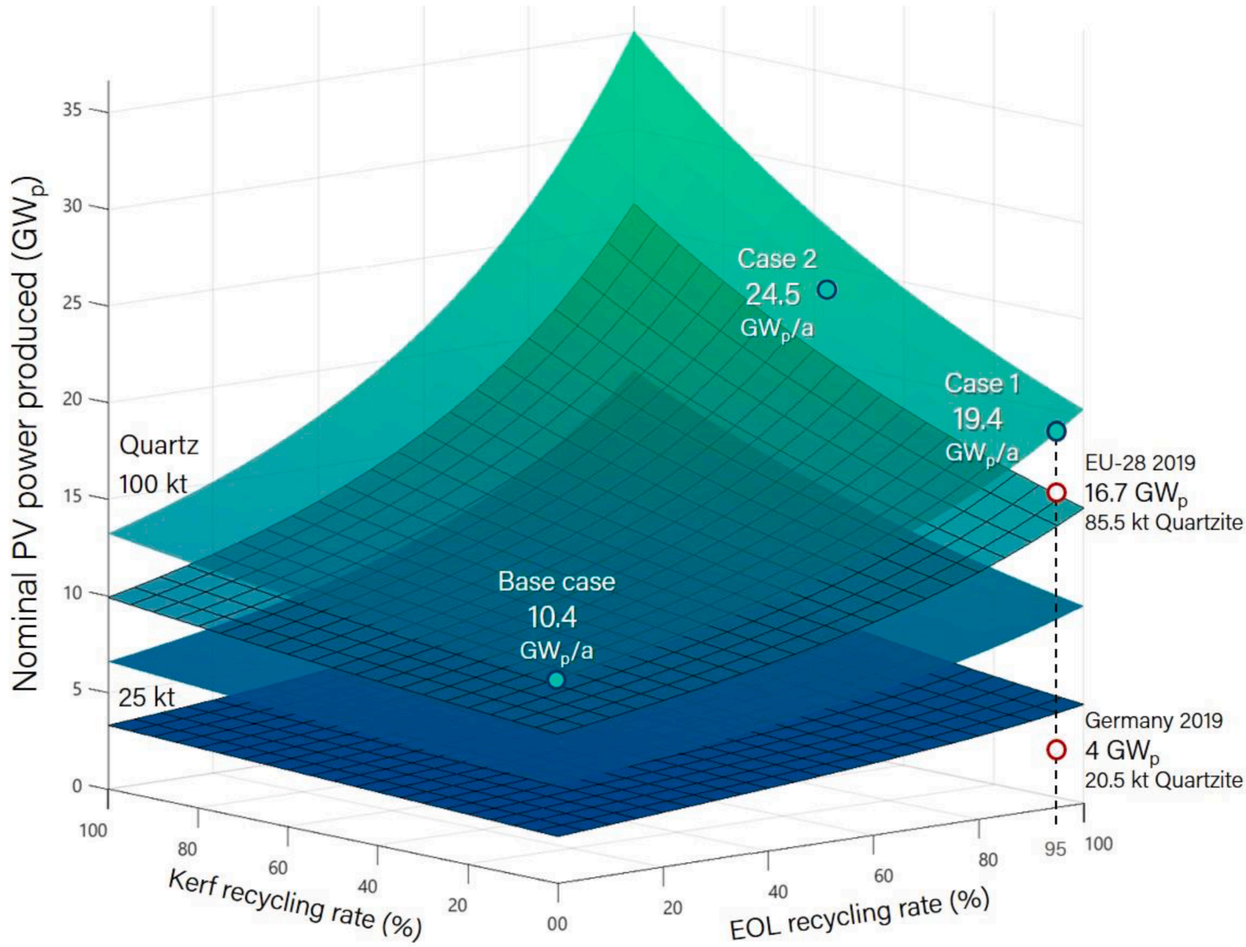

Fig. 3. The combined effects of EOL and kerf recycling on nominal PV power production at a nominal area efficiency of 230 W_p/m^2 (surfaces of constant quartz consumption are shown for reference).

used as a tool to help sustain projected PV deployment growth.

3.1.2. *Effects of recycling on power consumption*

Combining the above with a similar analysis of power consumption reveals the effects of recycling on power consumption per module area produced. Fig. 4 shows that EOL recycling reduces power consumption significantly, from 116 kWh/m^2 without recycling to 75 kWh/m^2 for Case 1, a 35% reduction. This can be explained by the fact that EOL-recycled Si bypasses both MG-Si production and the Siemens process, the largest electricity consumer in the system. The additional benefit of kerf recycling is small (73 kWh/m^2 compared to 75 kWh/m^2 for Case 1) because of kerf still going through the Siemens process (see Fig. 2). If future treatment processes were to recover kerf at SG-Si quality, energy savings would self-evidently increase. For Case 2, the simulation predicts a consumption of 64 kWh/m^2 when kerf is recycled into the Cz process instead—a 45% decrease from the base case. This excludes the energy consumption of any potential kerf treatment process, however, and may be overoptimistic.

3.1.3. *Material efficiency of the recycling process*

The recycling process (described in the Appendix) claims to recover 74% Ag and 83% Cu, and 85–90% wafer Si at SG-Si quality, and can remove at least 99% of Pb^{2+} from solution (Huang et al., 2017). Applying these recoveries to the EOL-recycled modules and assuming a 5% loss of wafers and strings during the dismantling and combustion steps, the overall metal recoveries shown in Fig. 5 are achieved.

Module composition and potential metal and other material recoveries are given in Table 2.

With an average Ag price of $521.48/kg in 2019 (macrotrends.net), potential revenue is approximately $109/tonne recycled ($2.60/module). Similarly, with the average Cu price of $7.29/kg, potential revenue amounts to $55/tonne recycled ($1.31/module) excluding cables, assuming that metals are recovered at saleable purity. This translates to an estimated revenue of $8.2 million per nominal GW_p for Ag and Cu alone. This is a rough indication based on present-day technology and economic conditions, however, as PV module lifetime, the cost of recycling, and technological developments such as the decreasing trends in Ag and Cu consumption (VDMA, 2020), amongst others, have not been considered in this paper.

3.2. *Exergy analysis (application of the SLT)*

3.2.1. *Exergy dissipation and efficiency*

As mentioned, exergy efficiency and cost are used as proxies for RE and RC, respectively, thus taking a thermodynamic perspective. Fig. 6 shows the relative magnitudes of exergy *flows* through the system for Case 2. Stream colours are designated as follows:

Dark blue: material and energy inputs from outside of the system (from the technosphere),

Yellow: waste streams and emissions (to the environment),

Green: byproducts and potentially useful streams to be treated outside of the system (to the technosphere),

Light blue: internal transfers of products,

Brown: internal recycle streams,

Orange: dissipated exergy i.e. the amount needed to close the exergy balance, representing the losses resulting from the creation of entropy.

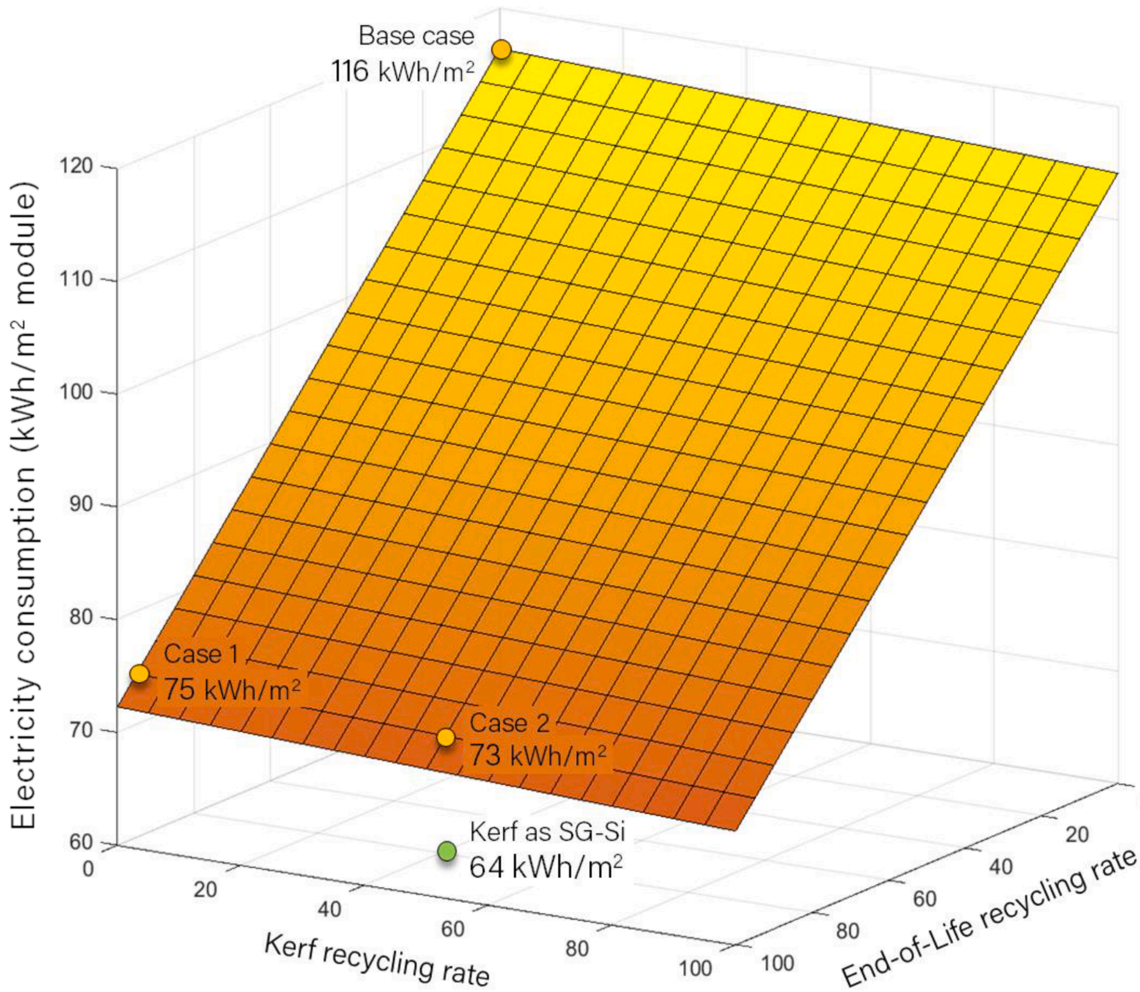

Fig. 4. Variation of specific power consumption with EOL and kerf recycling rate (axis directions are opposite to those in Fig. 3 for the sake of readability).

From Fig. 6, it is clear that the SG-Si and Cz processes consume large amounts of exergy (dark blue) relative to the intended onward exergy flows (light blue). These processes, followed by recycling and cell manufacturing, also dissipate the most exergy (orange). Module production, on the other hand, consumes a sizeable amount of exergy but does so efficiently with relatively little exergy dissipation (orange).

Associated exergy *efficiencies* are shown in Fig. 7. Also shown are efficiencies for a hypothetical zero-waste scenario in which all material and energy losses (yellow) are considered to be resources i.e. all waste have been "designed out" as promoted by prominent CE organizations. The difference between the two efficiencies represents, therefore, the theoretical RE improvement potential if it were possible to eliminate or transform all losses into resources. This should be what is aimed for in all the DfX domains. Highlighting the two process steps with the largest differences between the two efficiencies, and therefore opportunities for waste reduction: in the Cz and wafering processes, the difference is attributed to water treatment residues and the 50% kerf loss. In the PERC cells process it is the result of losses from the layer deposition steps and the wafer texturing liquor.

Important to note is that even zero-waste efficiencies are well below 100%, the primary cause being the inevitable dissipation of exergy. Thus, even if *material* loops could be closed, total circularity is impossible when exergy dissipation is accounted for. Fig. 7 is, therefore, a representation of the thermodynamic *limits of circularity*. However, by minimising exergy dissipation through optimisation and innovation in technology, processes and supply chains, the root causes of open loops e.g. downcycling, carbon emission and energy loss will be addressed automatically as these are the main dissipaters of exergy. Waste heat recovery and reducing the consumption of electricity and high-carbon feed materials like fossil fuels, polymers and other organic compounds are good starting points, as these carry significant amounts of exergy that are usually dissipated in their transformation processes. In this paper, the maximum efficiencies represent our simulation model in its current configuration only, and cannot be applied to other systems.

3.2.2. *Exergy cost*

Analogous to monetary cost, exergy cost is an accounting quantity that represents the thermodynamic cost of a product, based on the theory of exergetic cost (Lozano and Valero, 1993). While exergy itself is *not* subject to the conservation laws, exergy cost is additive i.e. the exergy cost of a product is the sum of the costs of its constituents. It allows for the specific causes of exergy dissipation and their relative contributions to be identified, as is shown in Fig. 8. For the production of PV modules, our simulation gives an exergy cost of 50.3 kWh/m^2 (or 98.8 kWh/module). Note that, while exergy cost is expressed in kWh, it represents exergy dissipation from both energy and material streams. For comparison, we found the specific exergy cost for CdTe PV to be 118 kWh/module (Bartie et al., 2020).

The largest contributor to module exergy cost is the Cz process (31%), followed by deposition of the AlO_x cell layer (14.5%), phosphorous deposition (9.6%), wafer sawing (8.4%), and SiN_x deposition (8.4%). Based on our simulation, innovation in these processes would realise the greatest decreases in RC. In the Cz process, irreversibility mainly stems from power consumption and the subsequent loss of waste

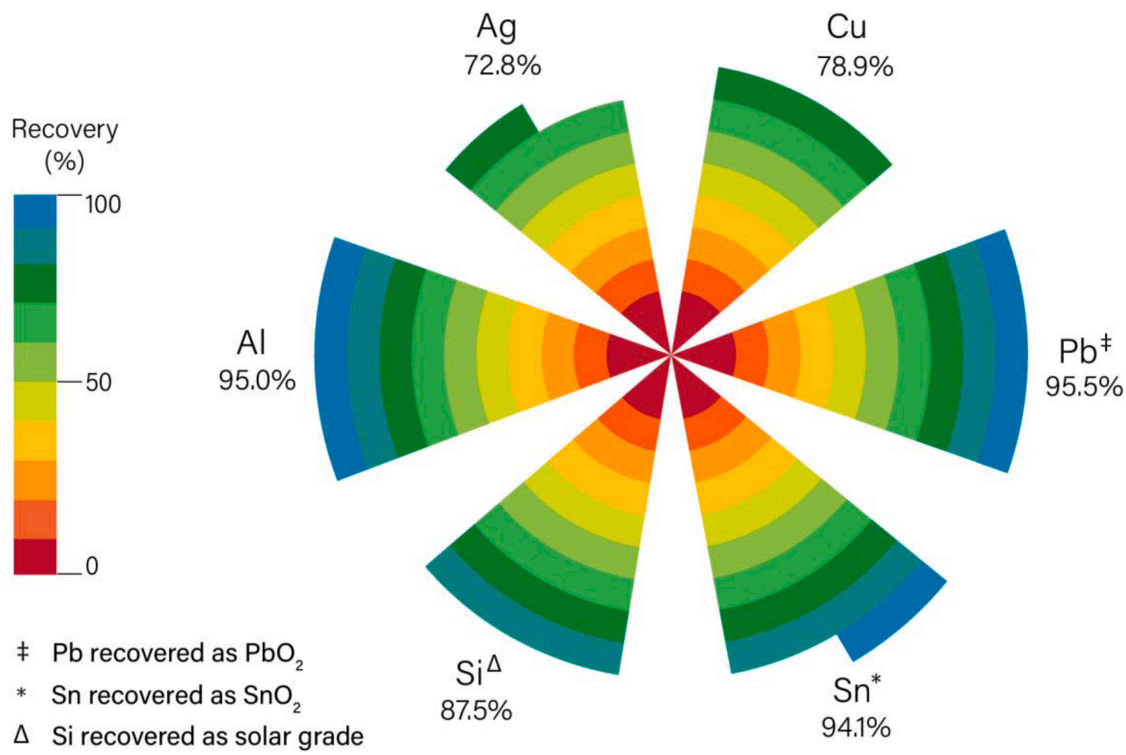

Fig. 5. Metal recoveries from the simulated recycling process.

Table 2
Module composition and material recovery.

Component	Content (kg/t module)	Recovered† (kg/t module)
Al (as Al_2O_3, excl. frame)	2.728	2.332
Ag	0.3313	0.2096
Cu	10.86	7.532
Pb (as PbO_2)	0.07797	0.06703
SnO_2	6.045	5.111
Si (solar grade)	30.98	24.15
Glass	661.0	654.4*
Polymers	111.3	–
Rest	1.243	–

† excluding intact cells (10% assumed in this paper).
* based on an estimated 99% glass recovery (Heath et al., 2020).

heat. This is also the case for the deposition and wafering processes, with smaller amounts of dissipation resulting from material losses. The *rest* contribution (13.5%) consists of those for the Siemens CVD reactor; module soldering, edge sealing and framing, and several others, each with an impact of less than 1%.

3.3. Carbon emissions

Total CO_2 generation is determined as the sum of (i) that generated directly in each process step (described in Section 2.2.4), (ii) 1.1 $kgCO_2$/kg glass (thinkstep, 2018), (iii) 62.4 $kgCO_2/m^2$ for the frame, mountings, cables, and connectors (Wernet et al., 2016; De Wild-Scholten 2013), and (iv) that associated with the electricity/fuel consumption of individual process steps. Fig. 9 shows the CO_2-equivalent emission per module area produced for the German energy grid mix and quantifies the sustainability increases potentially achievable via increased circularity. For the base case, emissions amount to 146 $kgCO_2/m^2$, decreasing by 14% (to 125 $kgCO_2/m^2$) for Case 1 and by 15% (to 124 $kgCO_2/m^2$) for Case 2. As for power consumption (Fig. 4), the decreases can be attributed to EOL recycling bypassing the Siemens process, while kerf recycling does not. Similarly, the upgrading of kerf residue to SG-Si purity before recycling could bring about further decreases in CO_2 emission, in this case a further 3% to 120 $kgCO_2/m^2$. It highlights and quantifies the potential benefits of innovation in kerf recycling, so as to upcycle it to higher value solar grade purity, in which case it would also bypass the Siemens process.

While direct CO_2 emissions remain constant for a given production configuration, total emissions depend strongly on the energy grid mixes at production locations. Fig. 10 depicts the same information as Fig. 9 for the energy grids of various other countries, assuming that all life cycle steps are co-located.

With the high proportion of hydroelectricity, Norway is clearly the best performer with the lowest absolute emissions. Interestingly, because process-generated (Scope 1) emissions comprise almost 88% of the total (Scope 1 + 2) on the Norwegian grid, the benefit of bypassing the Siemens process is outweighed by process-generated CO_2—EOL recycling *increases* net emissions slightly, hence the reversed slope for the Norway surface. For the other regions, the Scope 1 share ranges between 18 and 42%, making the effects of changes in grid-related emissions more pronounced. Australia's and China's high reliance on coal make them the worst performers. In a hypothetical situation where the entire system is located in either Norway or Australia, CO_2 emissions are a factor of 2.3 higher in Australia with no recycling, and 1.8 times higher for Case 2 because of the dominance of fossil fuels in the grid mix, as is also the case for China. Europe's Si metal imports originate primarily from Norway and Brazil, the two best performers, and to a lesser extent from China (European Union (EU), 2017b). At present, only China holds the full supply chain domestically.

3.4. Technology change

A total technology change can affect the system significantly in terms of RC and impacts. Fig. 11 depicts normalised response surfaces for absolute power consumption and CO_2 emission for the current system and shows that they trend in the same direction. Increased recycling leads to more modules being produced, higher CO_2 generation during

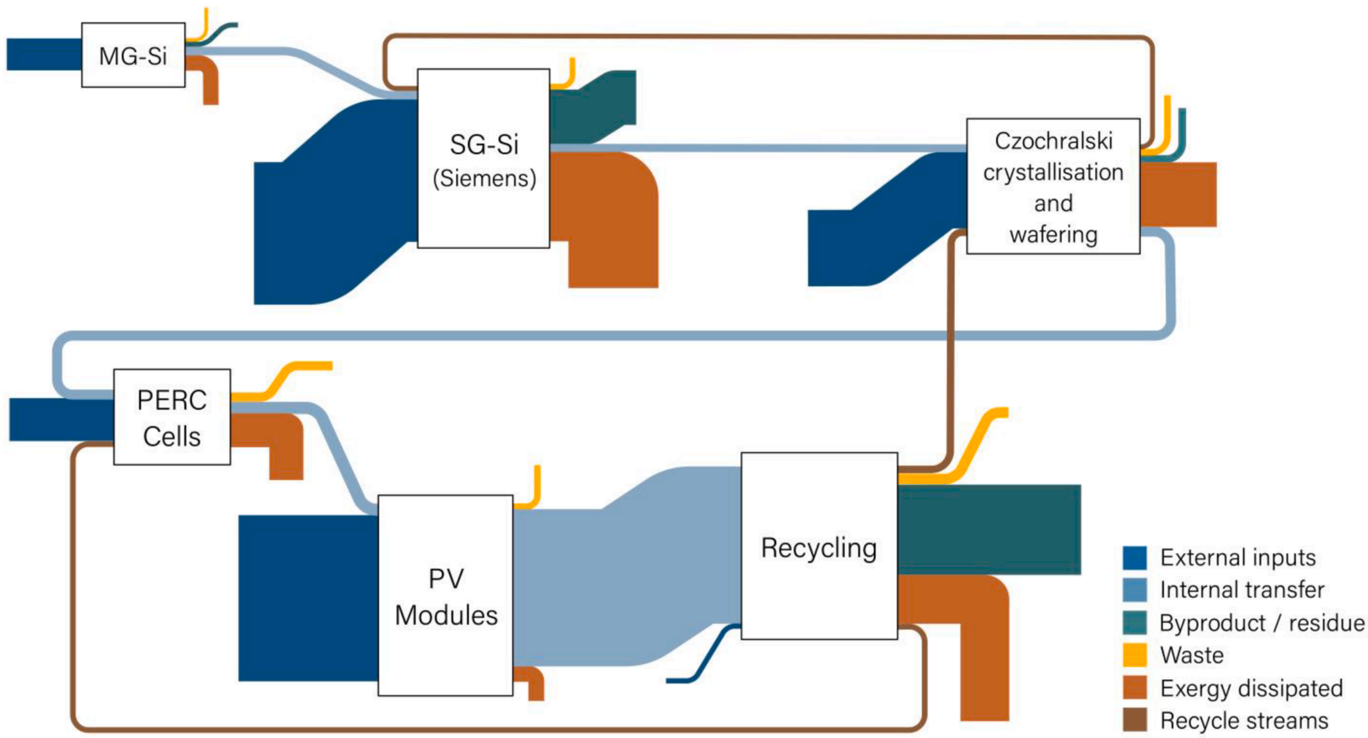

Fig. 6. Flows of exergy through the mono-Si life cycle (Case 2).

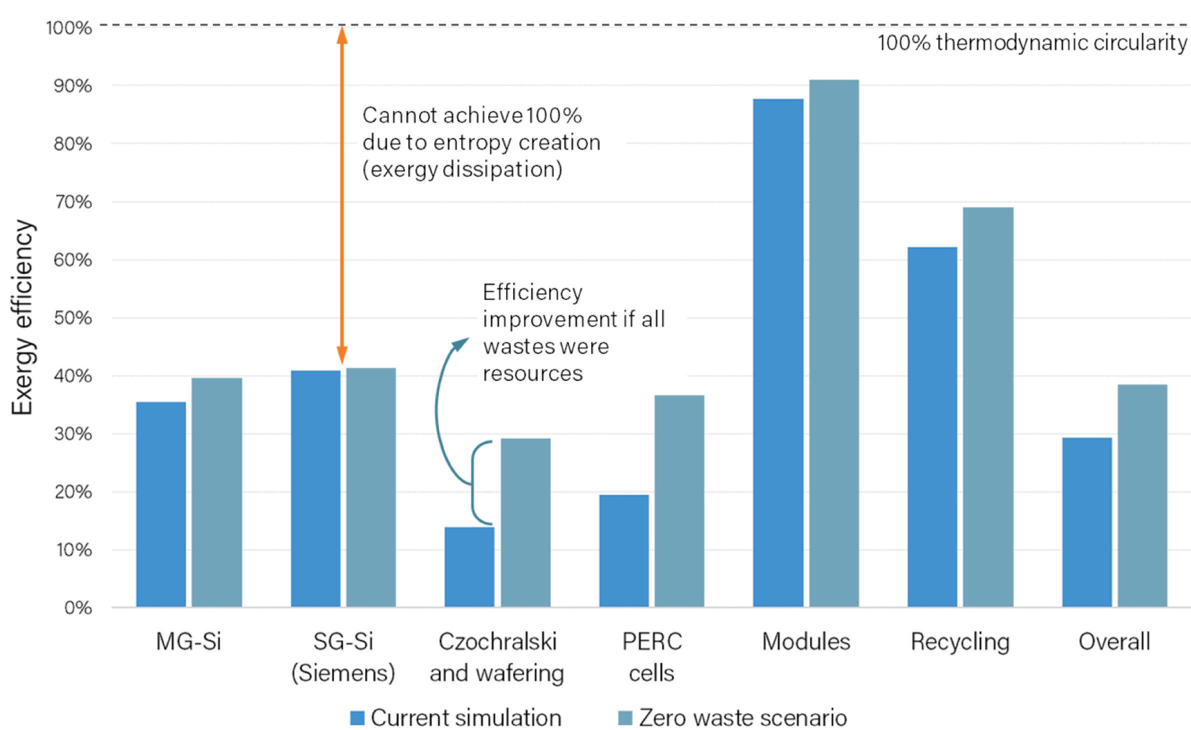

Fig. 7. Exergy efficiency for each process in the life cycle based on Case 2 and for a zero-waste scenario (note that overall efficiency is not the product of individual efficiencies, as each process step has external inputs).

polymer incineration, and higher net power consumption.

Fig. 12 shows the equivalent surfaces for a system in which SG-Si is produced via the silane FBR route, and mc-Si is used instead of mono-Si.

Here, surface slopes are *opposite* with respect to EOL recycling. As EOL recycling increases, net CO_2 emission increases due to increased polymer incineration. At the same time, the additional power generated via heat recovery decreases the net power requirement. In contrast, the Siemens process completely absorbs the power generated from waste heat in the mono-Si system. This highlights the importance of waste heat recovery in balancing overall system power consumption. To further illustrate potential differences, Fig. 13 shows that, on average, CO_2 emission per module area for the FBR process is around 27% lower than

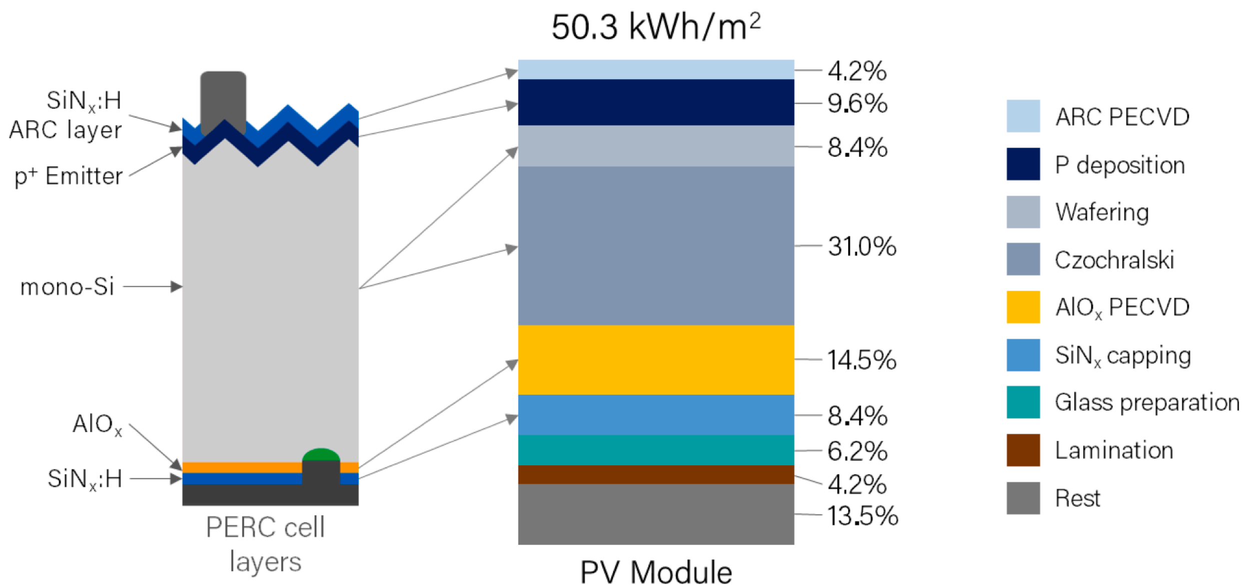

Fig. 8. Breakdown of contributions to overall module exergy cost (links to PERC cell layers shown on the left).

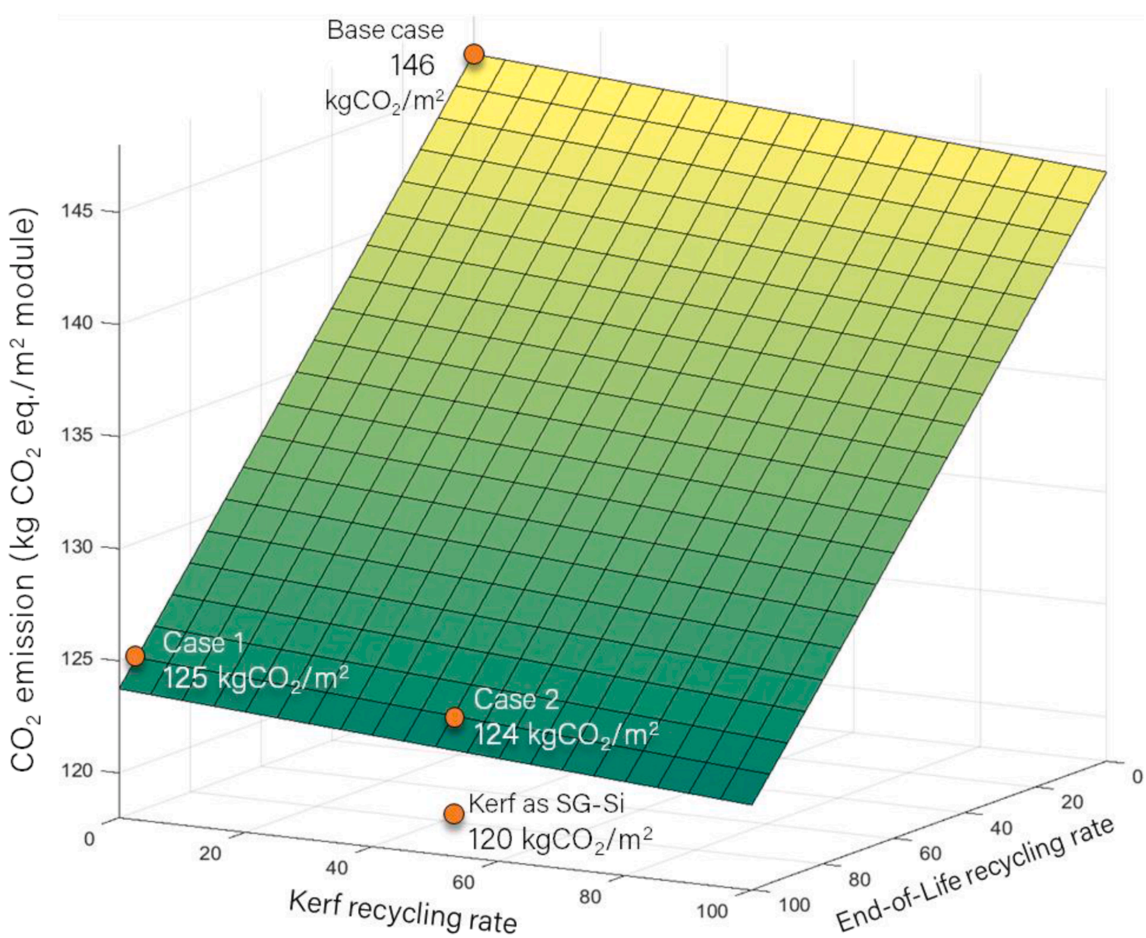

Fig. 9. CO_2-equivalent emission per m^2 module, with all life cycle steps co-located on the German energy grid.

that for the Siemens process.

Systems can be configured to combine any industrially viable technology combinations using the simulation approach. For instance, selecting a recycling process based on polymer pyrolysis rather than combustion would again change the picture as direct CO_2 emissions would not occur, while at the same time no additional power would be generated from waste heat. The approach presented here offers the flexibility to evaluate and compare the systemic effects of such changes relatively easily, whereas the data for such processes do not exist in the inventory databases as yet.

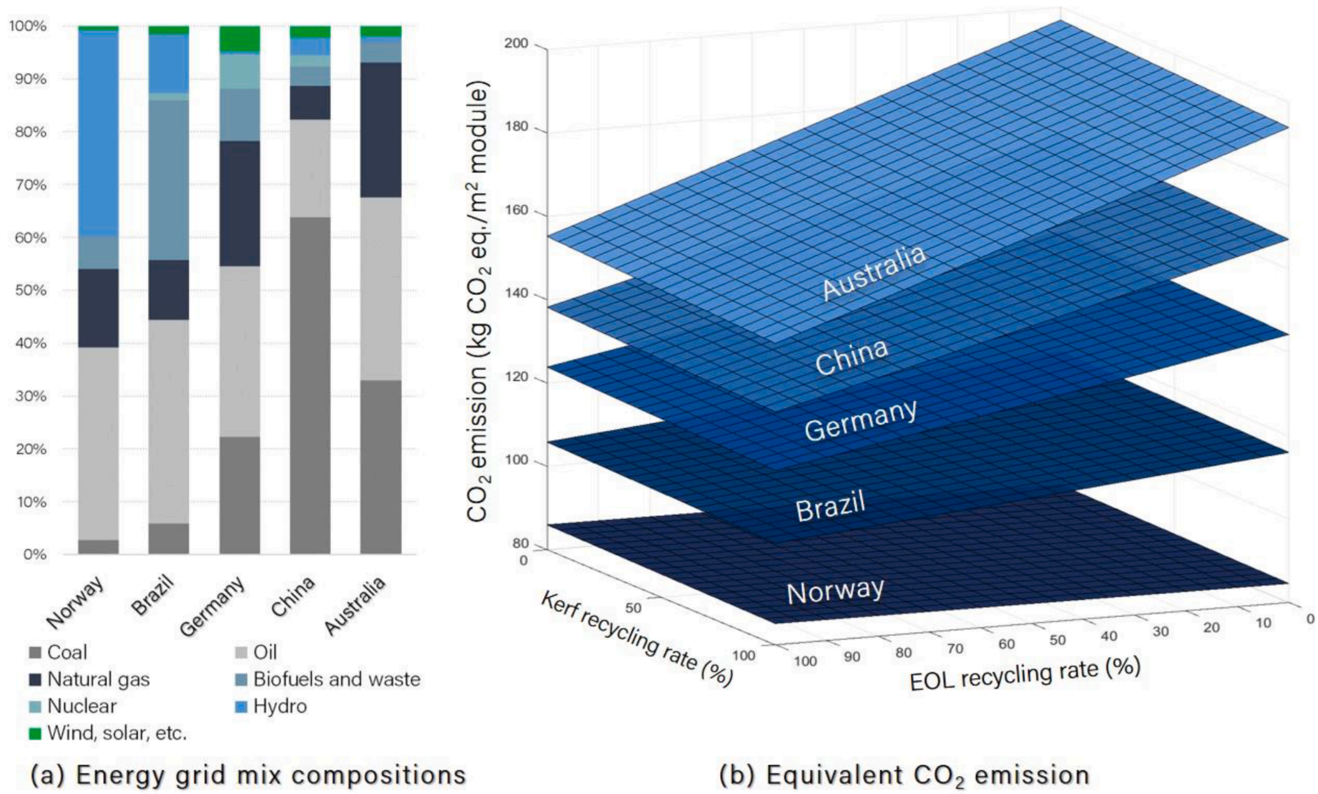

Fig. 10. CO_2 emissions for different energy grid mixes, assuming all life cycle steps are co-located. Based on 2017 data from the IEA (IEA, 2020b).

4. Conclusions

While the elaboration of the PV industry is primarily driven by cost, this study highlights that sustainable CE requires a wider lens. Resource extraction and waste treatment requirements, options for the recovery of secondary resources at high purity, and impacts in all the dimensions of sustainability need to be assessed and optimised if the PV industry is to not just deliver renewable energy, but do so in a sustainable way that does, in fact, facilitate decarbonisation. Industry performance against this goal can only be measured using a highly granular systems approach that considers the full life cycles that PV systems fit into. It is, therefore, recommended that life cycle systems be analysed using rigorous approaches such as that presented in this paper: opportunities for enhancing sustainability and circularity are identified down to the micro level for unit operations, for the processes they are the building blocks of, and the life cycle systems formed by connecting these processes. The non-linear nature of the life cycle is captured in a scalable, deterministic simulation model to predict the effects of various parameter changes on resource requirements and sustainability, to explore performance improvement options, and to compare different processing and design configurations in terms of RC, RE, and carbon footprint. Computational flexibility and efficiency are enhanced by employing NNs as surrogate representations of the simulation. The simulation approach makes it possible to compare and optimise current and future technology options based on up-to-date processes and operating parameters, rather than having to apply aggregated and sometimes outdated information, as is the status quo with most of the general MFA-based LCA approaches. These methods do not allow for delving into the process specifics of large systems to identify and address the root causes of material and energetic inefficiencies, entropy creation, and environmental emissions.

Simulation results establish the system configurations that would facilitate resource conservation and environmental impact reduction, and quantify the positive impacts of increased circularity on overall sustainability. Both EOL and kerf recycling increase PV module production capacity and hence, nominal PV power generation capacity without the need for additional primary material consumption (Fig. 3). Furthermore, both power consumption and CO_2 emission per module are driven down by increased EOL recycling because of recycled Si bypassing both MG-Si production and SG-Si production via the energy-intensive Siemens process. As kerf recycling only bypasses MG-Si production, its impact is less pronounced. However, if kerf residues were to be upgraded to SG-Si prior to re-entering the value chain, its recycling would provide significant additional reductions in power consumption and CO_2 emission (Figs. 5 and 10). There is a need for continued focus on developing high-value recycling processes for both EOL modules and kerf, on reducing kerf formation and contamination to the minimum practically and economically achievable, and/or eliminating kerf completely through alternative processing techniques e.g. Si deposition methods that eliminate the need for wafer cutting.

Power consumption-related CO_2 emissions (Scope 2) depend strongly on energy grid composition. We highlight the significantly poorer environmental performance of countries that still depend mostly on fossil fuels for power generation (Fig. 10). If energy-intensive production processes are to remain the norm, they should be geographically

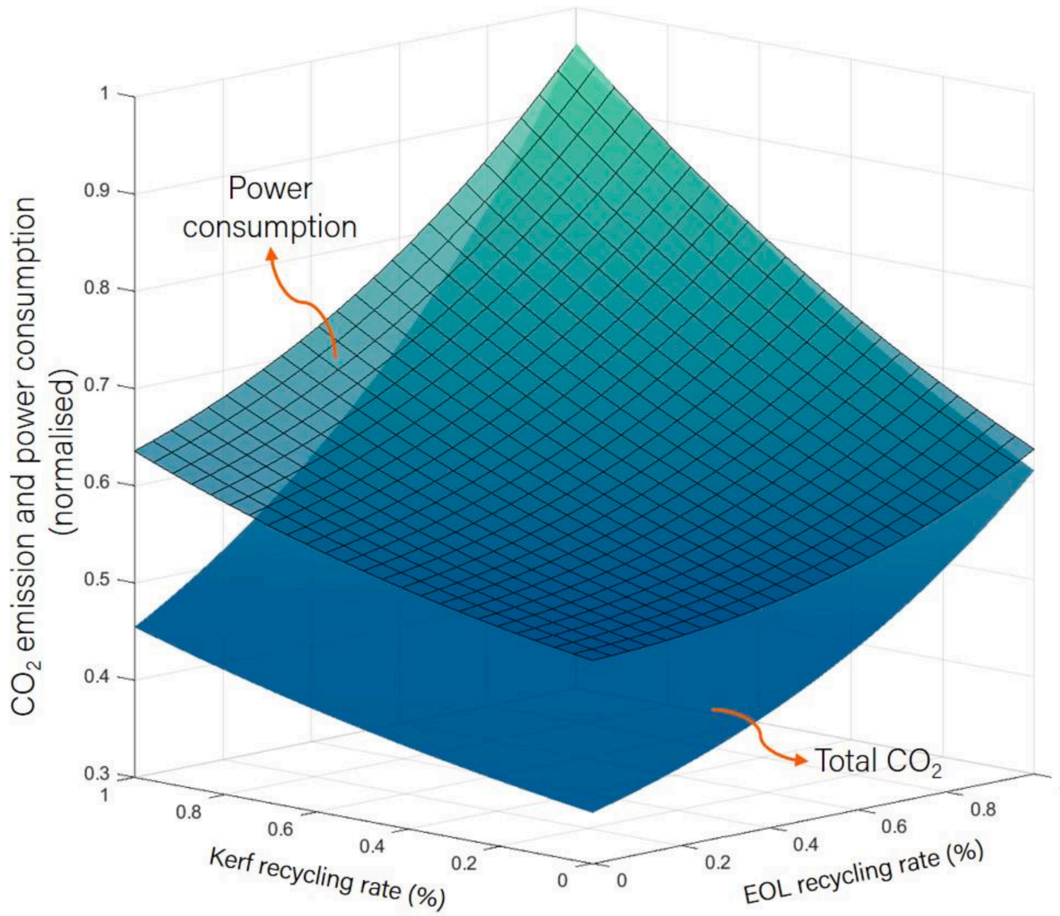

Fig. 11. Variation of power consumption and CO_2 generation (normalised) with recycling rate (Siemens Process SG-Si and Cz mono-Si).

located where energy grids are least dependant on fossil fuels. There is, of course, an economic trade-off to consider but there should be a shift from the primarily *profit*-orientated view to one that strikes a balance between *people, planet,* and *profit.* From a policy perspective, instruments such as life cycle carbon pricing likely have a role to play.

From a thermodynamic perspective, Cz crystallisation, wafering, and cell manufacture are the least resource-efficient processes in the closed-loop system (Fig. 7). Further analysis, using exergy cost as a proxy for RC, reveals that the specific causes are Cz crystallisation itself and AlO_x layer deposition, followed by the other cell layer deposition steps. Based on our assumptions in these unit operations, exergy dissipation mainly stems from power consumption without waste heat recovery, and losing materials due to deposition inefficiencies. To limit systemic entropy creation, and by implication the dissipative loss of resources, developing innovations that target these inefficiencies first is recommended.

Simulation results show that, amongst others, Ag and Cu to the value of approximately \$109 and \$55, respectively, can be recovered per tonne of modules recycled (based on 2019 prices). Technoeconomic analyses should be conducted to further explore revenue potential and the trade-offs between high-value recycling at (first) EOL and dismantling for re-use to extend the life cycle and delay recycling. Based on the chosen recycling route, we find that at least 95.5% of Pb introduced via solders can be removed from recycling solutions to prevent potential release into the environment.

To take advantage of these opportunities, supply chains need to be integrated or designed such that collaboration and quality-focussed recycling are stimulated. In Europe, the WEEE directive and organisations like PV Cycle (pvcycle.org) that facilitate the collection and recycling of EOL modules contribute significantly to these, albeit that WEEE directive targets are largely mass based. Our simulation shows that, by the time EOL modules have been thermally processed (after removal of the frame, glass and polymers, before commencement of metal recovery), current WEEE directive targets have already been exceeded. At the policy level, a shift from mass-based targets to targets for both quantity and quality recycling with respect to specific elements is needed. Policy makers should develop these targets guided by approaches such as that presented here, the contribution of which lies in its agility and its ability to translate the complexities of large non-linear product systems into a consistent physics-based foundation of information. From here, industry and policy makers can:

- make properly informed decisions about future directions for the PV and other industries,
- effectively communicate up-to-date sustainability-related information in a rapidly changing environment (e.g. through the development of performance indicators and labels), and

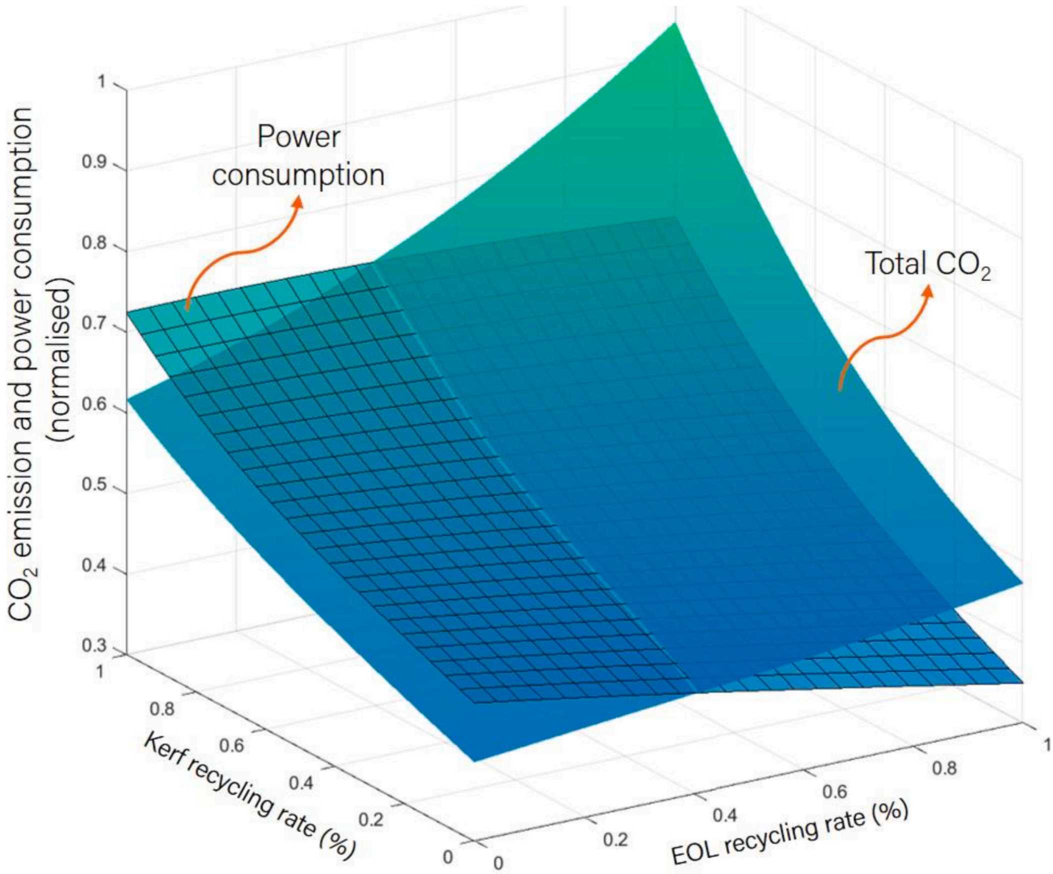

Fig. 12. Variation of power consumption and CO_2 generation with recycling rate (silane FBR SG-Si and mc-Si).

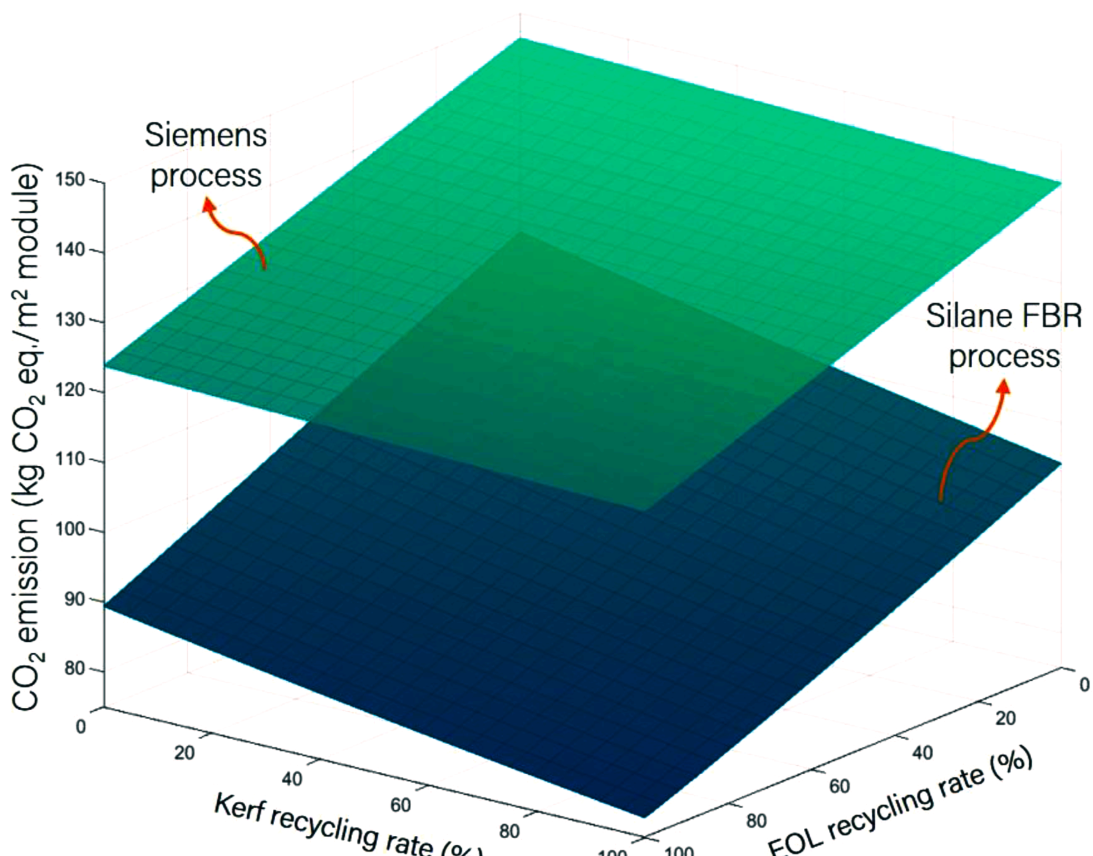

Fig. 13. Variation of CO_2-equivalent emission per m^2 with recycling rate (note that the top surface is the exact same as that shown in Fig. 9).

- take realistic and impactful operational and regulatory actions to stamp out barriers on the path to maximum sustainability and circularity.

It is, therefore, ideally placed to support decisions regarding decarbonisation, the transformation of global economies to CE, realising the EU Green Deal, and achieving the United Nations' Sustainable Development Goals. It is not limited to the systems covered in this paper, but can be applied to any other PV or complex product system.

5. Outlook

The results presented in this paper are for a static, steady state system based on the current status of the c-Si PV industry. The simulation model can, however, be adopted fairly easily to investigate the systemic effects of technological developments in e.g. wafer size and thickness, materials used, recycling strategy, and so on, as well as projected PV deployment rates, lifetimes and expected waste volumes over time.

We addressed resources and one dimension of sustainability i.e. the environment, and touched on some of the economic benefits of EOL metal recovery. Future work includes the additional assessment of economic and societal impacts, so as to assess and optimise the system across all three dimensions of sustainability.

We alluded to the fact that this approach can be applied to other systems. Significant R&D currently focusses on the development of tandem PV technologies that better exploit solar energy to bring about step changes in PCE. In particular, the development of perovskite-based PV is gaining significant traction. As such, the simulation of c-Si/perovskite tandem PV systems is currently underway. The methodology is being expanded to also include technoeconomic assessments and the optimisation of sustainability and circularity.

Credit author statement

Neill Bartie: Conceptualization, Methodology, Software, Validation, Formal Analysis, Investigation, Writing – Original Draft, Visualization; **Lucero Cobos-Becerra:** Conceptualization, Validation, Investigation, Writing – Original Draft, Writing – Review & Editing, Visualization, Project Administration; **Magnus Fröhling:** Conceptualization, Validation, Writing – Review & Editing, Supervision; **Rutger Schlatmann:** Conceptualization, Methodology, Validation, Writing – Review & Editing, Supervision; **Markus Reuter:** Conceptualization, Methodology, Software, Validation, Writing – Review & Editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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6. Appendix A

6.1. Methods

Fig. A1.1 shows a schematic representation of such a network and its relation to the process simulation.

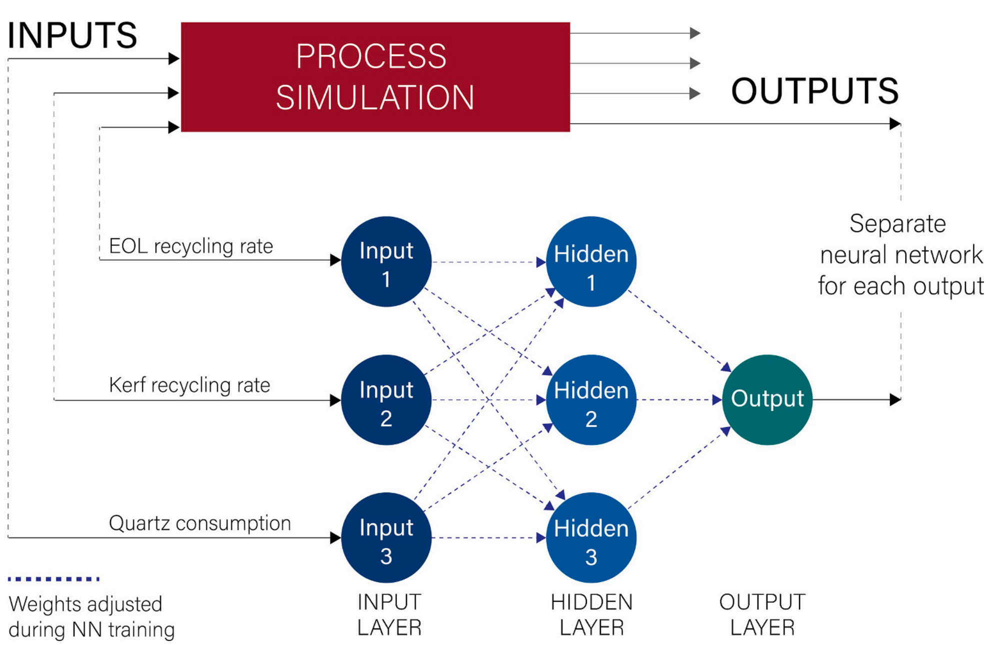

Fig. A1.1. Neural network structure and links to process simulation in/outputs (note that hidden and output layers each have a bias neuron, which are not shown).

6.2. Process Descriptions and Assumptions

The simulated life cycle consists of 75 process units and 333 process streams containing 155 compounds. The main process steps and assumptions are briefly described here.

6.3. Metallurgical grade silicon

Metallurgical grade silicon (MG-Si) is produced by the carbothermic reduction of silica (SiO_2). Quartzite sand is smelted in an electric arc furnace (EAF) with a number of other materials essential to the process (e.g. coke, charcoal, and woodchips) (Zulehner et al., 2000; Øvrelid and Pizzini, 2017).

As quartz (SiO_2) melts in the cooler zones (below about 1835 °C), it reacts with carbon to form silicon carbide (SiC), which acts as the reductant in producing the final silicon metal, according to reactions (1) and (2).

$$SiO_2 + C \ = \ SiO(g) + CO(g) \tag{1}$$

$$SiO(g) + 2\,C = SiC(s) + CO(g) \tag{2}$$

In the high-temperature zone in close proximity to the electrodes (at 1900–2100 °C), reactions (3) and (4) take place (PCC, 2017) to produce crude silicon at a typical yield of between 80 and 90% (Ciftja et al., 2008), and a gas phase containing primarily CO, SiO and H_2O. For the purposes of our simulation, we assume a yield of 85%.

$$2SiO_2(l) + 3SiC(s) = 4Si(l) + SiO(g) + 3CO(g) \tag{3}$$

$$SiO(g) + SiC(s) = 2Si(l) + CO(g) \tag{4}$$

Any SiO(g) that enters the offgas stream reacts according to reaction (5) to form a silica fume.

$$SiO(g) + CO(g) = SiO_2 + C \tag{5}$$

The fume byproduct—microsilica—is captured in the gas cleaning system and sold as a valuable additive in the construction and refractory industries (Ciftja et al., 2008). Crude silicon purity is typically 96 to 99% depending on the quality of raw materials and EAF electrodes used. The main impurities dissolved in the metal phase are given in Table A2.1 (Ceccaroli and Lohne, 2011).

Further purification is achieved through ladle refining, during which the addition of slagging agents such as SiO_2 and CaO allow for impurities less noble than Si (e.g. Al, Ca and Mg) to be oxidized. Temperature is maintained by blowing oxygen through the melt, leading to the exothermic oxidation of Si, so resulting in a loss of product Si. The reactions involved are listed below, with underlined elements dissolved in metal, and those in parentheses dissolved in slag (Ceccaroli and Lohne, 2011).

$$4Al + 3(SiO_2) = 3Si(l) + 2(Al_2O_3) \tag{6}$$

$$2Ca + (SiO_2) = Si(l) + 2(CaO) \tag{7}$$

$$2Mg + (SiO_2) = Si(l) + 2(MgO) \tag{8}$$

$$Si(l) + O_2 = (SiO_2) \tag{9}$$

Gibbs free energy minimization is used to estimate the thermodynamic equilibrium distributions of Al, Ca and Mg between the metal and slag phases. Carbon, in the form of suspended SiC particles, is also removed with ladle slag. Some dissolved C remains in the metal phase, however, at levels between 100 and 600 ppm (Ceccaroli and Lohne, 2011; Xakalashe and Tangstad, 2011). Our simulation gives a value of 182 ppm.

Boron (B) and Phosphorous (P) are electrically active elements that need to be removed to ultimately be added back into high-purity Si as dopants in carefully controlled quantities during PV cell manufacture. Boron is removed during ladle refining according to reaction 10 (Safarian et al., 2012).

$$2B + 11/2SiO_2 = (B_2O_3) + 11/2Si \tag{10}$$

The removal of B via ladle refining can be affected negatively by the presence of Al_2O_3 in the slag (Jakobsson and Tangstad, 2014). Applying these authors' findings, the refining slag Al_2O_3 content in our simulation results in a residual B content of 78 ppm in the metal phase. We assume a final P concentration of 25 ppm (Miki et al., 1996). The distributions of other contaminants are calculated such that the final product composition agrees with those in various academic publications (e.g. Xakalashe and Tangstad, 2011; Ceccaroli and Lohne, 2011; Zulehner et al., 2000; Øvrelid and Pizzini, 2017). After ladle refining, slag is removed and the refined Si metal is cast into ingots, cooled, and then crushed and sized. The process simulation flowsheet is shown in Fig. A2.1.

The quantities and compositions of feed materials and EAF electrodes relative to the input amount of quartzite are taken from literature, both academic and industrial (PCC, 2017; Zulehner et al., 2000; Chigondo, 2018; Chandrasekaran et al., 2012). EAF specific energy consumptions reported in the literature range from 11 to 14 MWh/t of metal produced (Zulehner et al., 2000; Xakalashe and Tangstad, 2011). We deduce a value of 11.4 MWh/t from information published by PCC BakkiSilicon hf. (PCC, 2017) and estimate power consumption and heat losses based on expected product temperatures and a closed energy balance. Heat is partially recovered from the hot offgas stream, and is used to generate electricity, which reduces the primary energy demand. Product, residue and direct emission (e.g. carbon dioxide) quantities are calculated from the chemical and physical transformations described above, validated and adjusted as appropriate to match industrial reality while maintaining closed mass balances.

Table A2.1
Impurities in crude metallurgical grade silicon.

Iron (Fe)	0.2 – 3 wt%	Titanium (Ti)	0.01 – 0.1 wt%
Aluminium (Al)	0.4 – 1 wt%	Carbon (C)	0.1 – 0.15 wt%
Calcium (Ca)	0.2 – 1 wt%	Oxygen (O)	0.01 – 0.05 wt%
V, Cr, Mn, Co, Ni, Cu, Zr and Mo	tens to hundreds of ppm(w)	B, P	10 – 100 ppm(w)

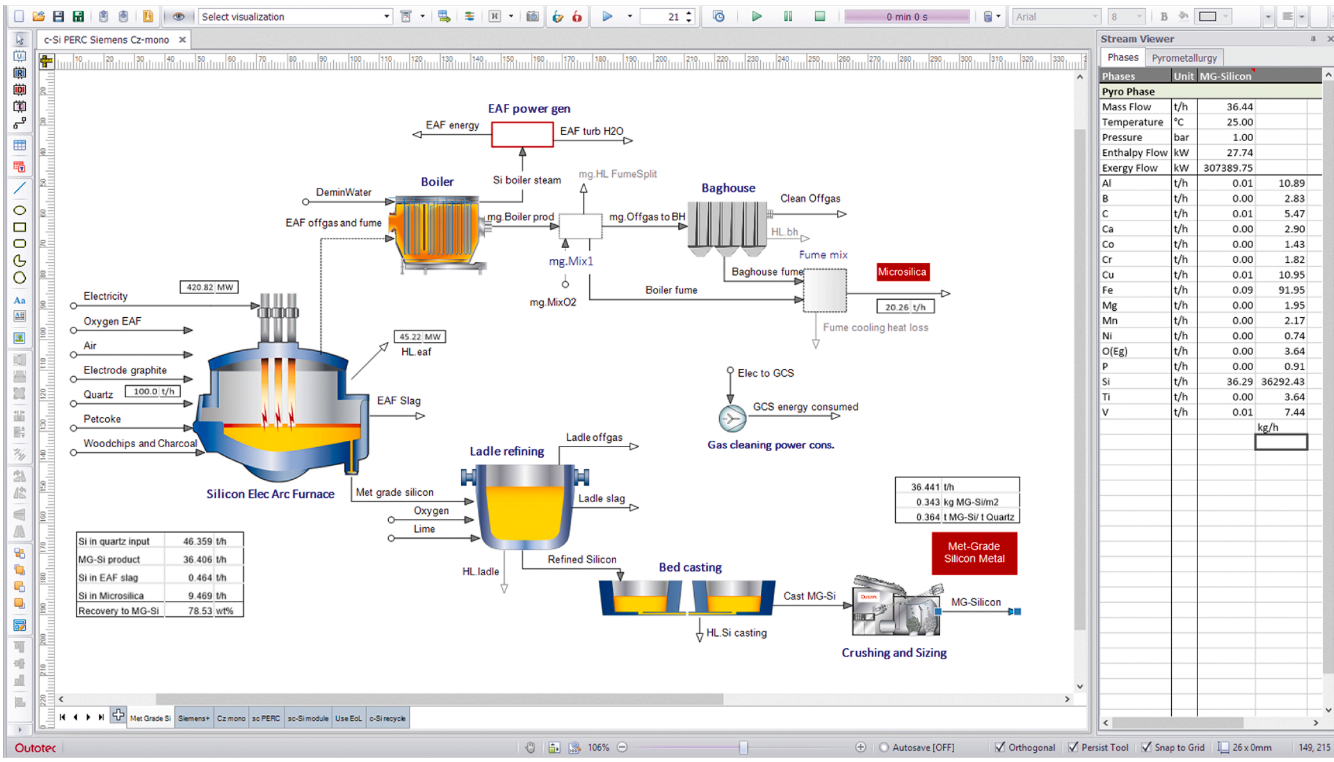

Fig. A2.1. Simulation flowsheet for metallurgical grade silicon production.

6.4. Solar grade silicon

Most manufacturers adopt the established, but energy intensive, Siemens process for polysilicon production. Process improvements have led to energy consumption decreasing from 200 kWh/kg Si in 2009 (Yan, 2017) to around 71 kWh/kg Si in 2018 (IEA PVPS, 2019). To date, less than 5% of manufacturers have adopted the far more efficient FBR process, which consumes significantly less energy (around 10 kWh/kg Si) (IEA PVPS, 2019). Furthermore, tariffs and trade tensions between the USA and China have put producers under pressure—REC Silicon, for example, in an effort to maintain liquidity, has had to halt FBR operations at its Moses Lake production facility due to insufficient access to Chinese polysilicon markets (REC, 2019). Nonetheless, greater adoption of lower-energy processes would reduce energy payback time (EPBT), a PV performance indicator that measures the time required for a PV system to generate the amount of energy consumed for its manufacture, so increasing sustainability. For the main simulation of this life cycle stage, the Siemens Process is used. The simulation flowsheet is shown in Fig. A2.2.

MG-Si is reacted with hydrogen chloride (HCl) in a FBR at temperatures between 300 and 500 °C and pressures between 1 and 5 bar to produce trichlorosilane (TCS - $SiHCl_3$) and some silicon tetrachloride (STC – $SiCl_4$) (Bye and Ceccaroli, 2014) according to reactions (11) and (12).

$$Si + 3HCl(g) = SiHCl_3(g) + H_2(g) \tag{11}$$

$$Si + 4HCl(g) = SiCl_4(g) + 2H_2(g) \tag{12}$$

A selectivity of 90% towards TCS, the desired product, is achieved by adding a 10% excess of HCl (Ramírez-Márquez et al., 2018). It is assumed that MG-Si impurities exit the system in this step with unreacted Si (assumed to be 5% of input Si) and in the offgas stream. In order to estimate HCl consumption more accurately, the compounds formed from the impurities are predicted using Gibbs free energy minimisation in FactSage (GTT-Technologies, 2020) and are as follows:

Solid phase: $FeCl_2$, $MnCl_2$, $CaCl_2$, $MgCl_2$, $CrCl_2$, Cu_3P, VCl_2, Co_2P, and Ni_5P_2

Gas phase: $AlCl_3$, BCl_3, $TiCl_4$

TCS is subsequently separated from STC and further purified in two fractional distillation steps. Si deposition takes place in a Siemens chemical vapour deposition (CVD) reactor, in which TCS is diluted in pure hydrogen (H_2) to decompose and deposit SG-Si onto pure Si filaments at 1000–1100 °C (Jiao et al., 2011; Safarian et al., 2012).

The main reactions that occur in the deposition reactor are as follows (Ceccaroli and Lohne, 2011):

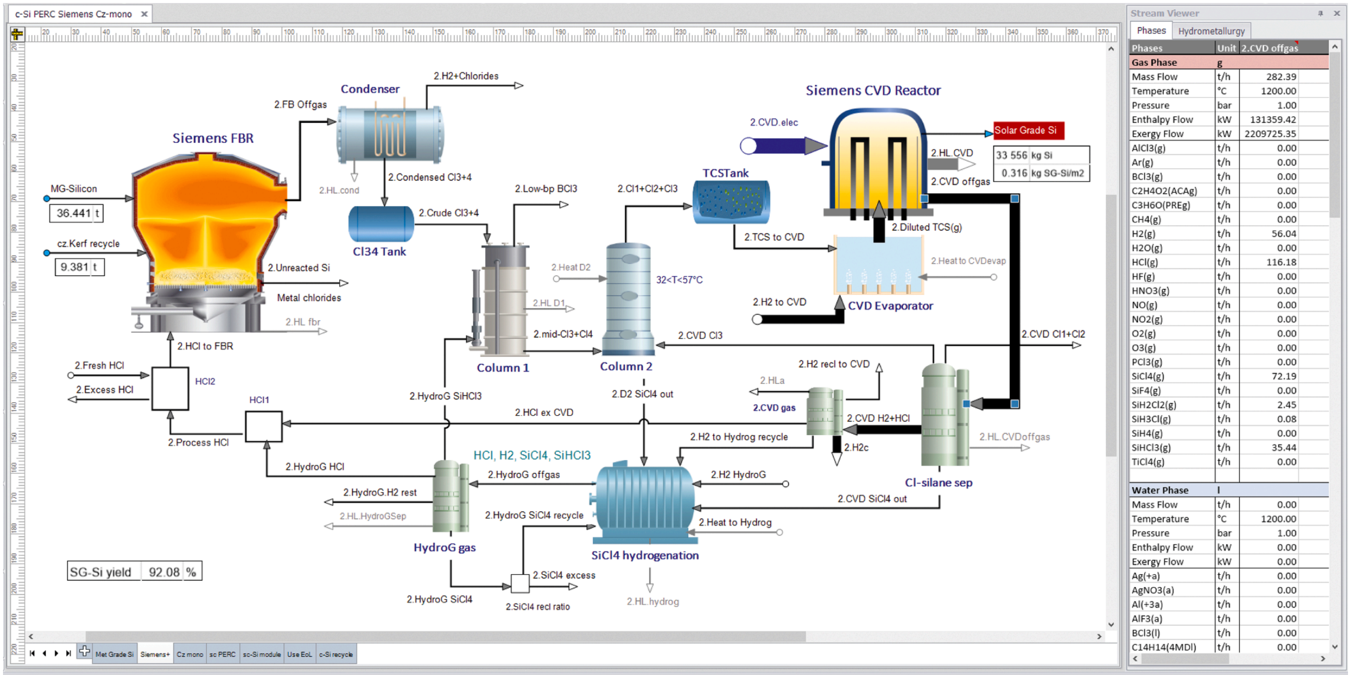

Fig. A2.2. Simulation flowsheet for solar grade silicon production via the Siemens process.

$$2\,SiHCl_3 = SiH_2Cl_2 + SiCl_4 \tag{13}$$

$$SiH_2Cl_2 = Si + 2HCl \tag{14}$$

$$SiHCl_3 + H_2 = Si + 3HCl \tag{15}$$

$$SiHCl_3 + HCl = SiCl_4 + H_2 \tag{16}$$

The offgas stream, therefore, contains $SiHCl_3$, SiH_2Cl_2, $SiCl_4$, HCl, and H_2, and we use Gibbs free energy minimisation to estimate the amounts of each exiting the CVD reactor. To improve process efficiency, STC is recycled by converting it into TCS by hydrogenation, according to reaction (17) (Seigneur et al., 2016).

$$SiCl_4 + 3H_2 = SiHCl_3 + 3HCl \tag{17}$$

As mentioned earlier, the process is energy intensive and inefficient with significant dissipative losses (Ceccaroli and Lohne, 2011). We assume an overall power consumption of 95 kWh/kg SG-Si produced but acknowledge that modern facilities achieve 71 kWh/kg, as mentioned in the Introduction. Depending on the case being investigated (refer to Table 1), kerf residue may re-enter the process here with MG-Si. All of the SG-Si produced is transferred to the next process for the production of mono-Si wafers.

In Section 3.4, a comparison is made with the alternative SG-Si production route mentioned earlier—the production of silane (SiH_4) from MG-Si and the subsequent production of granular SG-Si in a FBR. Silane is produced via the disproportionation of trichlorosilane ($SiHCl_3$), which involves the reaction of MG-Si with hydrogen (H_2) and silicon tetrachloride ($SiCl_4$) to produce $SiHCl_3$, and the subsequent catalytic redistribution of purified $SiHCl_3$ in fixed bed columns (Bye and Ceccaroli, 2014). In the silane FBR, seed particles of pure Si are fluidized in a preheated stream of SiH_4 and H_2. The SiH_4 decomposes unidirectionally into Si and H_2, and Si deposits on the seed particles, which grow until their weight causes them to fall out of the fluidized bed (Jiang et al., 2017). This process consumes 80–90% less electricity than the Siemens process and is a continuous rather than a batch process, which offers several advantages (Jiang et al., 2017) amongst which is significantly lower CO_2 (Scope 2) emissions. Detailed descriptions of these processes can be found in the cited references.

6.5. *Monocrystalline silicon ingots and wafers*

The Cz method is used to produce mono-Si ingots from which Si wafers are sawn. The process (depicted in Fig. A2.3) starts with the cleaning of Si feedstock and the removal of SiO_2 by etching in an acid bath consisting of a mixture of nitric (HNO_3), hydrofluoric (HF) and acetic (CH_3COOH) acids (Hirtz et al., 1992). The general etching reactions are:

$$3\,Si + 4\,HNO_3 = 3SiO_2 + 4\,NO + 2\,H_2O \tag{18}$$

$$SiO_2 + 6\,HF = H_2SiF_6 + 2H_2O \tag{19}$$

The role of acetic acid in the solution is as a diluent, oxidation promotor, and a wetting agent (Yifan et al., 2013). The quantities of reagents and products for this step are calculated using reactions (18) and (19) above and the HNA volumetric ratio of HF:HNO_3:CH_3COOH = 1:6:1 reported by Yifan et al. (2013). The offgas stream is cleaned in a scrubber where sodium hydroxide (NaOH) is used to remove NO_x, HF, HNO_3 and CH_3COOH (Jungbluth et al., 2010). The resulting effluent contains sodium nitrate ($NaNO_3$), sodium fluoride (NaF) and sodium acetate ($NaCH_3COO$) and is transferred to a water treatment unit. After a rinsing step in deionised (DI) water and drying with acetone (CH_3COCH_3), the Si feedstock proceeds to

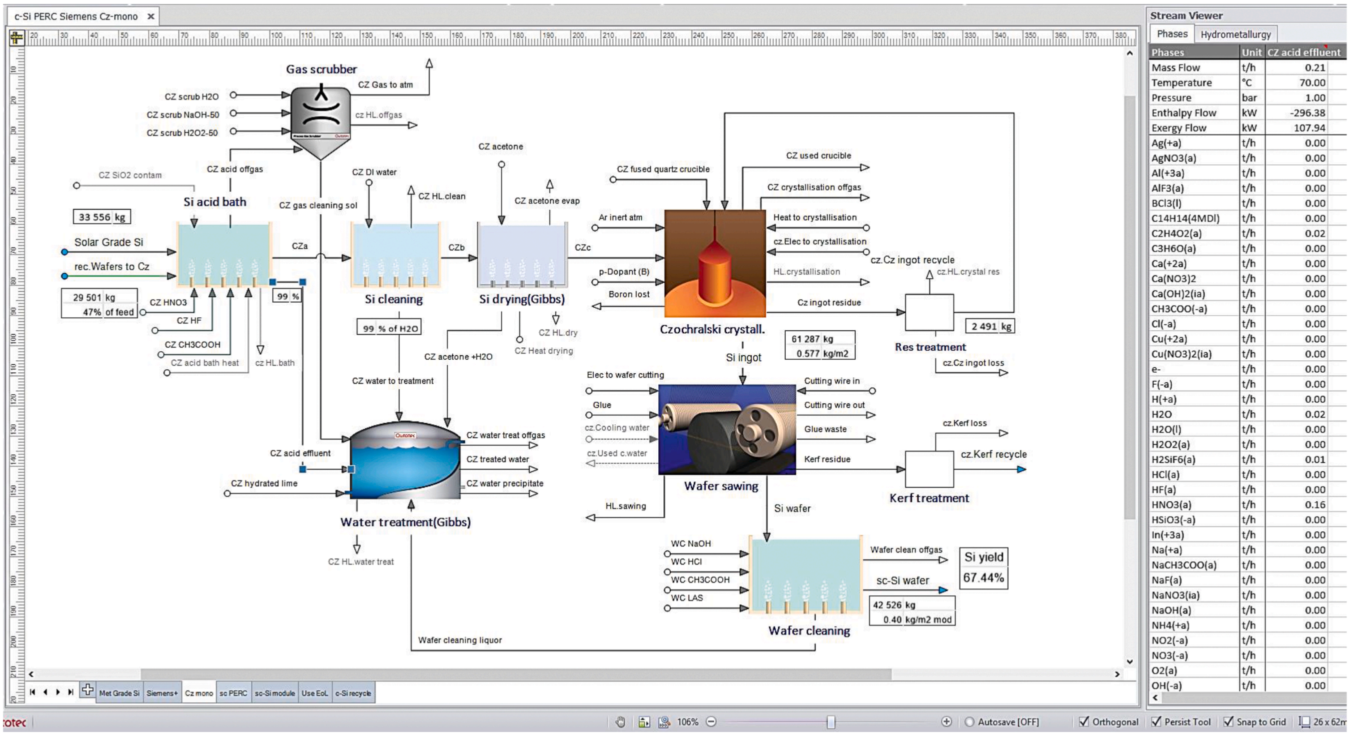

Fig. A2.3. Simulation flowsheet for Czochralski crystallisation and wafer cutting, showing some of the aqueous species tracked in the simulation, in the table on the right hand side (here showing some quantities for the acid bath effluent liquor).

the Cz crystallisation step.

Conventional Cz crystallisation of a mono-Si ingot entails melting the Si feedstock and controlled amounts of the required dopant (B in this case for a p-type ingot) in a quartz glass crucible, dipping a single crystal seed into the melt, and withdrawing it from the melt following carefully controlled speed and temperature profiles to start growing a cylindrical mono-Si ingot of the required diameter (Seigneur et al., 2016). Boron is added to achieve a dopant concentration of $1 \times 10^{16}/cm^3$ (Rodriguez et al., 2011), assuming a 50% loss of B during the process. The crucible is consumable and is lost together with some remaining solidified Si residue (Dold, 2015), while the susceptor can be reused for a number of cycles (Lan et al., 2009). The furnace chamber is flushed with argon (Ar) for the entire duration of the cycle to continuously remove SiO gas that forms as a result of contact between the melt and the quartz crucible, so preventing SiO_2 deposition in the furnace chamber (Dold, 2015) and lowering the risk of particulates falling into the melt, which could cause dislocations in the crystal, necessitating a restart of the growth process (Lan et al., 2009). Power consumption for the crystallisation process is taken as 33 kWh/kg crystal (VDMA, 2020).

Diamond wire sawing is used to cut wafers with a thickness of 170 μm (VDMA, 2020). The wire consumption rate is estimated using a wire performance of 250 cm^2/m (wafer area cut per metre of wire consumed) and a wire thickness of 120 μm (Peguiron et al., 2014). Kerf residue represents the largest and most expensive material inefficiency in the system with approximately 32% of mono-Si lost with a kerf thickness of 75 μm (VDMA, 2020). Recent European projects such as CABRISS (www.spire2030.eu/cabriss), amongst others, have demonstrated successful kerf recycling, achieving purities of up to 4 N, albeit with a requirement to manage a number of safety concerns (Halvorsen et al., 2017). Ongoing research projects like SELISI (selisi.eu) and collaboration with private companies aim to further this work, indicating that module and kerf recycling remain priorities from both industry and policy perspectives. ROSI Solar, for instance, has been one of the first to be awarded funding for their work in this field under the EU Green Deal (ROSI Solar, 2020). Furthermore, with Si metal listed as a critical metal and the forecast growth in the PV and other Si-consuming industries like electronics and batteries, future shortages of high purity metal may occur. For these reasons, kerf recycling is one of the main topics investigated in this paper.

After sawing, wafers are etched to remove surface damage and cleaned to remove any residues (Rodriguez et al., 2011). Reagent and energy consumptions for this step are estimated from Jungbluth et al. (2010) and Frischknecht et al. (2015). Finally, all acidic liquors from the crystallisation and wafering processes are neutralised in a water treatment unit using hydrated lime ($Ca(OH)_2$). All of the wafers produced in this process are transferred to the cell manufacturing process.

6.6. Cell manufacture

The PERC solar cell architecture (shown in Fig. A2.4a) uses an advanced silicon cell architecture, the key improvement being the integration of a back-surface passivation layer. This layer of material on the back of the cell that is able to improve its PCE to between 21% and 24%, compared to 18 to 19% for conventional aluminium back surface field (Al-BSF) cells (Blakers, 2019). The passivation layer increases the overall cell efficiency in three key ways: (i) it reduces rear-side recombination losses; (ii) it increases the absorption of light and (ii) it enables higher internal reflectivity (Allen et al., 2019; Blakers, 2019; Mandal et al., 2020). In line with current PV industry and market trends, the PERC cell design is used in our simulation. The process steps in our simulation follow, for the most part, those described by Werner et al. (2017) and are shown in Figs. A2.4b and A2.5. Based on industry trends (VDMA, 2020), we consider cells 166 mm in length and 166 mm in width (size M6). Indication are, however, that larger M12 (210 mm x 210 mm) cells may gain market share sooner than expected (e.g. PVTECH, 2020a, 2020b), in which case it would be relatively straightforward to modify wafer size and to rerun our simulations.

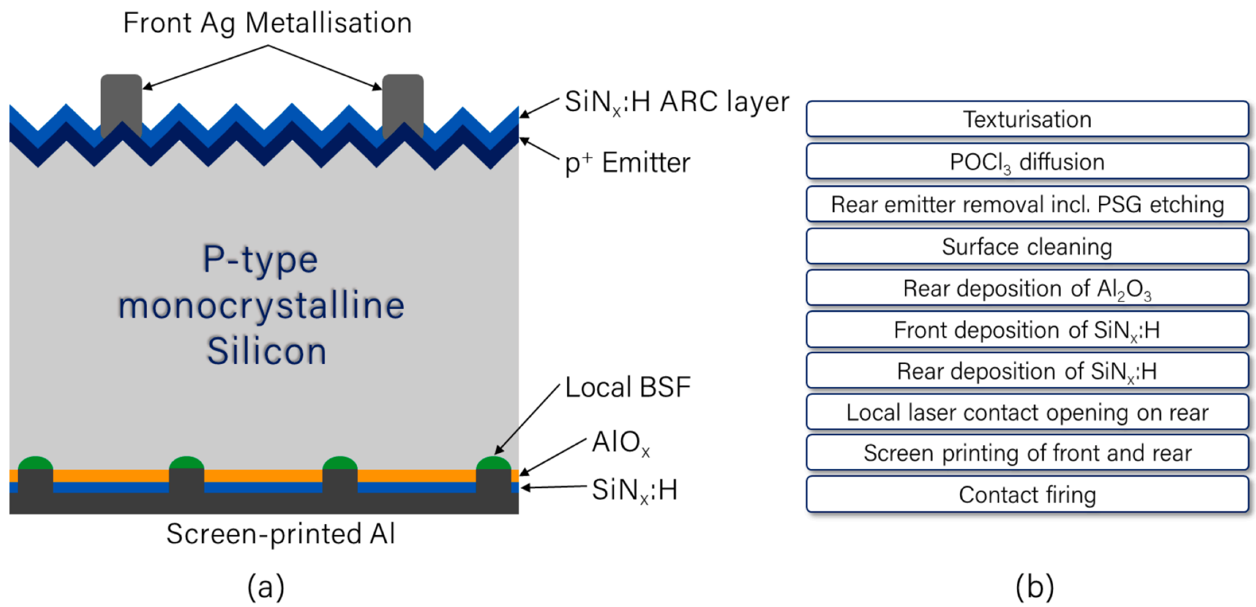

Fig. A2.4. (a) Passivated emitter rear contact (PERC) PV cell architecture, and (b) production steps.

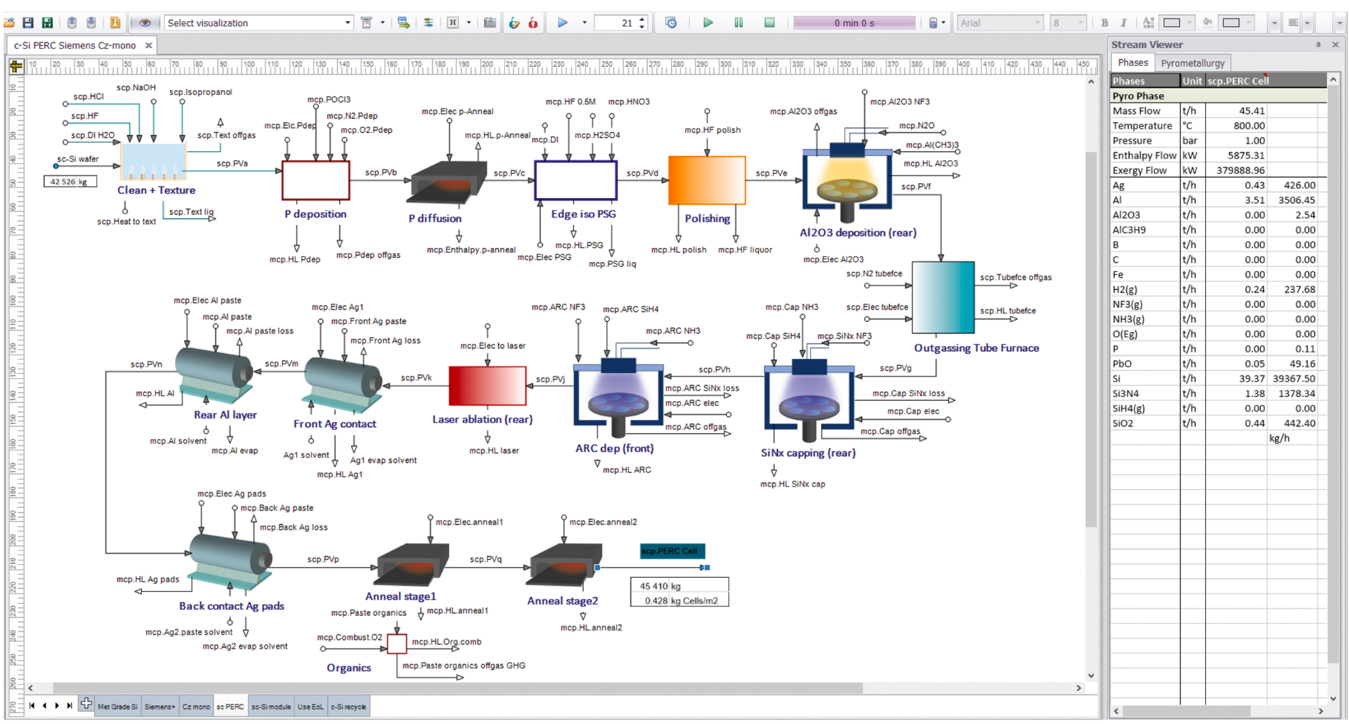

Fig. A2.5. Simulation flowsheet for PERC PV cell manufacturing showing mass, element, compounds, enthalpy, and exergy flows.

The process starts with the cleaning and texturing of the wafers produced in the previous step. Etching in HNO_3 and HF removes approximately 5 μm of Si on either side of the wafer, followed by a cold KOH etch to remove a thin layer of remaining porous Si. Further washing and etching steps (in HCl, HF and DI water) remove remaining residues (Hahn and Joos, 2014). The consumptions of HNO_3 and HF are calculated from the volume of Si to be etched and the etching reactions. Water consumption is taken as 33.4 L/m^2 wafer (Louwen et al., 2015) and other reagent consumptions are based on inventory data reported by Frischknecht et al. (2015). Energy requirements are determined by closing the energy balance. Products and effluents from this step include NO, H_2SiF_6, unused HCl, HNO_3, HF, KOH, isopropanol and used DI water.

During the next group of steps, the P-doped homogeneous emitter is deposited. The dopant precursor, phosphorous oxychloride ($POCl_3$), reacts with O_2 at 830 °C (Li et al., 2017) to form P_2O_5 and Cl_2. The O_2 oxidises the wafer surface, and the resultant SiO_2 - P_2O_5 combination forms a phosphor-silicate glass (PSG) layer, which then acts as the actual dopant source (Hahn and Joos, 2014). Relevant chemical reactions used in our simulation are:

$$4POCl_3 + 3O_2 = 2P_2O_5 + 6Cl_2 \quad (20)$$

$$2P_2O_5 + 5Si = 5SiO_2 + 4P \tag{21}$$

The deposited dopant is subsequently driven further into the silicon through annealing in the absence of $POCl_3$ (Li et al., 2017). We assume an active doping depth of 1 μm (Hahn and Joos, 2014) and an average P concentration of $10^{19}/cm^3$ in the active volume to calculate the $POCl_3$ consumption and PSG formation according to reactions 15 and 16. The PSG layer on the front surface is removed using dilute HF and the emitter deposited on the rear of the wafer in a wet etching process which involves the use of H_2O, HF, HNO_3, and H_2SO_4 (Hahn and Joos, 2014). A further polishing step takes place in a dilute (1% in H_2O) HF solution (Dingemans et al., 2010). Products and effluents from this step include $POCl_3$, O_2, N_2, P_2O_5, Cl_2, P, SiO_2, H_2SO_4, HNO_3, HF, and used DI water.

Rear passivation is achieved through PECVD of a 6 nm AlO_x (expressed as Al_2O_3) layer (Werner et al., 2017) with $Al(CH_3)_3$ as the precursor according to reaction 22 (Hofmann et al., 2013).

$$2Al(CH_3)_3 + 24\,N_2O = Al_2O_3 + 24N_2 + 6CO_2 + 9H_2O \tag{22}$$

After an outgassing step using N_2, the Al_2O_3 layer is capped with a 100 nm (minimum) layer of SiN_x (expressed as Si_3N_4), deposited by PECVD. Similarly, a 75 nm SiN_x layer deposited on the front surface serves as both a passivation and an anti-reflective layer (Werner et al., 2017). The precursors for the SiN_x layer are SiH_4 and NH_3, which form Si_3N_4 according to reaction 23.

$$3SiH_4 + 4NH_3 = Si_3N_4 + 12H_2 \tag{23}$$

PECVD deposition efficiency is assumed to be 25% and deposition chambers are cleaned using NF_3 gas (Louwen et al., 2015). The main products and effluents from the PECVD steps are Al_2O_3, $Al(CH_3)_3$, N_2O, NF_3, NH_3, SiH_4, Si_3N_4, N_2. Because the GWP of NF_3 gas is 16,100 times that of CO_2 (Greenhouse Gas Protocol, 2016), it is extracted from the offgas stream and does not contribute to Scope 1 emissions in our simulation.

The PERC-specific local contact openings are then created by laser ablation. Metallization of the front (busbar grid) and rear surface (Ag and Al, respectively), and the rear contact pads (Ag) are achieved by screen printing. Dry metallization pastes are assumed to contain 80 wt% of the relevant metal (Ag or Al), 10 wt% glass frit, and 10 wt% organic binders (Hahn and Joos, 2014) and pastes are assumed to contain 30 wt% solvent, which is taken to be butyl acetate ($C_6H_{12}O_2$) (Gong et al., 2015). Total paste consumptions (including losses during manufacturing) are calculated from VDMA (2020).

After drying and the evaporation of the solvents at 150–200 °C, curing takes place following a firing program that removes the organic binders (below 600 °C) and forms the Ag and Al contacts following a temperature profile up to 800 °C before cooling and recrystallization (Hahn and Joos, 2014). Typical power consumptions for wet etching, PECVD, screen printing and annealing are taken from Louwen et al. (2015).

6.7. Module assembly

PV module assembly commences with cleaning of the glass substrate, and the preparation of fluxed Sn-coated Cu ribbons, which are used to string sets of 10 to 12 cells together by soldering (Wirth, 2013). After placing the first EVA encapsulant layer with a thickness of 450 μm (VDMA, 2020) on the glass substrate, 5 or 6 cell strings are placed and soldered together in series. Another encapsulant layer is then placed, followed by the back sheet, which consists of a layer of PET (250 μm) sandwiched between two layers of PVF (40 μm each), the so-called TPT configuration (Blieske and Stollwerck, 2013; Frischknecht et al., 2015). The completed layup is then laminated under vacuum at 150 °C. After cooling, module edges are trimmed and sealed using PVB, the module is framed with an aluminium alloy ($AlMg_3$) frame, and the junction box is attached (Wirth, 2013). As a final step, the module is tested. The process is depicted in Fig. A2.6.

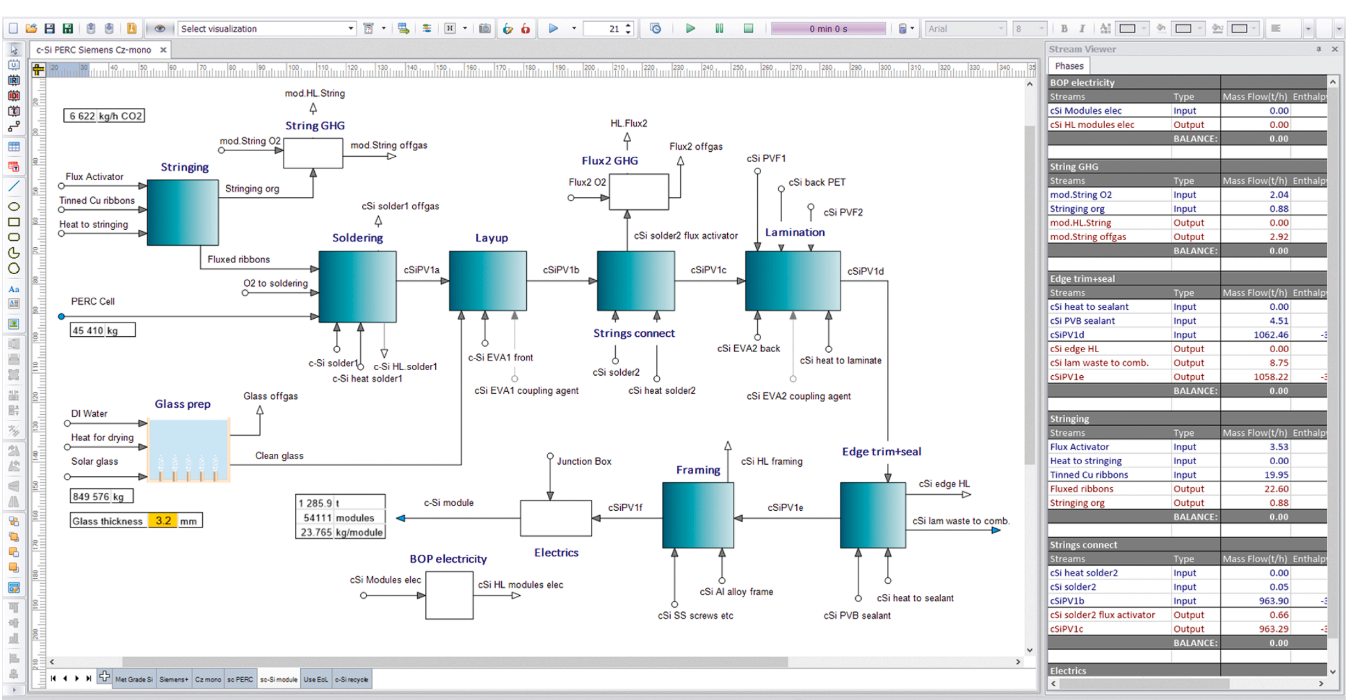

Fig. A2.6. Simulation flowsheet for PV module assembly steps.

We use the following additional parameters based on the cited references:

- Module dimensions are 1981 mm x 991 mm (energysage, 2020).
- Each module contains 72 cells (Frischknecht et al., 2015; VDMA, 2020).
- Glass thickness is 3.2 mm (Frischknecht et al., 2015; VDMA, (2020) specifies >3 mm).
- Solder type Sn63 (63 wt% Sn, 37 wt% Pb) is used throughout, and its consumption is deduced from the aggregated data provided by Frischknecht et al. (2015).
- The soldering flux activator and solvent are assumed to be adipic acid and isopropanol, respectively (Wirth, 2013), and are included for the purposes of estimating potential environmental impacts.
- Total power consumption is calculated using the specific consumption (3.73 kWh/m^2 module) reported by Frischknecht et al. (2015).

6.8. End-of-life module recycling

Under the European Waste Electrical and Electronic Equipment (WEEE) Directive (2012/19/EU), at least 85% of the PV modules put on the market shall be collected, and at least 80% “prepared for reuse and recycled” (European Union, 2012). Other than landfilling, dominant recycling approaches for c-Si PV modules involve their treatment in general recycling facilities with other waste electronic goods or laminated glass (Duflou et al., 2018). With these, the mass-based targets of the directive can be met without having to pay considerable attention to the quality of recyclates and as a consequence, components like Ag, Cu, Si, and Pb are lost and their potential environmental impacts not mitigated (Heath et al., 2020). More sophisticated, PV-dedicated upcycling processes generally consist of two main steps: (i) module delamination, achieved through mechanical, thermal, or chemical methods, and (ii) cell recycling, consisting of wafer/metal separation to recycle Si, and the subsequent extraction of metals (Deng et al., 2019). Heath et al. (2020) identify the FRELP[1] (Latunussa et al., 2016; Ardente et al., 2019) and ASU[2] (Huang et al., 2017) processes to have the potential to advance PV recycling. Despite not recovering the whole suite of minor elements in the c-Si system, these aim to integrate the delamination and cell recycling steps and achieve high recoveries of Ag, Cu, Al, Si, glass, and insulated cable (Heath et al., 2020).

For this study, the recycling simulation (depicted in Fig. A2.7) consists of dismantling, thermal de-encapsulation and glass separation, polymer combustion, string dissolution and metal recovery, wafer etching, and neutralisation of the leach liquors. Dismantling involves the removal of the junction box and frame and the thermal processing step involves the separation of the glass, backsheet, and polymer layers from the cell strings. Glass is assumed to be recovered intact, but is not recycled directly into new modules in the current simulation. The metal and wafer recovery steps follow that proposed by Huang et al. (2017), i.e. the ASU process. It was selected because of its ability to recover Si at SG-Si quality and its self-limiting

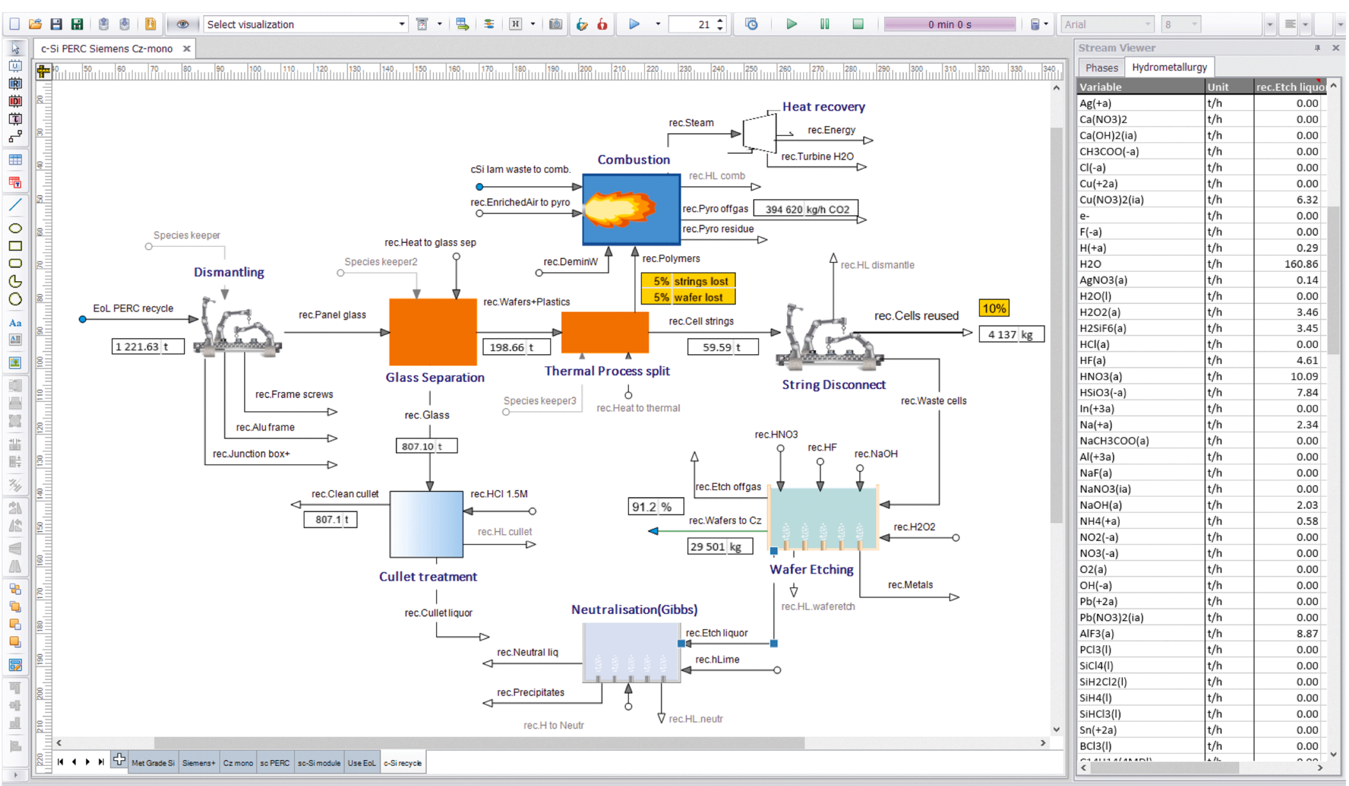

Fig. A2.7. Simulation flowsheet for PV module EOL recycling (wafer etching liquor composition shown on the right).

[1] FRELP – Full Recovery End of Life Photovoltaic process, developed by SASIL (https://frelp.info)

[2] ASU – Arizona State University (Huang et al. 2017)

chemistry for metal extraction, which allows for more control over the process (Huang et al., 2017). The authors report recoveries of 74% and 83% at purities of more than 99% for Ag and Cu, respectively, and the removal of more than 99% of Pb^{2+} from solution. Furthermore, 85–90% of Si can be recovered as SG-Si. In a previous study (Bartie et al., 2020), we modelled a PV-dedicated recycling process for CdTe PV modules based on First Solar's processing route, which also consists of delamination, glass recovery, and metal recovery steps (Wade, 2014).

Complete combustion of the polymers (EVA, PET, PVF, and PVB) and a 5% loss of cells and strings are assumed to occur in this step. Polymer combustion is used to estimate direct CO_2 generation (i.e. Scope 1 emission) according to the following chemical reactions:

$$\text{EVA} \quad C_2H_4 + 3O_2 = 2CO_2 + 2H_2O \tag{24}$$

$$2C_4H_6O_2 + 9O_2 = 8CO_2 + 6H_2O \tag{25}$$

$$\text{PVB} \quad C_8H_{14}O + 11O_2 = 8CO_2 + 7H_2O \tag{26}$$

$$\text{PET} \quad C_{10}H_8O_4 + 10O_2 = 10CO_2 + 4H_2O \tag{27}$$

$$\text{PVF} \quad 2C_2H_3F + 5O_2 = 4CO_2 + 2H_2O + 2HF \tag{28}$$

The assumption of complete combustion is conservative in terms of GHG emissions as it likely overestimates the quantity of CO_2 generated. Hydrogen fluoride (HF) formed during the combustion of PVF is absorbed in a scrubber for neutralisation. Heat generated during this combustion process is partially recovered to generate electricity.

From string dismantling, it is assumed that 10% of cells are recovered intact for direct reuse. The remaining cells strings proceed to the leaching and etching steps to recover metals and wafer Si for reuse. Firstly, leaching in HNO_3 dissolves only Ag, Pb, and Cu, and precipitates Sn as SnO_2 i.e. the Sn/Pb solder, Sn-coated Cu ribbons, and the Ag contacts. Huang et al. (2017) then recover Ag, Cu in sequential electrowinning (EW) steps with Pb preciptating as hydrated PbO_2 during Cu recovery (see also Mecucci and Scott, 2002), while the SnO_2 precipitate is recovered by filtration. The EW steps are not included in the simulation, but their reported metal recoveries are taken into account in overall material efficiency calculations. Next, HF is used to dissolve only the SiN_x and Al-containing layers from the cells. The emitter and BSF are etched away in NaOH, a process that needs careful control as it is not self limiting (Huang et al., 2017). The leaching steps are simulated using the following reactions:

$$3Ag + 4HNO_3 = 3AgNO_3 + 2H_2O + NO \tag{29}$$

$$Pb + 4HNO_3 = Pb(NO_3)_2 + 2NO_2 + H_2O \tag{30}$$

$$Cu + 4HNO_3 = Cu(NO_3)_2 + 2NO_2 + H_2O \tag{31}$$

$$Sn + 4HNO_3 = SnO_2 + 4NO_2 + 2H_2O \tag{32}$$

$$2Al + 6HF = 2AlF_3 + 3H_2 \tag{33}$$

$$Si_3N_4 + 18HF + 4H^+ = 3H_2SiF_6 + NH_4^+ \tag{34}$$

$$Si + NaOH + 2H_2O = NaSiO_3 + H_2 \tag{35}$$

A stream of clean wafers is produced and recycled to the Cz crystallisation process as SG-Si feedstock. The remaining acidic liquor is neutralised with hydrated lime ($Ca(OH)_2$) to form calcium fluoride (CaF_2). Gibbs free energy minimization is used in this unit operation to estimate the compositions of the remaining neutral liquor and precipitates.

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Annexure C: Article 3

Metallurgical infrastructure and technology criticality: the link between photovoltaics, sustainability, and the metals industry

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Metallurgical infrastructure and technology criticality: the link between photovoltaics, sustainability, and the metals industry

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ORIGINAL PAPER

Metallurgical infrastructure and technology criticality: the link between photovoltaics, sustainability, and the metals industry

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Abstract
Various high-purity metals endow renewable energy technologies with specific functionalities. These become heavily intertwined in products, complicating end-of-life treatment. To counteract downcycling and resource depletion, maximising both quantities and qualities of materials recovered during production and recycling processes should be prioritised in the pursuit of sustainable circular economy. To do this well requires metallurgical infrastructure systems that maximise resource efficiency.To illustrate the concept, digital twins of two photovoltaic (PV) module technologies were created using process simulation. The models comprise integrated metallurgical systems that produce, among others, cadmium, tellurium, zinc, copper, and silicon, all of which are required for PV modules. System-wide resource efficiency, environmental impacts, and technoeconomic performance were assessed using exergy analysis, life cycle assessment, and cost models, respectively. High-detail simulation of complete life cycles allows for the system-wide effects of various production, recycling, and residue exchange scenarios to be evaluated to maximise overall sustainability and simplify the distribution of impacts in multiple-output production systems. This paper expands on previous studies and demonstrates the key importance of metallurgy in achieving Circular Economy, not only by means of reactors, but via systems and complete supply chains—not only the criticality of elements, but also the criticality of available metallurgical processing and other infrastructure in the supply chain should be addressed. The important role of energy grid compositions, and the resulting location-based variations in supply chain footprints, in maximising energy output per unit of embodied carbon footprint for complete systems is highlighted.

Keywords Circular economy · Sustainability · Process simulation · CdTe and Silicon photovoltaics · Life cycle assessment (LCA) · Technoeconomics

Introduction

Metals, their production, and system interactions

Technologies that enable the harnessing of renewable energy contain various metals that facilitate specific functionalities. These include a number of precious and special metals/metalloids, often in miniscule—but essential—quantities. Most of the minor metals are only produced as co-products of their carrier metals (Bleiwas 2010), thereby drawing all associated carrier metal value chains, including the associated infrastructure, into the life cycle. As demand for some of these metals is on the increase (UNEP 2013), several of them have been classified as *critical raw materials* (CRMs) because of the combination of their economic importance, supply disruption risk, and the resource intensity associated with their extraction from lower-grade deposits (Frenzel et al. 2017; Nassar and Fortier 2021). Not all critical materials are necessarily scarce—in some cases, supply risk can be alleviated by having and keeping the right infrastructure in the right locations. Without the associated critical infrastructure, however, critical materials cannot be produced.

Manufacturing processes cause pure metals and other materials to become heavily intertwined, the degree to

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which directly impacts the effectiveness of recycling—in terms of quantity and quality—and thus circularity potential. Upon entering the end-of-life stage, devices collected for recycling become complex urban minerals in which no elements are present in pure form. As with the extraction of valuable metals from primary geological minerals, urban mineral beneficiation and extraction processes need to be designed to run at their thermodynamic limits, so optimising resource efficiency and consumption. This complexity has been visualised in the Metal Wheel, which can be found in various publications, e.g. Reuter et al. (2019) and Verhoef et al. (2004). Negative impacts on sustainability, i.e. the environmental, economy, and society, need to be minimised at the same time, while still increasing economic welfare for the stakeholders operating in the life cycle. Before such optimisation can be performed, all material and energy flows (losses and entropy creation included), and potential environmental, economic, and social impacts need to be quantified for the entire system (Reuter et al. 2019). Of particular importance is also the location of infrastructure in the supply chain, as it strongly affects sustainability—environmental impacts related to power consumption can change dramatically depending on the combination of fossil- and non-fossil energy sources in the local electricity grid. Production and recycling costs and social impacts can also change significantly between locations. With this information in hand, the potentially many trade-offs in this complex optimisation problem can be identified and quantified.

Good separation of the multitude of intertwined materials, compounds, alloys, and others usually cannot be achieved in only one reactor, but rather in a system of reactors (i.e. a plant) or a system of plants and processes which then form part of the circular economy. It is self-evident that the production and recycling of metals are not only about the technology but how best to manipulate the exchange of materials between different phases in individual reactors, between different reactors, and between systems of reactors (Reuter 2016; Reuter et al. 2021). To achieve this, mass and heat transfer between different phases in a reactor, process kinetics and dynamics, chemistry, and thermodynamics need to be understood well. Furthermore, it is about the exchange of information between life cycle stages, e.g. making available recycling data to facilitate design-for-recycling, and financial exchanges to keep the life cycle going. Two PV technologies, cadmium-telluride (CdTe) and monocrystalline Si (mono-Si) PV, will be presented to illustrate these concepts. This paper demonstrates the benefits of simulating these large life cycle systems at the process level, and linking simulations with environmental assessment and cost models, and provides insights on their sustainability and circular economy potential. For the results presented here, the authors expanded the methodologies presented in previous publications, in which the resource consumption (RC), resource efficiency (RE), and environmental impacts of the CdTe (Bartie et al. 2020) and mono-Si (Bartie et al. 2021a) PV module life cycles were assessed.

The role of metals in photovoltaics

Dominant PV technologies include first-generation wafer-based crystalline silicon (c-Si) and second-generation CdTe and copper-indium-gallium-diselenide (CIGS) thin-film cells in which several semiconductor and other metal layers are deposited onto sub- or superstrates and laminated into modules with another layer of glass polymer sheet as backing. Between these three technologies, the metals and other elements needed include Ag, Al, Au, B, Cd, Cu, Ga, In, Mg, Mo, Ni, P, Pb, Se, Si, Sn, Te, Ti, Zn, and others in various combinations. Between the CRM lists of the USA and Europe, more than half of these are considered critical (European Commission 2020; Graedel et al. 2022). Significant research and development are also underway to further develop tandem modules, which consist of combinations of these and new third-generation technologies that utilise, e.g. Pb-based perovskites to maximise power conversion efficiency (PCE) (Lal et al. 2017; Mohammad Bagher et al. 2015). As the production of tandem modules effectively combines the life cycles of two different PV technologies, the resulting step changes in PCE come with increased life cycle complexity, resource requirements, losses, and impacts. Therefore, it is important to analyse PV life cycles at a detail level that enables the identification and optimisation of relevant sustainability and CE-related hotspots and trade-offs. This is important for the PV industry, to be able to assess resource and recycling requirements in the long run, and to optimise and strategise accordingly (e.g. Haegel et al. 2019).

Process simulation provides a platform for creating digital twins of systems of linked value chains and allows one to capture the detail level often missing from approaches that use aggregated input data (Jacquemin et al. 2012; Reuter 2016; Reuter et al. 2015) and often consider quantity, but not quality (Reuter et al. 2019). The work discussed in this paper has been realised using the HSC Chemistry (Metso:Outotec 2021) process simulation software. The complete supply chains have been mapped to consider all the complex non-linear combinations and chemistries in a large number of different reactors and systems. Following this approach permits calculating both energy and exergy flows (enthalpy and entropy of all streams) and linking them to power using a common unit, i.e. kW. Figure 1 shows the typical stages in a circular life cycle of a PV module with materials and energy also entering from outside the system to keep it functioning. The energy balance is represented by the orange ring, and the inevitable dissipation of exergy along the life cycle by the changing thickness of the red ring. Running the life cycle at its thermodynamic limits would minimise the thinning of this ring and reduce the amounts of external energy and

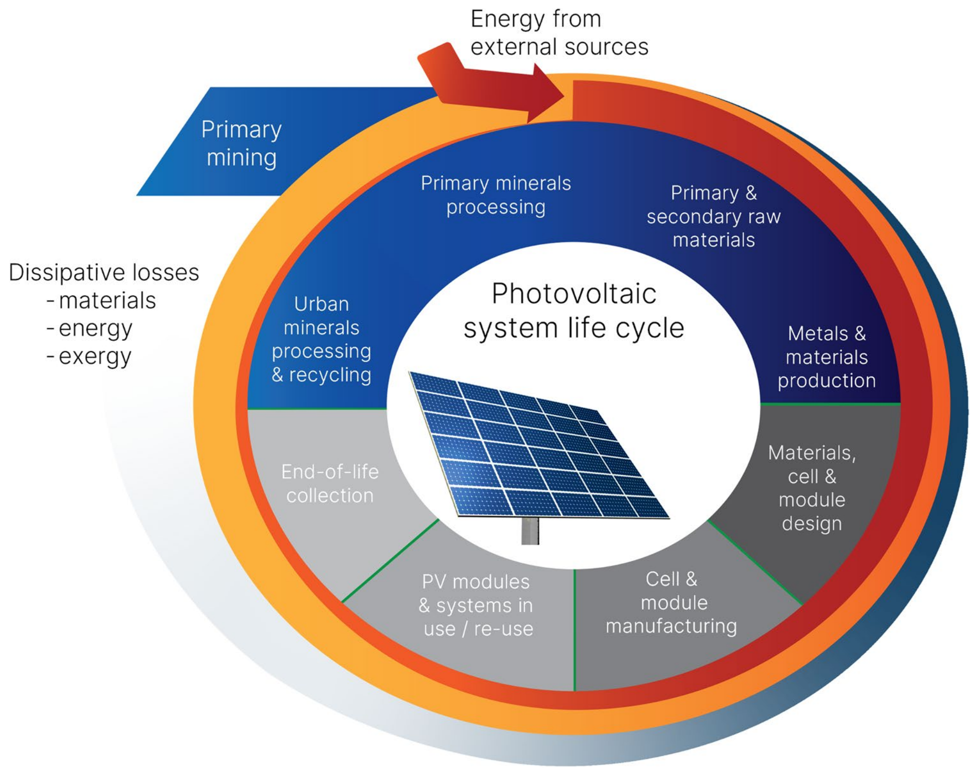


Fig. 1 PV module life cycle stages showing the consumption of external materials and energy, and their dissipation along the life cycle

materials that need to enter the system to slow down losses and keep value within the life cycle. Note that the resulting environmental and other impacts are strongly influenced by the location of each part of the complete CE infrastructure. This is discussed in Sects. 3.2.2 and 3.4.

High-detail simulations such as these facilitate simultaneous assessments of RC, RE, and potential environmental, social, and economic impacts using up-to-date operational data for existing technologies and relatively straightforward adaptation to create alternative datasets for the exploration of future scenarios. By linking these dimensions, the approach lends itself to design for sustainability and design for circularity by complementing design activities with relevant resource and sustainability information from the outset.

The Cd-Te PV system

The life cycle of a CdTe PV module is presented here as the first example of a system that encompasses several base and minor metal production chains. To manufacture a state-of-the-art CdTe PV cell, the metals needed include, among others, Al, Cd, Cu, Se, Sn, Te, and Zn. In this system, Cd is a co-product of Zn and Pb, while Te and Se are co-products of Cu (Bartie and Reuter 2021). A schematic diagram of the CdTe module life cycle is shown in Fig. 2. A thin-film PV module typically consists of a glass substrate onto which several semiconductor, metal contact, and other layers are deposited. Apart from the encapsulant and back glass, all layers contain metals (First Solar 2020a, b).

As mentioned, Cd, Te, and Se are mainly produced as by-products, and therefore, the life cycle includes the production of their carrier metals, and not just finished semiconductors. By expanding the foreground system boundaries in this way, resource, environmental, and other hot spots can also be identified within individual processes in metal value chains, opening up more opportunities for improvement in sustainability throughout the value chain. Recycling closes the life cycle loop, albeit only partially due to material and energy inefficiencies and losses, and the creation of entropy.

The mono-Si PV system

Silicon PV technologies dominate the PV market and also rely on the availability of various materials. Despite its abundance in the earth's crust, Si metal itself is considered a critical raw material because of its economic importance and potentially increased supply risk (European Commission 2020). Continued research and development over the last decades have resulted in higher cell and module efficiencies and considerable decreases in the amount of solar grade Si (SG-Si) required to generate a Watt of power, reaching 3.6 g/W_{DC} in 2019, half of the consumption a decade prior (IEA-PVPS 2019). At the same time, however,

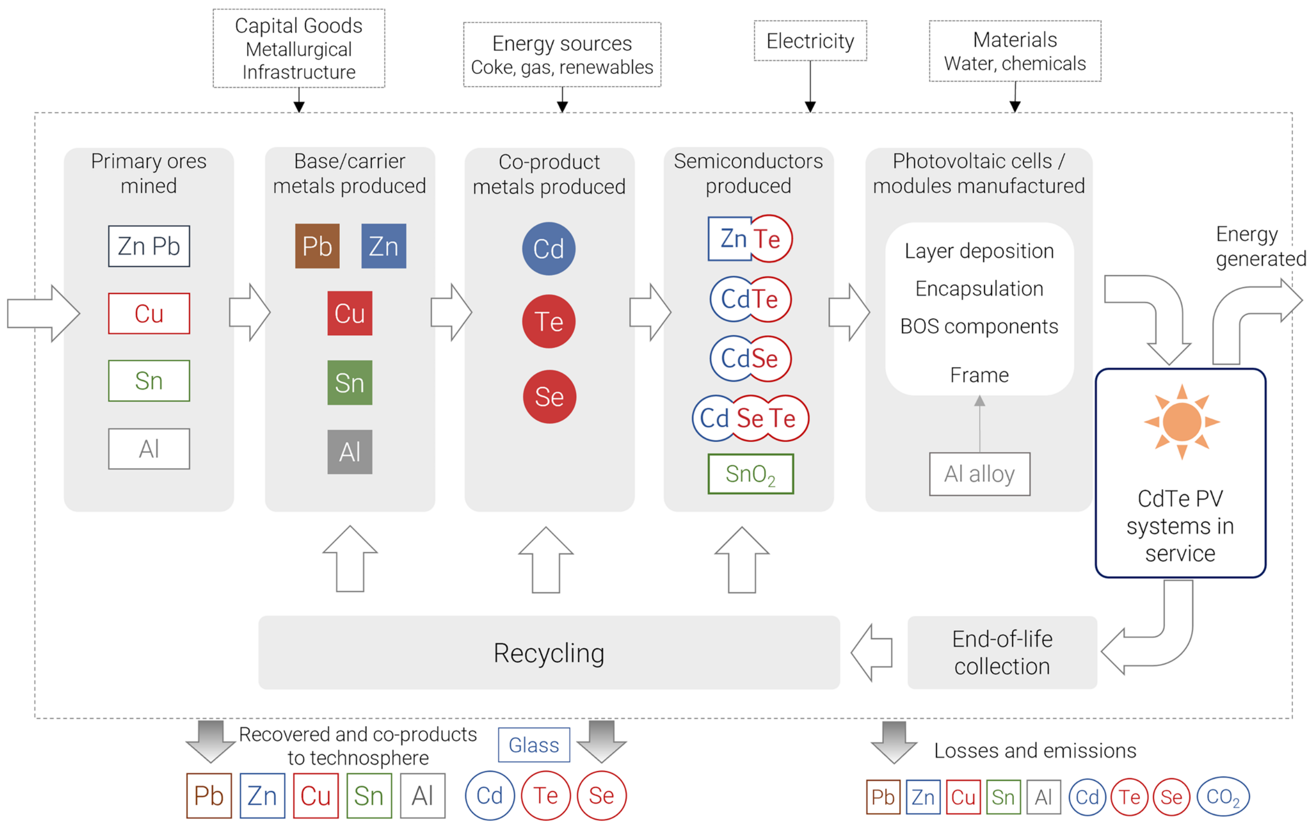


Fig. 2 Physical flows in the CdTe PV module life cycle (Bartie and Reuter 2021)

PV deployment is growing rapidly, the net effect being an increase in demand from 33 kt in 2015 to 235 kt in 2030 in the European PV industry (Gislev and Grohol 2018).

Opportunities are emerging to compensate, at least partially, for the net growth in demand from secondary resources. End-of-life (EOL) PV module quantities are expected to increase significantly between 2025 and 2030 with cumulative global PV waste quantities forecast to reach eight million tons by 2030 and almost 80 million tonnes by 2050 (IRENA and IEA-PVPS 2016). It is clear that design for circularity and sustainability and the development of complementary business models and high-quality recycling processes need to be prioritised.

The objective of recycling is to maximise RE by closing material loops. What cannot be ignored is that losses occur at every step along the way, meaning that these loops can only ever approach closure up to limits determined by life cycle design and the laws of nature. These losses cannot be designed out if they are not identified in the first place. Therefore, while EOL recycling is critically important, losses and inefficiencies need to be identified and minimised throughout the life cycle, in manufacturing processes as well as during product design. A block flow diagram of the mono-Si system analysed is shown in Fig. 3.

Methods and approach

Digital twinning of the systems

To create a steady-state process model of the entire CdTe life cycle system shown in Fig. 2, Cu production was modelled as a sulphide flash smelting operation with electric furnace slag cleaning, Peirce Smith converting, and anode furnace refining followed by electrolytic refining to Cu cathode. Te and Se are produced through further treatment of anode slimes, and all Cu scrap is recycled internally. A combination of conventional roast-leach-electrowinning (RLE) and direct Zn smelting was used for Zn production, with Cd recovered from residues generated during RLE purification stages. Lead production was modelled as a direct smelting process with bullion refining through conventional Cu and sulphur drossing, the removal of As, Sn and Sb (and the recovery of Te from recycled semiconductor material) in the Harris process, desilvering in the Parkes process, and Zn removal by vacuum distillation.

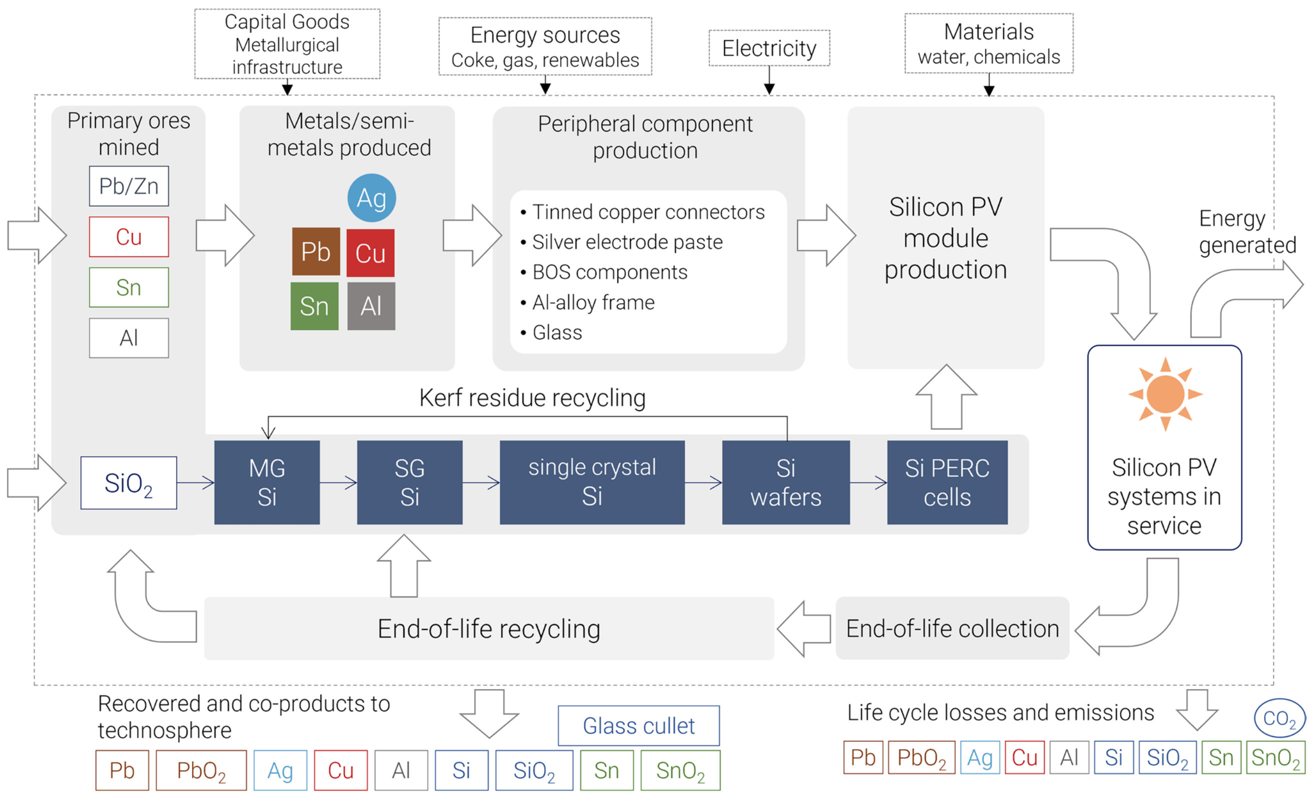


Fig. 3 Physical flows in the mono-Si PERC PV module life cycle system where PERC refers to passivated emitter and rear cell, the current industry-standard Si cell configuration (VDMA 2021)

The manufacturing and recycling of CdTe solar cells were modelled using process descriptions and data from published literature (e.g. Fthenakis 2004; Sinha et al. 2012; Wade 2013) and product specifications from the largest producer of CdTe PV technology (First Solar 2020b). Processes were linked through the exchange of compatible residues, so creating a closed loop integrated metallurgical production system that aims to minimise untreated residues and waste, and to maximise the quantities and qualities of products. Detailed descriptions of this model and all included process flowsheets can be found in previous publications (see Abadías-Llamas et al. 2019, 2020; Bartie et al. 2020). Models for the production of Pb, Zn, and Cd, and the production and recycling of CdTe PV modules are available online (Heibeck et al. 2020; Bartie and Heibeck 2020).

The same approach was used to create a detailed digital depiction of the mono-Si PV life cycle system shown in Fig. 3 to assess its RE, carbon footprint, and technoeconomic performance. As with the CdTe system, the process units in each of the process blocks were modelled separately and connected to create a model for that process. The processes were then connected to create the life cycle system, and material loops closed as far as possible by means of recycling. In this simulation, metallurgical grade Si (MG-Si) is produced via the carbothermic reduction in quartz with ladle refining for impurity removal. Solar grade Si (SG-Si) is produced with the Siemens process and monocrystalline Si (mono-Si) with the Czochralski method. Diamond wire sawing is used for the cutting of Si wafers, which then proceed to PV cell production. It is assumed that the residue that forms during wire sawing, the so-called kerf residue, is recycled to the SG-Si production process as MG-Si. For the recovery of metals and SG-Si from used wafers, a process developed by Huang et al. (2017) was simulated.

The simulation models capture the complexity of the processes and systems by considering relevant physical relationships, chemical reactions, thermodynamics, and process constants that define their response to changes in inputs or other process parameters. Results were compared with published operating points and known industrial reality for validation. Using this approach, the dependence on aggregated models for the foreground system is significantly reduced.

The Si PV simulation was expanded to include capabilities beyond that of the CdTe system simulation. These included the ability to predict the life cycle system's response to changes in wafer thickness and two recycling rates—kerf residue as MG-Si and EOL wafers as SG-Si. By additionally linking simulation results and a bottom-up cost model, the effects of recycling and a potential carbon tax on the PV module minimum sustainable price (MSP) and levelised cost of energy (LCOE) were estimated.

To improve computational efficiency for carrying out parameter studies, which, in this case, involved the simultaneous variation in three independent variables (wafer thickness and the two recycling rates), neural networks were created as surrogate functions that represent the process simulation output. Neural networks allow for generalised nonlinear process modelling without the need to predefine regression equations (Reuter et al. 1992). To achieve this, the simulation model was run several thousand times using uniformly distributed random combinations of the three independent variables as inputs. Subsequently, a large dataset containing the corresponding simulation results for all relevant dependent variables was created. MATLAB's neural network tool was used to generate the code that initiated, trained, tested, and validated the networks using this dataset. The neural networks then allowed for the individual and simultaneous effects of EOL and kerf recycling rates, and wafer thickness throughout the system to be evaluated in a fraction of the time it would have required using only the simulation model.

Resource consumption and efficiency

The production and recycling of metals and PV modules come at a cost. Resource throughput and efficiency are often quantified separately for material and energy streams using the laws of conservation. These indicators are important but do not capture changes that may have occurred in the utility of these streams (Gößling-Reisemann 2008). Applying the second law of thermodynamics using exergy analysis provides a way to track resource quality and its inevitable degradation along life cycles. While mass and energy always balance, exergy does not and this imbalance (i.e. irreversibility) over any process represents its degradation of the thermodynamic quality of materials and energy combined, due to the creation of entropy (refer to Fig. 1). In the approach presented here, exergy cost—the cumulative irreversibility associated with the product of interest (Szargut 2007)—serves as a proxy for resource consumption. Exergy efficiency is taken to represent the thermodynamic resource efficiency of a process. As mentioned, the advantage of exergy as an indicator is that material and energy streams are combined, and all expressed in units of measure for energy (Ayres 1998).

Environmental impact

Life Cycle Assessment (LCA) is the standardised and well-established method for assessing environmental impact (ISO 2006; ILCD 2010, 2011) and was used to estimate Global warming potential (GWP) and Acidification potential (AP) for the CdTe system using GaBi LCA software (sphera 2020). Total carbon emissions for the mono-Si system were determined as the sum of CO_2 generated directly in each process step (Scope 1 emissions) that associated with power consumption (Scope 2 emissions), and published values for the glass, Al alloy frames, mountings, cables, and connectors (Scope 3 emissions) (de Wild-Scholten 2013; Frischknecht et al. 2015). Direct CO_2 emissions remain constant for a given production configuration, but Scope 2 and 3 emissions depend on the composition of the energy mix at the production location.

In multi-product systems such as the carrier/co-product systems described earlier, substitution cannot be applied to avoid having to allocate overall impacts to products (ISO 2006), as many of the co-products cannot be produced in alternative, standalone processes that can be used to estimate their individual impacts. According to the standard, allocation should be based on physical relationships as far as possible. In the metals industry, however, economic allocation is applied frequently. It is a hotly debated topic (e.g. Finnveden et al. 2009; Heijungs et al. 2021), especially when there are large differences between the products' economic values (Valero et al. 2015). For comparison and to highlight some of the challenges, the distribution of impacts between products in the CdTe system was calculated using mass, exergy cost, exergy content, and economic value as allocation factors.

Technoeconomic assessment (carbon pricing, MSP, and LCOE)

Technoeconomic assessments were carried out for the mono-Si system to investigate the combined effects of circularity and a hypothetical carbon tax on module cost and LCOE. A bottom-up cost model, with assumptions partly adopted from Liu et al. (2020) and Sofia et al. (2019), was utilised to estimate the effects of recycling rates, wafer thickness, and carbon taxation on MSP and LCOE. MSP, the minimum module price at which manufacturers can meet investment return expectations, was calculated using discounted cash flow analysis—the MSP is iteratively calculated as the price at which the sum of the present values of all projected future cash flows breaks even with the initial investment, i.e. the price at which the so-called net present value (NPV) is zero (Zweifel et al. 2017). LCOE, the ratio of total energy generated and total cost, was calculated for a system lifetime of 30 years, a power conversion efficiency of 21.7% (Sofia et al. 2019), an average annual irradiation of 1,500 kWh/(m^2.year), and an annual degradation rate of 0.5%. To create a link between the process simulation and the cost model, MG-Si and SG-Si prices were updated based on recycling rates under the assumption that the prices of secondary Si (SG-Si from EOL wafers and MG-Si from recycled kerf) are two thirds of that produced from primary raw materials. Life cycle carbon taxes of 50 and 100 \$/tonne CO_2-equivalent (\$/tCO_2e) were considered.

Results and discussion

Resource consumption and efficiency

Resource flows in carrier/co-product metal systems for CdTe raw material production

The CdTe system is a good example of a system in which the key raw materials are produced as by-products of other systems. Approximately 40% of Te produced in the world today is used in CdTe PV applications (USGS 2021). To produce the Te and Cd, the prior production of Cu, Zn, and Pb cannot be avoided. The Cu system is required for Te production and the Zn system for Cd production. The Zn and Pb systems are linked, and the Pb system is also needed for the recycling of Te. The quantities and overall recoveries for relevant metals produced in the system to manufacture one CdTe module, as predicted using the simulation model, are shown in Table 1.

Table 1 shows that, to produce the CdTe needed for one PV module, also taking into account Cd and Te needed for non-PV uses, the system represented by this simulation would automatically also produce 104 kg Cu, 74 kg of Zn, and 35 kg of Pb due to the interconnectedness of the metal production systems. Without production infrastructure for the carrier metals, it would not be possible to bring PV modules to market. With the strong forecast growth in PV deployment (IRENA 2019), carrier- and co-metal production requirements could be challenging to meet. For CdTe PV, it has been shown that meeting even conservative demand forecasts would be limited by the periodic availability of Te, rather than its scarcity, which is strongly dependent on the supply chain and production methods of Cu (Fthenakis 2012; Bustamante and Gaustad 2014).

Table 1 Total mass and recovery of metals produced

Product	Quantity produced	Recovery (%)
Zinc	74 kg	90.8
Copper	104 kg	95.5
Lead	35 kg	94.1
Cadmium	267 kg	79.4
Tellurium	55 kg	86.9
CdTe PV modules	1 unit	

updated from Bartie and Reuter (2021)for material flows that produce one PV module

Resource flows and efficiencies in the mono-Si system

Figure 4 shows a Sankey diagram representing the closed, steady state Si balance for the system, with line widths proportional to the elemental Si content of each stream. It provides a visualisation of the locations and relative magnitudes of Si-containing streams, including losses, in the life cycle for a case in which 50% of the kerf residue and 95% of EOL wafers are recycled.

Considering the streams exiting the wafer cutting process, the magnitude of the loss of high-grade, expensive SG-Si as kerf becomes clear and highlights the opportunity to increase material efficiency through kerf recycling at MG-Si quality. A second option, the vertical line between wafering and the Czochralski process, is shown for recycling kerf at the higher SG-Si quality. This option will be highlighted again in the carbon footprint analysis. Based on the configuration of our simulation, considerable amounts of Si also leave the system as microsilica, a useful byproduct used as an additive in refractories and concrete (Ciftja et al. 2008), and in residues from various other processes. By identifying and quantifying losses throughout the life cycle, a more realistic view of RE is obtained.

For the scenario shown in Fig. 4, the recoveries of materials as a percentage of the quantity entering the assumed recycling process are 86.9% Si from wafers, 70.3% Ag, 82.2% Cu, 98.9% Al (including module frames), 94.1% Sn (as SnO_2), and 94.0% Pb (as PbO_2). Note that these values have been updated from a previous version (Bartie et al. 2021a)—we have removed the assumption that 10% of recycled Si wafers can be re-used directly, and have included Al recovery from module frames.

Effects of closed-loop Si recycling on PV power potential in the mono-Si system

The use of NNs as surrogates for the simulation model allows for the effects of parameter ranges to be analysed relatively easily. Figure 5 shows the combined effects of closed-loop EOL and kerf Si recycling, at constant primary quartzite consumption, on the nominal PV power that could be generated from all the Si available in the system at a PCE of 21.7%. As one would intuitively expect, increased circularity increases power generation potential without the need for increased primary resource consumption. As described in Sect. 2.1, this effect can be quantified realistically using the thermodynamic process simulation approach. Three scenarios are shown as points in Fig. 5 at a constant quartzite consumption of 100 kt—the reference scenario with no recycling, a 95% EOL recycling scenario, and one in which

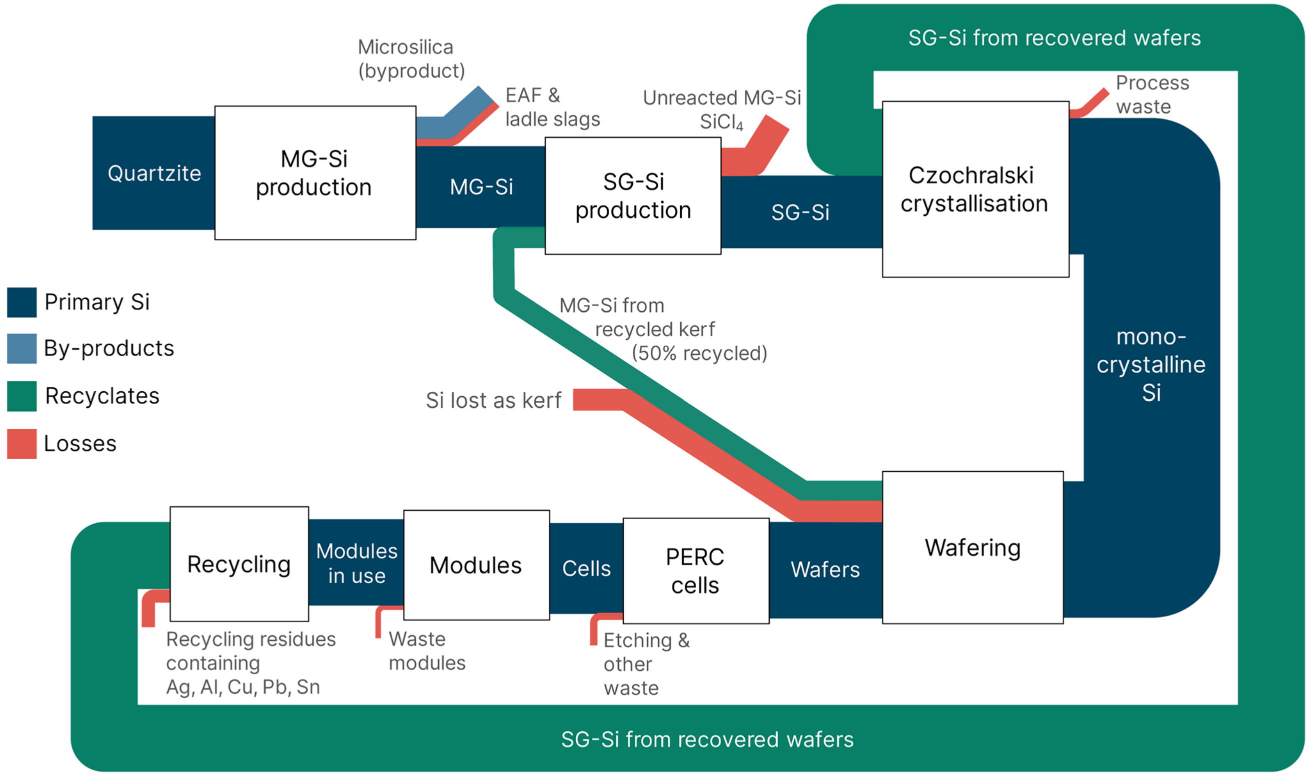
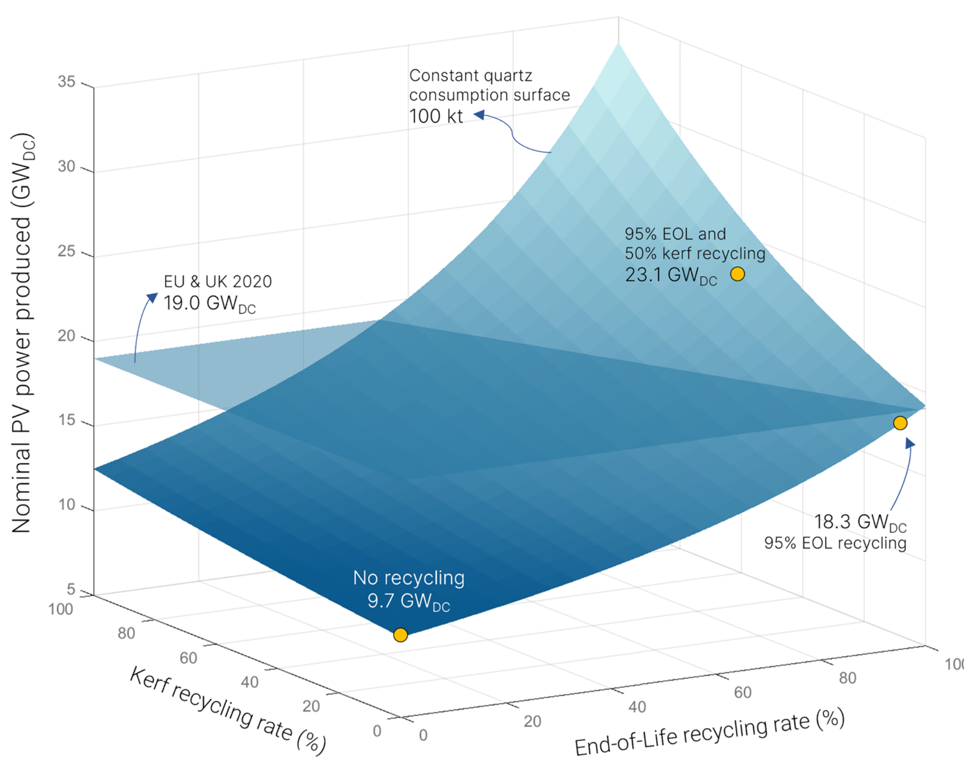


Fig. 4 Sankey diagram of the balanced flows of Si through the life cycle with 95% EOL recycling and 50% kerf recycling (Bartie et al. 2021b)

Fig. 5 Combined effects of EOL and kerf recycling on nominal PV power production at constant primary quartzite consumption (updated from Bartie et al. (2021a): PCE changed from 23% to 21.7% for consistency in this paper and actual annual PV deployed updated to 2020; underlying data can be found in the Supporting Information)



50% of the kerf residue is additionally recycled. While the simulation model allows for the curved surface to be generated for any quantity of quartzite consumption, 100 kt is used in Fig. 5 to allow for a horizontal surface representing the total nominal PV power generation capacity deployed by the EU and UK in 2020 (IEA 2021), to be shown as a tangible reference.

For the consumption of 100 kt of quartzite with no recycling, the equivalent nominal PV power generation potential is 9.7 GW_{DC}. With 95% EOL recycling, this value increases by 88%, less than 95% because of accounting for losses in the system. Adding the 50% kerf recycling results in a 137% increase from the reference scenario. In this simulation, an increase greater than 100% is achieved because primary quartzite consumption is not displaced by the equivalent amount of recycled Si but is added to the available Si in the system, representing growth in PV deployment. The 16.7 GW_{DC} of nominal PV power reference can be achieved via any combination of raw material consumption, EOL recycling rate, and kerf recycling rate on the horizontal plane at that value. However, potential trade-offs with environmental impact and economic viability must also be evaluated. More detailed results can be found in Bartie et al. (2021a).

Using exergy to identify sources of resource inefficiency

The exergy flows that occur during CdTe and mono-Si PV manufacturing are shown in Fig. 6. Of the exergy inputs (material and energy streams combined), 41% and 24% are lost irreversibly in the CdTe and mono-Si systems, respectively, which equate to specific exergy dissipations of 19.5 and 15.5 kWh/m^2 of these modules produced, respectively. Module sizes are based on the specifications of commercial units—0.72 m^2 for CdTe (First Solar 2018) and 1.96 m^2 for Si (Frischknecht et al. 2015). Reducing the amounts of Al used for module frames (assumed for both systems) or producing frameless modules, reducing the use of adhesives and polymer foils, and the incorporation of more renewable energy sources into electricity grid mixes are highlighted as potential opportunities for RE optimisation under the operating conditions specified in the models presented here.

This type of analysis can be done for any process or combination of processes in the life cycle. In the CdTe system, for example, a Zn fuming furnace was introduced to connect the Pb and secondary Cu systems and this resulted in a 4% increase (from 53 to 57%) in overall system exergy efficiency (Bartie et al. 2020). At the same time, however, this resulted in a 7% increase in GWP and a 9% increase in AP, highlighting the interaction and trade-offs between RE and environmental impacts that need to be optimised. It should be noted that although this system is based on best available techniques, it has not yet been optimised. Therefore, there is a high probability that efficiencies could be further improved through innovation while also reducing the magnitudes of any trade-offs. Figure 7 shows the contribution of subsystems to the total exergy cost for the production of Te and Cd. Exergy cost is expressed as exergy (in kWh) dissipated per tonne of metal produced.

For both Te and Cd, more than half of the total exergy dissipation originates from energy-intensive electrochemical refining processes. When electricity is used, its exergy (which equals its energy) is completely dissipated, regardless

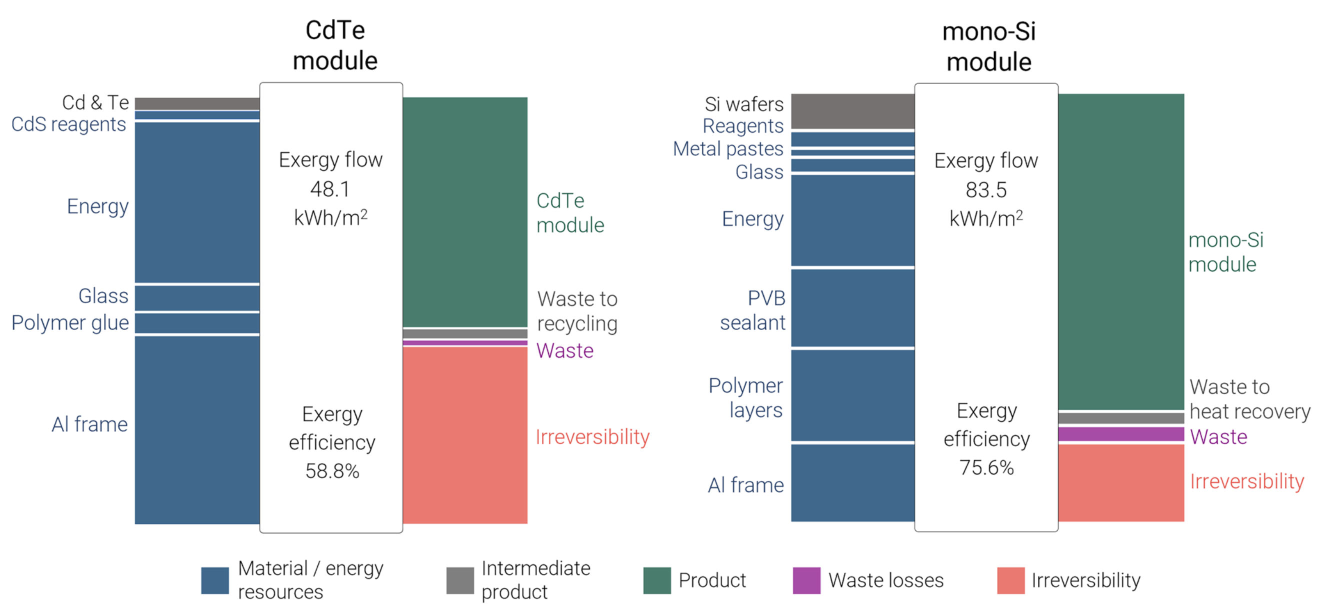


Fig. 6 Exergy flow, irreversibility, and efficiency for CdTe and mono-Si PV module manufacturing (underlying data can be found in the Supporting Information)

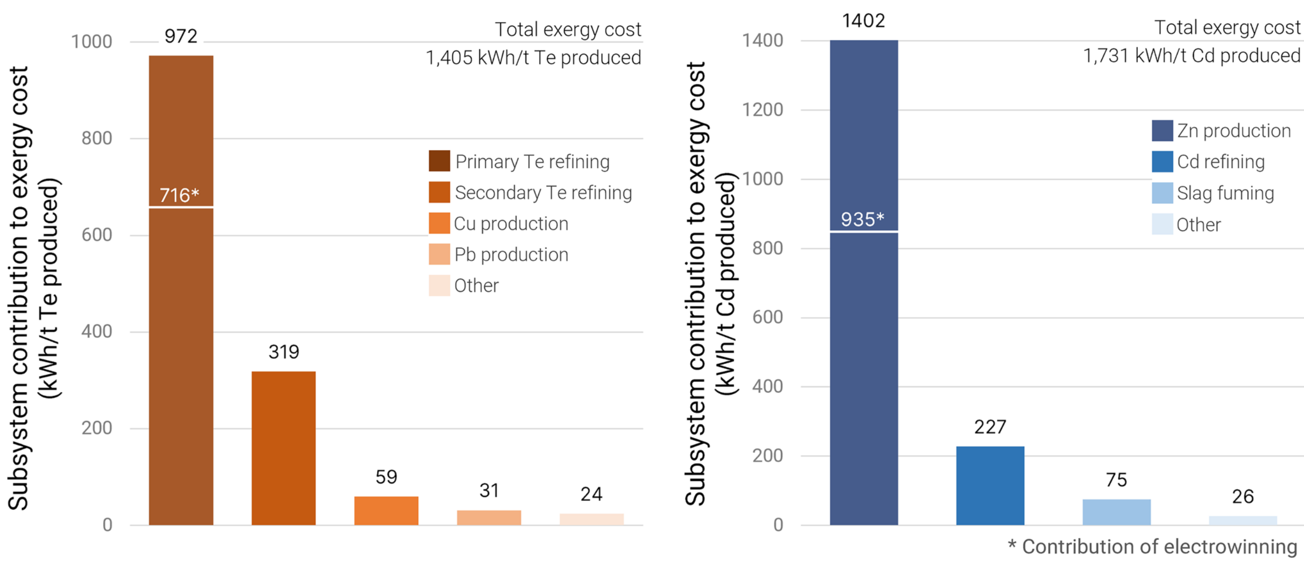


Fig. 7 Subsystem contributions to exergy cost for the production of Te (left) and Cd (right), all values determined within the process simulation model (Heibeck et al. 2020)

of how it was produced. However, its embodied environmental impact is strongly influenced by *how* it was produced (e.g. the mix of fossil and non-fossil resources used), which depends strongly on *where* it was produced. Therefore, the most effective way to improve the net sustainability of these processes would be to locate them where electricity grid mixes are made up of predominantly renewable energy sources and not in locations where carbon-based electricity grids are still the norm. This is discussed further in Sect. 3.4.

Environmental impact

Impacts and allocation challenges in the CdTe system

To produce all the quantities in Table 1, the total system GWP has been estimated at 733 kg CO_2-equivalent ($kgCO_2e$) and the total AP at 7.7 mol H^+ equivalent (mol H^+-eq.) using the ILCD midpoint v1.09 (ILCD 2011) life cycle impact assessment method. To be able to state the environmental impact associated with individual products in this large system, the overall impacts need to be distributed in an appropriate way. As mentioned, LCA guidelines recommend that, if it cannot be avoided, allocation should be based on physical relationships between the products and their environmental impacts or on economic value, the former the preferred option. In multi-metal systems such as that presented here, allocation cannot be avoided as subdivision of the production processes is not possible (Ekvall and Finnveden 2001). Following these guidelines, the distributions of overall impacts to the system's products were calculated by quantity produced, exergy cost, exergy content, and economic value for comparison (see Table 2).

As is evident from Table 2, the results are generally inconsistent—it is difficult to decide which set of distributed

Table 2 Total emission distribution between outputs by mass, exergy cost, exergy content, and value of system products

	Allocation parameter			
	Mass	Exergy Cost	Exergy Content	Economic value*
System output	Percentage of total impact allocated to output			
Copper	41.7	38.0	24.0	62.9
Zinc	29.7	19.2	43.3	16.4
Lead	14.0	6.8	4.9	6.2
Cadmium	0.097	0.033	0.071	0.055
Tellurium	0.011	0.017	0.0078	0.22
CdTe PV modules	14.4	35.8	27.7	14.1#

*Based on average commodity prices for 2020 (statista.com)

Based on average selling price ($0.345/W) for 2019 (seekingalpha.com)

impacts is most likely to be representative of reality. Similar challenges have been reported by others (e.g. Stamp et al. 2013; Bigum et al. 2012). Furthermore, various additional calculation approaches are recommended in LCA guidelines to account for EOL impacts (i.e. cut-off/recycled content, EOL recycling/avoided burden), which are applied differently for open loop and closed loop recycling, and in attributional and consequential LCAs (Nordelöf et al. 2019). Detailed descriptions are beyond the scope of this paper but suffice it to say that these add further complexity and can be counterintuitive (Guinée and Heijungs 2021). Difficulties also arise when attempting to compare results with those of other researchers, as the studied systems are often not directly comparable (Farjana et al. 2019). In this study, only some of the allocated values agree with those published by e.g. Nuss and Eckelman (2014), Van Genderen et al. (2016), and Ekman Nilsson et al. (2017), and only if mixed allocation methods are used. A hybrid allocation method could, therefore, be implemented in some way, but would likely have to be based on somewhat arbitrary and subjective assumptions.

For the current system, as defined in the simulation: the price of Cd is only 3% of that of Te. To produce the CdTe semiconductor, however, Cd is clearly just as important as Te. In this case, mass-based allocation would be more appropriate. Looking at the overall system in which much larger amounts of Cu, Zn, and Pb are produced with Cd and Te for applications other than PV, value-based allocation would probably make more sense as the producer's objective would be to maximise profit. Because Te is significantly more expensive than all the other metals, a portion of the environmental impact would be allocated to it—in this case an order of magnitude more than with the other allocation factors. Such a small quantity is produced; however, that its impact is virtually negligible relative to that of the system (0.2% based on economic allocation). Similarly, and even though eleven times more Cd than Te leaves the system as a product, its allocated impact is even smaller (less than 0.1% for mass, exergy, and economic allocation). Allocation based on exergy cost gives impacts several orders of magnitude higher for both Cd and Te, but it is unclear how a sensible choice between the allocation factors would be made. Subjective or arbitrary decisions would have to be made in this scenario to generate an uncertain result that would likely carry low credibility.

The simplest and clearest way to avoid having to choose between various EOL and allocation methods and/or combinations of them is to make use of detailed process models such as those presented here and in other recent work (Abadías-Llamas et al. 2019; Bartie et al. 2020; Hannula et al. 2020; Fernandes et al. 2020). The flowsheet models contain all the necessary detail to determine the absolute emissions from every process in the system as and when they really occur, eliminating the need to divide the overall emission between outputs.

Effects of circularity on carbon footprint in the mono-Si system

Following the same approach as for nominal power generation, Fig. 8 shows the CO_2-equivalent emissions per nominal kW power generated for the German electricity mix and quantifies how increased circularity could increase sustainability. With no recycling, emissions amount to 659 $kgCO_2e/kW_{DC}$ based on the assumptions in our simulation, decreasing by 13% with 95% EOL recycling and an additional 1% by adding 50% kerf recycling. The decreases are mainly due to reductions in Scope 2 emissions—in the system as defined here, EOL recycling bypasses the Siemens process, which is the most energy-intensive process in the life cycle, while kerf recycling only bypasses MG-Si production. An additional 3% decrease in emissions could be achieved by recycling kerf at SG-Si quality, in which case the recyclate would also bypass the Siemens process (see Fig. 4). This analysis highlights and quantifies the effects of recyclate quality on sustainability and the potential benefits innovation and development of high-quality recycling processes could bring, albeit that the potential environmental footprint of such upcycling processes has not been considered here.

The locations of the energy-intensive processes have a strong influence on emissions. Although it is assumed in Fig. 8 that the entire life cycle is co-located on the German electricity grid, it is instructive to point out that moving it to Australia, for example, would result in a 32% increase in overall CO_2-equivalent emissions, while moving it to Brazil would result in an 26% decrease. There are, of course, other factors at play, such as where material resources are geographically located, production costs at different locations, transport costs, trade regulations, etc. No one conclusion should be viewed in isolation, but rather as part of the overall system that needs to be optimised.

Combined effects of Si wafer thickness and EOL recycling on carbon footprint in the mono-Si system

Over the last decade, improvements in cell and module efficiencies have resulted in a 50% reduction in the amount of Si needed to generate a Watt of power and this trend is expected to continue. The average thickness of the most frequently used Si wafers is currently between 170 and 175 µm, accounting for about 72% of the weight of a standard mono-Si cell (VDMA 2021). This value is expected to decrease to between 150 and 160 µm by 2031 (VDMA 2021), further reducing the consumption of Si for PV systems. Figure 9 shows the variation in CO_2-equivalent emissions with EOL and kerf recycling rate for wafer thicknesses of 150 and 175 µm. The reduction from 175 to 150 µm (without recycling) results in a 5% reduction in emissions. However, combined with an EOL recycling rate of 95%, emissions decrease by 15%. Compared to recycling alone (Fig. 8), the contribution of a 25 µm

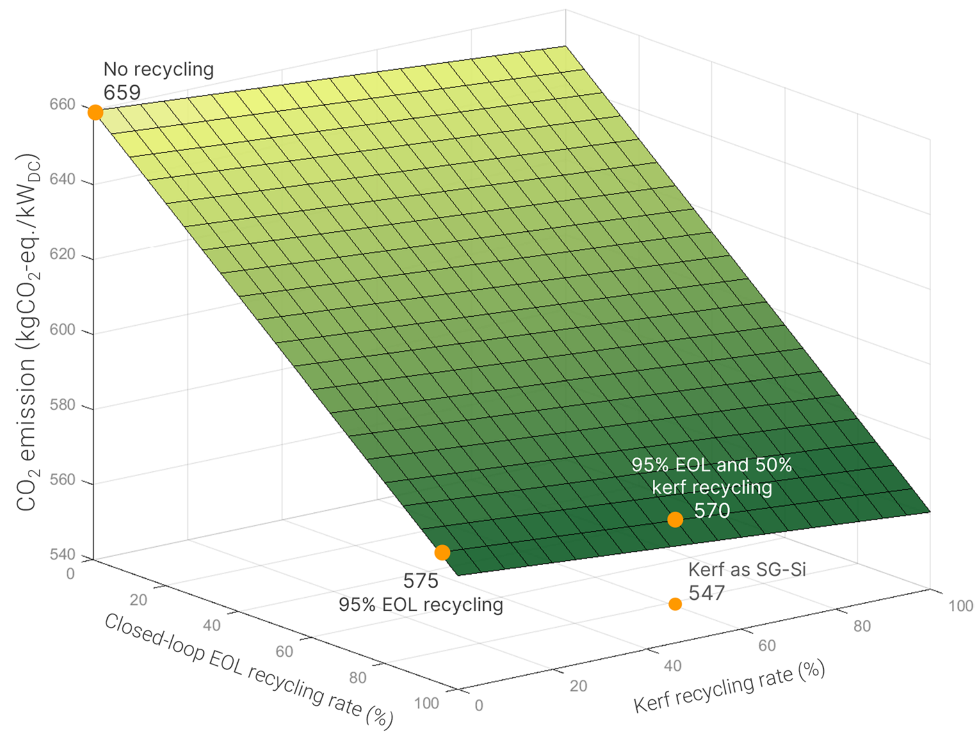


Fig. 8 CO2-equivalent emissions per nominal kW of power generated for module production on the German electricity grid (updated from Bartie et al. (2021a) for a PCE of 21.7% and an electricity supply emission factor of 0.558 kgCO2e/kWh (Treyer 2021); underlying data can be found in the Supporting Information)

reduction in wafer thickness to decreasing carbon footprint is relatively small. This analysis of the effects of wafer thickness highlights one of the advantages of the process simulation approach—the ability to change process parameters and generate new process inventory data to assess the impacts of expected technology developments.

Technoeconomic assessment and the effects of carbon taxation in the mono-Si system

Figure 10 shows the impacts carbon taxation on MSP and how it is influenced by closed-loop recycling. A carbon tax of \$100/tCO_2e increases MSP by 24% (from \$67/m^2 to \$83/m^2) when no closed-loop EOL recycling takes

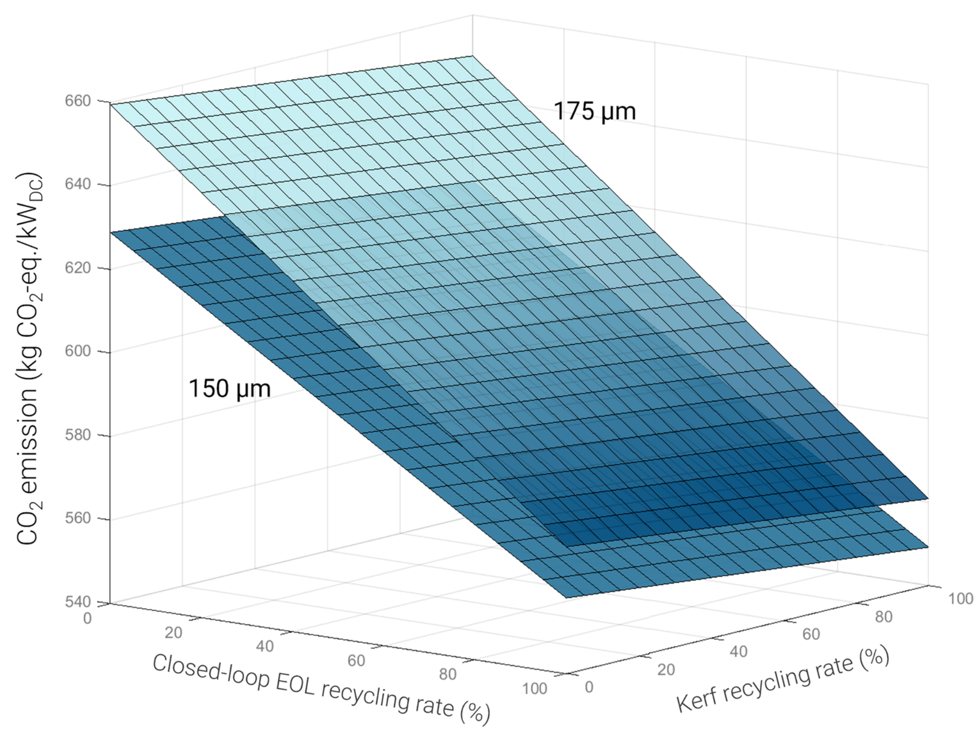


Fig. 9 Variation in CO2 emissions with EOL and kerf recycling rate for wafer thicknesses of 150 and 175 µm (for a PCE of 21.7% and an electricity supply emission factor of 0.558 kgCO2e/kWh (Treyer 2021); underlying data can be found in the Supporting Information)

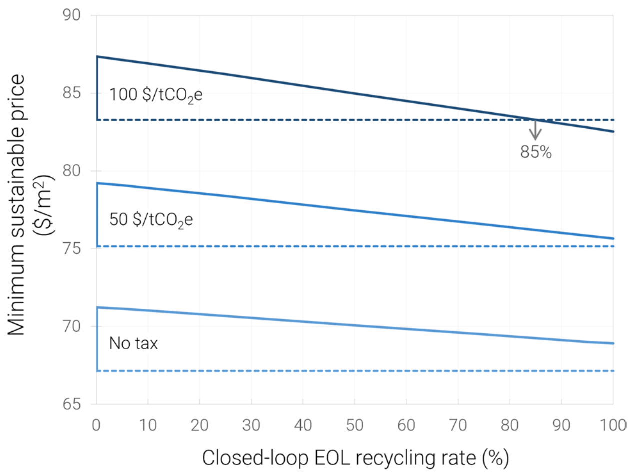


Fig. 10 Variation in manufacturing cost with EOL recycling rate at hypothetical carbon tax rates of 0, 50, and 100 $/tCO2e emitted (adapted from Bartie et al. 2021b; underlying data can be found in the Supporting Information)

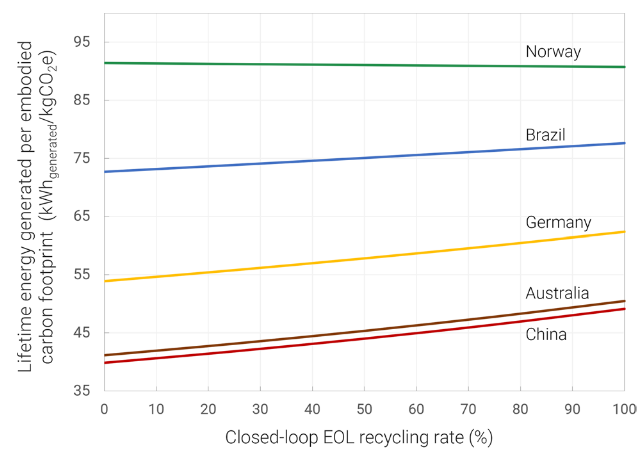


Fig. 11 Ratio of lifetime PV energy generated and its manufacturing carbon footprint as a function of EOL recycling rate (underlying data and electricity supply emission factors can be found in the Supporting Information)

place, and by 20% at a 95% recycling rate. As soon as recycling is introduced, an upwards step change in MSP occurs due to the fixed costs of the recycling process. As recycling rate then increases, MSP decreases but can only break even with the original MSP when the carbon tax is higher than approximately \$75/tCO$_2$e. Below this level, recycling cannot compensate fully for its cost. At a tax level of \$100/tCO$_2$e, however, the minimum recycling rate needed to break even is a relatively high 85%. These effects follow the same pattern for the LCOE, but are less significant. A \$100/tCO$_2$e tax results in a 6.6% increase in LCOE, from 7.81 to 8.33 c/kWh. However, the effects of balance-of-system items such as land, concrete, support structures, and others on the overall carbon footprint have not been included in our LCOE calculations yet. With these included, the effect of carbon tax on LCOE would be larger.

Both recycling and carbon taxation aim to reduce environmental impacts and lead to increased cost. The linking of process, environmental, and technoeconomic models allows for analyses such as this; however, that shows that there are conditions under which increased recycling could reduce costs to below their original values despite the carbon tax.

The location dependence of the embodied carbon footprint of PV energy in the mono-Si system

As highlighted throughout this paper, infrastructure location plays a pivotal role in life cycle sustainability. Figure 11 shows the ratio of power generated over the lifetime of a mono-Si PV system and its manufacturing carbon footprint as a function of closed-loop EOL recycling rate for different locations. It is assumed that manufacturing takes place in the country indicated that the PV system has a 30-year lifetime with a 0.5%/year degradation rate and a PCE of 21.7%. The average annual insolation is assumed to be 1,500 kWh/(m^2.year).

Figure 11 clearly shows how manufacturing location influences carbon footprint. Moving manufacturing from China or Australia to Germany, for example, would have a positive effect by increasing the ratio of energy generated and CO_2 emitted by more than 30% in the case without recycling. In China and Australia, 71% and 77% of electricity were generated from fossil resources in 2020, respectively (IEA 2022). Countries like Brazil and Norway, on the other hand, respectively, generated 75% and 98% of their electricity using wind, solar PV, and hydropower in the same year (IEA 2022). To maximise energy output per embodied carbon footprint, it is clear that infrastructure development should occur away from countries still largely dependent on fossil fuels for power generation.

Increased circularity lowers the embodied carbon footprint of PV energy, but this effect is weaker the lower the carbon intensity of the relevant electricity grid. The reason is that the strongest effect of Si recycling is its contribution to avoiding electricity consumption in energy-intensive processes and hence avoiding Scope 2 emissions. The carbon intensity of Norway's electricity grid is an order of magnitude lower than those of the other countries, and as a result, this life cycle's Scope 2 emissions are lower than its Scope 1 emissions, the latter constant regardless of location. In this case, the increase in Scope 1 emissions brought about by increased recycling is higher than the simultaneous decrease in Scope 2 emissions (explained in Sect. 3.2.2), resulting in a slightly negative slope for Norway in Fig. 11. To take full advantage of the Si circularity effect, it would make sense to focus on using secondary Si in locations where electricity grids are most carbon-intensive.

These comparisons are based on recently published emission factors for the *generation, supply, and distribution* of electricity from the ecoinvent (version 3.8, 2021) database (Wernet et al. 2016). It should be noted that these are higher than the carbon intensities of electricity *generation* reported by the European Energy Agency (EEA) and the International Energy Agency (IEA), among others. The EEA reports a carbon intensity of 0.311 $kgCO_2e/kWh$ for Germany (EEA 2021), for example, compared to the 0.558 $kgCO_2e/kWh$ from the ecoinvent database (Treyer 2021). While the former is more recent (2020 vs. 2018), it does not include emissions associated with supply chains and electricity losses during transmission across networks. As a result of the time lag, the values used to generate the results presented here (and used for environmental impact assessments in general) may not fully reflect recent progress in reducing the carbon intensities of electricity consumption.

Conclusions and outlook

This paper discussed the links between the metals and PV industries with specific reference to the CdTe and mono-Si PV life cycles. The approach presented here allows for evaluation of complex systems in terms of resources, environmental impact, technoeconomic performance, and their interactions simultaneously. The use of a physics-based foundation of inventory data on a process simulation platform ensures consistency in assessments of these dimensions, so facilitating rigorous life cycle sustainability assessment. Simulation results identify system configurations that enhance RC and RE and reveal the system-wide effects of changes in these on environmental impact and technoeconomic performance, so quantifying the positive impacts of increased circularity—therefore, CE—on the system's sustainability. The following conclusions are drawn from the work to date:

- Of the minor metals needed for CdTe and other PV module manufacturing, most are co-products of other production systems and many are CRMs. Their availability must be ensured by designing and building the necessary infrastructure without delay. The location of the infrastructure plays a decisive role in life cycle sustainability.
- In the CdTe system, an increase in overall RE was achieved by linking the Pb and Cu production subsystems by means of additional metallurgical infrastructure. The increased efficiency, however, resulted in increased environmental impact. Introducing the other dimensions of sustainability (society, economy) as well as other system improvements creates various trade-offs that need to be optimised for overall sustainability and CE.
- The analysis of recycling in the mono-Si system, of both the internal kerf residue and EOL wafers, quantified how circularity and the quality of recycled Si influence RE and carbon footprint. Both kerf and EOL wafer recycling increase the potential to generate PV power without additional consumption of the primary mineral resource. At the same time, the overall carbon footprint per module is reduced, and more so when the recyclates are of higher purity. Si recovered from wafers at SG-Si quality bypass the primary production of both MG-Si and SG-Si, resulting in significant reductions in Scope 2 (i.e. power consumption-related) emissions. For Si recovered from kerf at MG-Si quality, this effect is smaller as only the MG-Si production process is bypassed. This highlights the benefits of keeping recycling loops in the life cycle as small as possible and the importance of innovation and investment in recycling infrastructure capable of producing high-purity secondary resources for sustainable CE.
- Analysis of the combined effects of recycling and a hypothetical carbon tax in the mono-Si PV system showed that increased recycling alone is unlikely to be successful at balancing the increase in MSP caused by such a tax. Recycling itself initially increases cost, which then decreases as recycling rate increases. However, it only breaks even with the original cost at high recycling rates and high taxes. As intended with such a tax, the best remedy would be to avoid emissions in the first place, so increasing sustainability. The follow-on effect on LCOE is smaller due to other costs over and above that of the modules.
- As also reported by others, the sensible allocation of environmental impacts to products in multi-output systems remains challenging. The clearest and most efficient solution is to use process simulations that give the real emissions for every process from which impacts can be calculated directly instead of having to distribute overall emissions between products, so avoiding allocation altogether, even for very complex systems. For cases that include EOL treatment, the simulation approach provides the same clarity as recycling is modelled in the foreground system, negating the need for assumptions about what might be occurring in the background system.
- While numerous factors are at play, real RC and RE can only be quantified if entropy creation (i.e. exergy dissipation) is also accounted for. This "hidden" dissipative energy flow is usually where costs are incurred and, if not accounted for rigorously, may result in faulty policy and economic models.
- This work also suggests that supply chains for PV systems should be positioned in low environmental impact energy infrastructures to ensure that the embodied footprint of the system, including recycling, is as low as possible and enhances the performance of the system as

a whole, i.e. to maximise the ratio of energy delivered by the PV system over its lifetime and its embodied carbon emissions.

In summary, the infrastructure needed to enable the circular economy and to power sustainability is as critical as the critical materials it needs to produce from primary and secondary resources. It needs to facilitate the running of the life cycle shown in Fig. 1 at its thermodynamic limits—at the highest possible efficiency and lowest possible footprint—within prevailing social and economic constraints. It is envisaged that optimisation frameworks and results from this work would contribute to guiding strategy and policy in the renewable energy arena. Specifically, it also clearly shows quantitatively that the system, including recycling, must be positioned in energy landscapes with the lowest possible impact to ensure that the footprint of the system itself is as low as possible. The presented approach is not limited to the analysis of PV systems and can be applied to any product life cycle.

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Author contributions *Neill Bartie* involved in conceptualisation, methodology, software, validation, formal analysis, investigation, writing—original draft, visualisation; *Lucero Cobos-Becerra* took part in conceptualisation, validation, investigation, project administration; *Magnus Fröhling* involved in conceptualisation, validation, writing—review & editing, supervision; *Rutger Schlatmann* took part in conceptualisation, methodology, validation, writing—review & editing, supervision; *Markus Reuter* involved in conceptualisation, methodology, software, validation, writing—original draft, review & editing, supervision.

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Data availability The underlying data for Figs. 5 to 11 can be found in the Supporting Information.

Code availability As mentioned and cited in Sect. 2.1, two simulation models are available online (Heibeck et al. 2020; Bartie and Heibeck 2020).

Declarations

Conflicts of interest/Competing interests Not applicable.

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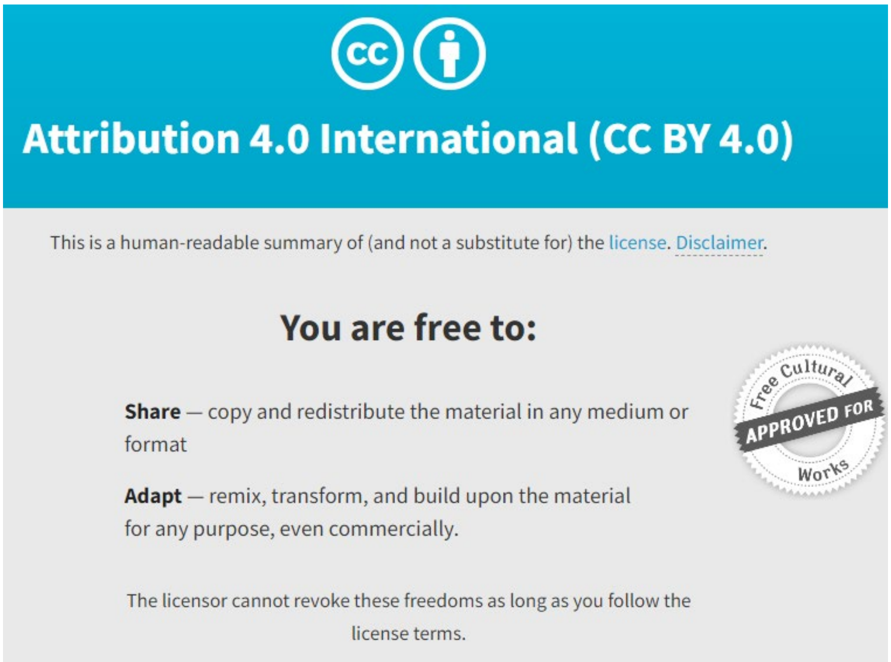
Annexure D: Article 4

Cost versus environment? Combined life-cycle, techno-economic, and circularity assessment of silicon and perovskite based photovoltaic systems



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RESEARCH ARTICLE

Cost versus environment?

Combined life cycle, techno-economic, and circularity assessment of silicon- and perovskite-based photovoltaic systems

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Abstract

Photovoltaics will play a key role in future energy systems, but their full potential may not be realized until their life cycles are optimized for circularity and overall sustainability. Methods that quantify flows of compound and minor element mixtures, rather than non-mixed elemental flows, are needed to prospectively analyze and predict inventory and performance for complex technology life cycles. This study utilizes process simulation to resolve the mass and energy balances needed to rigorously analyze these complexities in circular systems. Using physics-based prospective inventory data, we simultaneously assess the environmental and techno-economic performance of three photovoltaic life cycles and predict the effects of circularity on resource efficiency, carbon footprint, and levelized cost of electricity. One inventory dataset is generated per life cycle to ensure alignment between assessments and to identify trade-offs between environmental and techno-economic performance with respect to circularity, so linking circularity and sustainability. The linked material and energy resource and techno-economic models allow for the impacts of carbon taxation and the moderating effects of circularity to be explored. In addition to the clear environmental benefits of increased circularity, we find that it could dampen the cost impact of taxation. While confirming that perovskite-based modules, single junction or in tandem with silicon, clearly outperform the silicon market standard both techno-economically and environmentally, we show that maximum circularity does not automatically deliver the most sustainable outcome. The approach enables assessment of the combined impacts of specific technological, commercial, and policy choices made by different actors along the photovoltaic value chain. This article met the requirements for a gold–gold *JIE* data openness badge described at http://jie.click/badges.

KEYWORDS
circularity, industrial ecology, photovoltaics, process simulation, sustainability, techno-economics

1 | INTRODUCTION

1.1 | Current and emerging photovoltaic technologies

Manufacturers of photovoltaic (PV) technology have been successfully improving techno-economic performance with materials and cell architectures that increase power conversion efficiency (PCE), while reducing material consumption and production costs. This has resulted in levelized costs of electricity (LCOE) lower than that of fossil-based power generation (Fraunhofer-ISE, 2021). Wafer-based crystalline silicon (Si) PV, in particular the "passivated emitter rear cell" (PERC) architecture, is expected to remain the market leader for at least the next decade (VDMA, 2021). A promising emerging technology is based on mixed lead (Pb) halide compounds that crystallize in the "perovskite" structure. Perovskite absorbers have seen the steepest rise in PCE with the record single-junction cell efficiency currently 25.7%, up from 14% less than 10 years ago (NREL, 2022). Their high PCE and low cost, among others, allow perovskite cells to compete with existing commercial technologies (Liu et al., 2021). Challenges that still prevent commercialization are the long-term stabilities of some component materials and interfaces. Including a cesium cation (Cs^+) in certain perovskite structures has been shown to improve thermal and moisture stability (Saliba et al., 2016). A detailed comparison of PV types, advantages, and disadvantages is provided by Muteri et al. (2020).

The rapid progress in perovskite research has also driven fast development of perovskite-based tandem devices (Werner et al., 2018). Perovskite/perovskite and perovskite/Si tandem configurations achieve higher efficiencies by taking advantage of perovskites' tunable bandgap to better exploit short-wavelength photon energy (Leijtens et al., 2018). In four-terminal tandem configurations, two independently manufactured sub-cells are stacked on top of each other. They can operate independently these to maximize performance (Leccisi & Fthenakis, 2020). With a theoretical four-terminal efficiency limit of approximately 46% (Eperon et al., 2017), these devices exceed the theoretical single-junction limit of 33% (Shockley & Queisser, 1961) by far. Liu et al. (2022) recently reported a Cs^+-doped perovskite module efficiency of 21.08%, and Si and perovskite/Si tandem efficiencies are expected to reach 22.2% and 28% by 2031, respectively (VDMA, 2021).

Figure 1 depicts the structures of the (a) perovskite, (b) silicon, and (c) perovskite/silicon tandem modules described above and discussed in this paper.

1.2 | End-of-life PV and circular flows

Solar modules deployed in the 1990s, primarily Si based, are reaching the end of their useful lives, with rapid increases in PV waste volumes expected by 2030. It has been estimated that the recyclable materials in end-of-life (EoL) devices accumulated by 2050 could be used to produce about 630 GW of new capacity (IRENA & IEA-PVPS, 2016). That is, if all of it could be recovered at purities high enough for re-use in PV systems. Despite the high energetic and economic value of contained Si, most recycling facilities presently only recover bulk materials like glass cullet, cabling, and aluminum frames (Isherwood, 2022). Integrated processes aimed at recovering Si and other elements are complex, and while some have been demonstrated at laboratory or pilot scale, commercial examples barely exist (Deng et al., 2022). Promising recycling options for perovskites have only been investigated at laboratory scale (Liu et al., 2021). The further development of these processes, complemented by innovative design-for-recycling to simplify dismantling and separation processes, will maximize the quantities and purities of materials brought back into the economy (Norgren et al., 2020). However, the recovery of materials from EoL devices is subject to limits imposed by the laws of physics, including solution thermodynamics. Complete "unmixing" and recovery of individual elements is impossible due to the irreversibility of processes, as described by the second law of thermodynamics. The effects of these limitations on material and energy flows must be analyzed using fit-for-purpose tools to assess the contribution of circular strategies such as recycling to overall sustainability in detail, as circular flows do not necessarily guarantee sustainable outcomes (Geissdoerfer et al., 2017; Korhonen et al., 2018).

Besides the physical limits, circular flows are unlikely to occur unless driven by economic or regulatory incentives. In the Si PV case, for instance, still-low waste volumes and low demand for high-quality integrated recycling have limited investment in innovation and, as a consequence, recycling costs remain high (Cui et al., 2022). With the expected increase in waste volumes, programs like the European Green Deal (European Commission, 2020a) that promote circular economy (CE) and circular business models are important to stimulate investment and accelerate development despite the limited present demand. Also important are regulations that stipulate recovery targets for specific materials and penalize pollutant emissions. The effects of such measures also need to be quantified to assess whether they do, in fact, enhance overall sustainability.

Lindgreen and colleagues highlight the lack of assessment approaches that quantify the links between circularity and sustainability, that is, the environmental, economic, and social impacts of circular strategies. Such approaches are needed to ensure systemic change for sustainable development rather than mere incremental improvements driven by *"promises of economic gains through resource efficiency"* (Lindgreen et al., 2020).

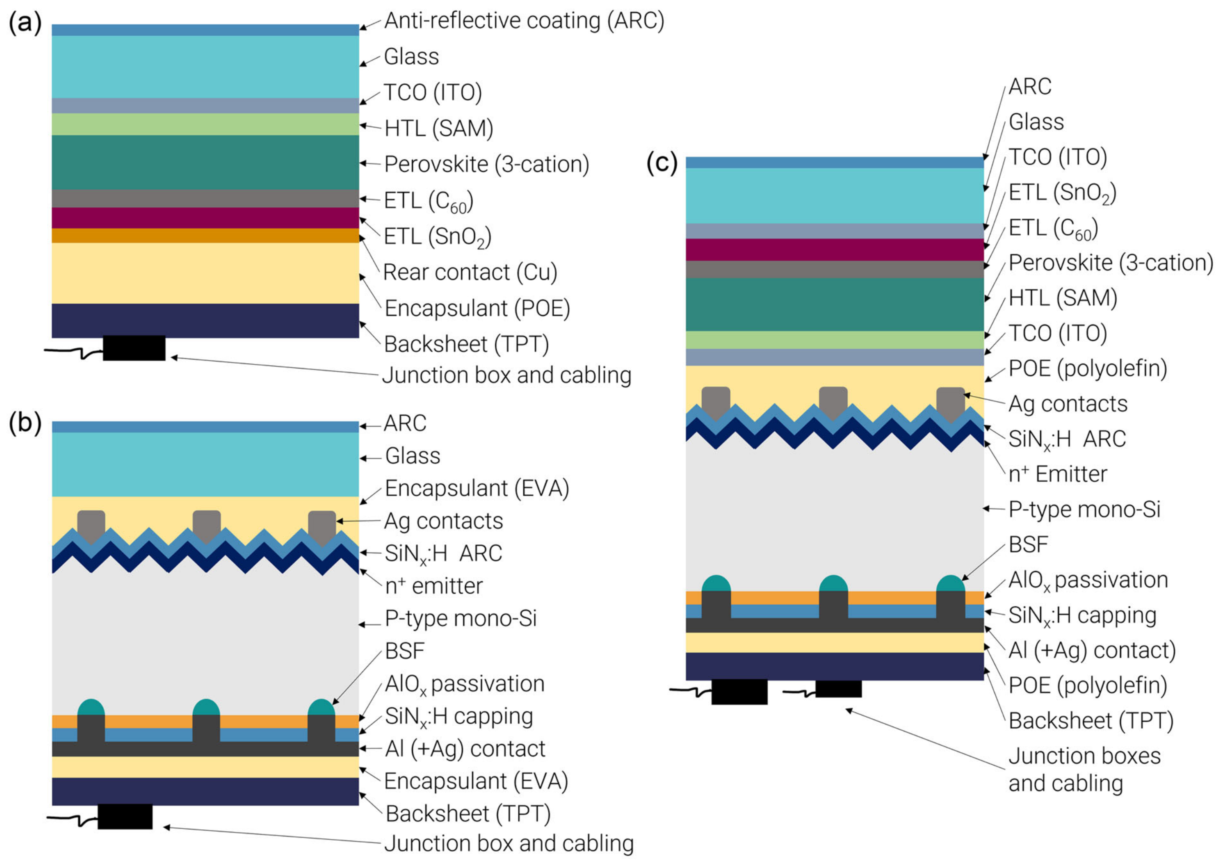

FIGURE 1 (a) Perovskite (single-junction), (b) Silicon (passivated emitter rear cell architecture), and (c) perovskite/silicon tandem (four-terminal) module configurations. We assume reference power conversion efficiencies of 18%, 21.7%, and 27.3% for perovskite, silicon, and tandem modules, respectively, based on previous publications for similar modules (Sofia et al., 2020). Explanations of the acronyms used in Figure 1 are given in Supporting Information S2.

1.3 | Assessing resource, environmental and economic performance

Environmental and economic performance are usually estimated using standardized life cycle assessment (LCA) (ISO, 2006) and techno-economic analysis (TEA), respectively, which are well-known approaches. After goal and scope definition, the foundation of LCA and TEA is the “inventory” of the system at hand, which refers to the mass and energy flows into and out of a system. Material flow analysis (MFA) is most often used and touted as the ideal tool to map life cycle inventory (Graedel, 2019). At its core, MFA refers to doing a mass balance, a fundamental concept that ensures adherence to the law of mass conservation. While suitable for single-element bulk material flows, methodological limitations hamper its use in prospective assessments when not integrated with high-detail approaches like process simulation (Baars et al., 2022). MFA does not suffice when the system includes complex material and minor element combinations such as those found in PV and other technologies, as it does not consider solution chemistry and cannot predict the distribution of minor elements between process outputs (Reuter et al., 2019). Process simulation implicitly performs MFA, but considers the enthalpy, entropy, and thus, free energy of all compounds and solutions, which is necessary to resolve complex mass and energy balances. This allows it to generate physics-based inventory data and gives it predictive capabilities, which is particularly relevant in the context of prospective assessments in which mass and energy flows are not based on historical data but need to be predicted. For these reasons, process simulation is the foundation of the work presented in this paper. More detail is provided in Section 2.2.1.

The integration of LCA and TEA is done in various ways and to various degrees and can enhance decision making in technology development (Wunderlich et al., 2021). Methodological challenges remain, often associated with inconsistent functional units and system boundaries, and discrepancies in assumptions when combining standalone LCAs and TEAs; there is a research gap with respect to tools that simultaneously perform LCA and TEA, while allowing for the influence of changes in process parameters to be investigated (Mahmud et al., 2021). Examples of integrated resource, environmental, and economic performance assessments of PV systems are scarce. Zhang and colleagues compare the environmental impacts and costs of three perovskite technologies to identify material and manufacturing method combinations that could deliver the best environmental and economic performance. The authors identify trade-offs *within* sustainability dimensions and highlight that additional methods are needed to quantify trade-offs *between* them (Zhang et al., 2022).

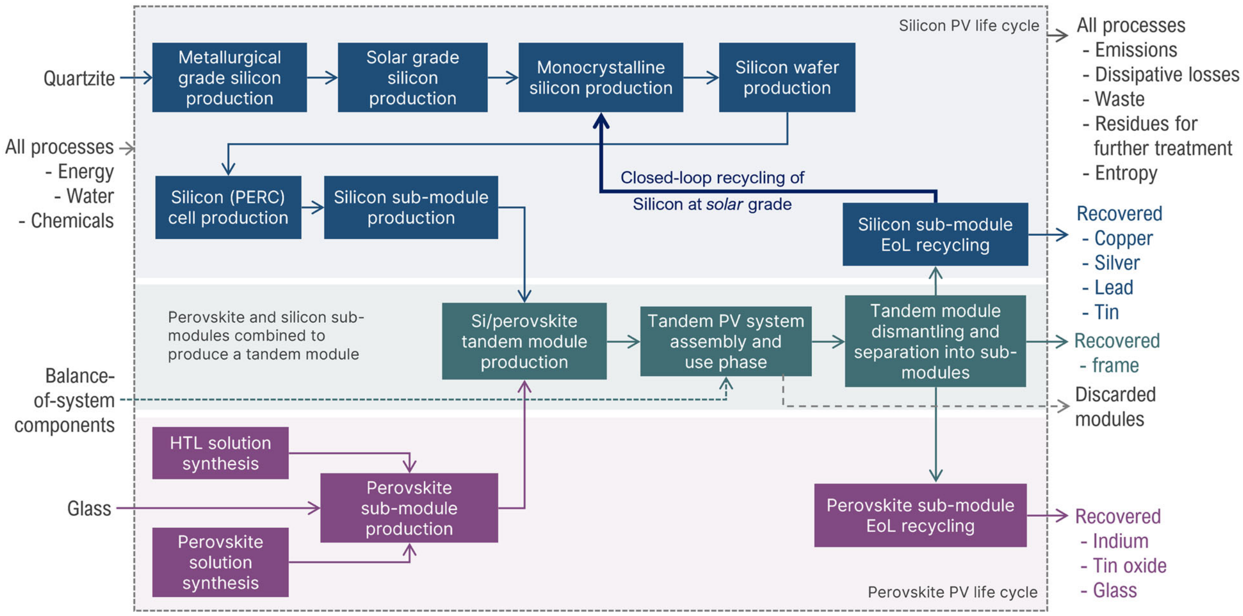

FIGURE 2 Four-terminal silicon/perovskite tandem life cycle and system boundary with the silicon and perovskite subsystems shown in the top and bottom sections, respectively. All arrows represent mass flows between processes. The silicon and perovskite systems can be visualized by connecting the respective sub-module production step with its end-of-life recycling process. The system can be considered cradle-to-cradle with respect to Si, and cradle-to-gate for other recovered products.

1.4 | Purpose of this paper

The cost of solar PV energy is already well below that of traditional power sources. While it is generally accepted that PV systems have negligible environmental impact during use (Battisti & Corrado, 2005; Lunardi et al., 2018; Muteri et al., 2020), their production and recycling processes introduce additional costs and environmental impacts. These need to be analyzed rigorously to avoid burden shifting between life cycle stages and to identify trade-offs between them. Whether increased circularity increases overall sustainability needs to be confirmed, as it is not guaranteed. By establishing its influence on life cycle inventory, circularity can be linked to environmental and economic performance via LCA and TEA to determine if and how it contributes to sustainability.

In this paper, process simulation is used to generate physics-based inventory datasets for each of the described PV systems to assess resource, environmental, and techno-economic performance consistently. We then analyze system responses to changes in the closed-loop recycling of solar-grade Si—our measure of circularity—to link sustainability and circularity via the inventory. To analyze and compare the circular PV systems, material recoveries and the energy return on investment ($EROI_{PE\text{-}eq}$) are used as indicators of resource efficiency. Carbon footprint, that is, CO_2-equivalent (CO_2e) emissions, is used as the environmental impact indicator, and LCOE as techno-economic performance indicator. The approach is further applied to evaluate the potential impacts of carbon taxation on cost, and how circularity might function as a moderator of its effects. To the best of our knowledge, this is the first study to employ thermodynamic process simulation to analyze the effects of circularity on resource, environmental, and techno-economic performance in a single assessment to compare the three contemporary PV systems, and to identify trade-offs between sustainability dimensions.

2 | METHODS

2.1 | Defining the life cycle systems

The Si system is expected to remain the market leader for at least the next decade and is the reference to which the other systems are compared. The overall tandem system and main production and recycling steps included are shown in Figure 2.

The top section in Figure 2 represents the tandem's silicon subsystem, including the shown production steps and a recycling process that recovers solar-grade Si, silver, copper, aluminum, lead, and tin. The closed-loop recycling of Si connects EoL Si recycling and monocrystalline silicon production (the thicker arrow in the top section of Figure 2). This loop is the focus of this paper and represents any reference to *circularity*. The bottom section depicts the perovskite subsystem, where indium, tin, and glass are recovered but not returned to the life cycle, as the focus of this paper is on Si circularity. Combining the sub-modules into the tandem is shown in the middle section. EoL tandem modules are disassembled into the two sub-modules and recycled in dedicated processes to maximize the quantities and qualities of recovered materials, also preventing cross-contamination with Pb compounds (Kadro & Hagfeldt, 2017). The top, middle, and bottom sections combined represents the tandem life cycle.

2.2 | Resource flows

2.2.1 | Process simulation-based life cycle inventory

Process simulation is indispensable in any process design activity and the value it adds to CE life cycle assessments has been recognized (Reuter, 1998; Reuter et al., 2019). To develop the simulation models, all unit operations that make up the aggregated blocks in Figure 2 are modeled separately and linked by material flows to create a model for that process. Process blocks are connected to create a deterministic simulation model of the whole life cycle, in the tandem system case comprising 122 unit processes, 653 material and energy flows, and 226 compounds, ions, and elements. The model automatically enforces the laws of mass and energy conservation. Because stream compositions and enthalpies are available, solution chemistry can be accounted for—the true losses from the system can be quantified via "excess" enthalpy and entropy, quantities that represent additional, usually unaccounted-for losses that occur when materials are joined in complex solutions rather than simply blended. This reveals the true non-circularity of systems. Furthermore, thermodynamic equilibrium relationships can be used to predict element distributions where process data are not available.

Closed-loop recycling and its system-wide effects are modeled in the foreground system, which avoids having to select EoL calculation approaches that are often unnecessarily complicated or counter-intuitive (Guinée & Heijungs, 2021). We do not intentionally apply the EoL ("avoided burden") or cutoff ("recycled content") recycling approach, as they merge when modeling closed loops in the foreground (Nordelöf et al., 2019). This simplifies direct assessment of the effects of CE strategies on sustainability performance. As the recycling processes do not exist commercially, simulation models are based on combinations of processes described in the literature, the authors' industry experience and own calculations, and thermodynamic equilibrium predictions to fill data gaps. Simulation models are created using HSC Chemistry (Mogroup, 2021). Detailed process descriptions are provided in Supporting Information S1 (Section S1).

We create neural network (NN)-based surrogate functions as proxies for simulation results using MATLAB (MathWorks, 2021). NNs enable generalized nonlinear process modeling of complex systems without having to define regression equations beforehand (Reuter et al., 1992). Computational efficiency is thereby enhanced to analyze inventory over parameter ranges. Simulations are run with random combinations of the independent variables of interest and the corresponding updated mass and energy flows recorded in datasets, which are then imported into MATLAB to create NNs that reliably reproduce simulation results in a fraction of the time it would take the simulation itself. This allows quantification of the selected sustainability indicators over ranges of, for example, closed-loop recycling rate, PCE, and PV system lifetime. The procedure is described in more detail in a previous publication (Bartie et al., 2021a).

2.2.2 | Resource efficiency

$EROI_{PE\text{-}eq}$, the ratio of energy delivered by, and that harvested to produce a PV system (Raugei et al., 2016) is used as an indicator of energetic resource efficiency assuming an average irradiation of 1700 kWh/(m^2 year), performance ratio (PR) of 0.75, a 30-year lifetime, and grid efficiency (η_{grid}) of 0.30. PR refers to the ratio of a PV system's rated power and that which it delivers, and η_{grid} to the efficiency with which a particular grid converts all energy harvested from the environment into an energy carrier, in this case electricity (Ibid.).

We distinguish between collection, recycling, and recovery rates. The *recoveries* of Si and other materials are determined by the efficiency of the recycling processes. The *recycling* rate is an independent variable used to specify the quantity of recovered Si returned to PV production and the quantities of other recovered materials sold. For clarity, we assume a *collection* rate of 100% so that the quantity of Si recycled also represents the quantity of originally consumed Si returned for re-use. At a recycling rate of 0%, however, nothing is returned or sold despite the hypothetical 100% collection rate, because all recoverable materials remain locked in unliberated "urban minerals," that is, spent PV modules. Until they pass through the recycling process, they only have potential value—we do not pre-emptively assign cost or environmental impact credits unless recycling actually occurs.

2.3 | Estimating carbon footprint

Distinctions are made between Scope 1, 2, and 3 CO_2e emissions (GHG Protocol, 2011) to assess carbon footprints. Scope 1 refers to direct CO_2e emissions from manufacturing and recycling, as calculated in the simulation models. Scope 2 refers to indirect emissions associated with the consumption of purchased energy. We convert power consumption quantities into Scope 2 emissions using the carbon intensity of the German electricity market mix (0.55 kgCO_2e/kWh; Wernet et al., 2016). Scope 3 refers to the embodied emissions of materials and components not modeled in the foreground. We include these for glass, aluminum frames, mounting systems, and cabling using published emission factors (de Wild-Scholten, 2013; Frischknecht et al., 2016; Stolz et al., 2017, 2020). In line with the majority of PV system assessments, two functional units—per m^2 of modules produced, and per kWh energy generated over the lifetime—are used to express carbon footprint. The former is useful for comparisons of production and recycling emissions. The latter considers PCE, lifetime, and solar irradiation, thereby accounting for PV system performance during its use phase.

It is acknowledged that considering only one impact category carries the risk of shifting burdens to from one environmental issue to others (Rosenbaum et al., 2018). The Product Environmental Footprint Category Rules (PEFCR) for PV electricity consider climate change, particulate matter formation, and resource use the most relevant impact categories (European Commission, 2020b). These and human toxicity impacts have been shown to tend in the same direction for PV (Laurent et al., 2018). Therefore, the risk of burden shifting is considered low.

2.4 | Cost calculations

2.4.1 | Minimum sustainable price

Discounted cash flow and net present value (NPV) analyses are used to estimate the minimum sustainable module price (MSP, expressed as $/Watt), which is needed to calculate LCOE. A description of the NPV calculation is given in Supporting Information S1 (Section S2). The module price at which NPV is zero is the minimum price that sustains the manufacturer while providing investors with their expected return. We assume a 6% weighted average cost of capital (WACC) for MSP calculations, in line with recently published analyses (KPMG, 2020; Roth et al., 2021; Steffen, 2020). To estimate potential revenue from recycled products, it is assumed that silver, copper, indium, and tin dioxide are recovered at saleable purities in accordance with the recycling process developers' claims of recovering pure indium metal sponge (Li et al., 2011) and other metals at purities greater than 99%, all of which can be sold to the PV industry (Huang et al., 2017). Module frames are recovered as aluminum scrap and glass as cullet. The link between Si circularity and cost is established by adjusting the total Si cost for PV production based on the share of recycled Si content. Taking an integrated life cycle perspective, the hypothetical revenue from recovered Si breaks even with the cost of recycled Si, which is assumed to be two thirds of the global Si MSP of $15/kg suggested by Woodhouse et al. (2020). For materials other than Si, potential revenue is estimated using calculated recoveries and current prices.

2.4.2 | Levelized cost of electricity

LCOE is a useful indicator of the economic performance of power supply technologies and a decision support tool when comparing them. It is the ratio of total economic investment in, and total energy generated by a PV system over its lifetime and is represented by Equation (1) (Sofia et al., 2018).

$$\mathrm{LCOE} = \frac{I_{\mathrm{system}} + \sum_1^n \frac{\mathrm{OM}}{(1+r)^n}}{\sum_1^n \frac{E(1-d)^n}{(1+r)^n}} \tag{1}$$

I_{system} is the initial PV system investment, OM is the annual operation and maintenance cost, n is the system lifetime, E is the energy yield in the first year, r is the nominal discount rate, and d is the annual degradation rate. The initial investment and *OM* were estimated using recently published breakdowns of area- and power-related costs (Zafoschnig et al., 2020). To calculate E, we assumed an average insolation of 1700 kWh/(m^2 year), a 0.5%/year degradation rate, and a performance ratio of 0.75. Cost estimation methods and assumptions are summarized in Supporting Information S1 (Section S2 and Table S2).

3 | RESULTS

The detailed mass and energy balance data for all 122 unit processes derived from, among others, reaction equations, distribution coefficients, Gibbs free energy minimization, and published information are available in a data repository (see Bartie et al., 2022). Also provided are separate inventory datasets for the perovskite, silicon, and tandem systems, each with EoL recycling rates of 0%, 50%, and 100%, as examples. These data form the basis of all results presented in this paper.

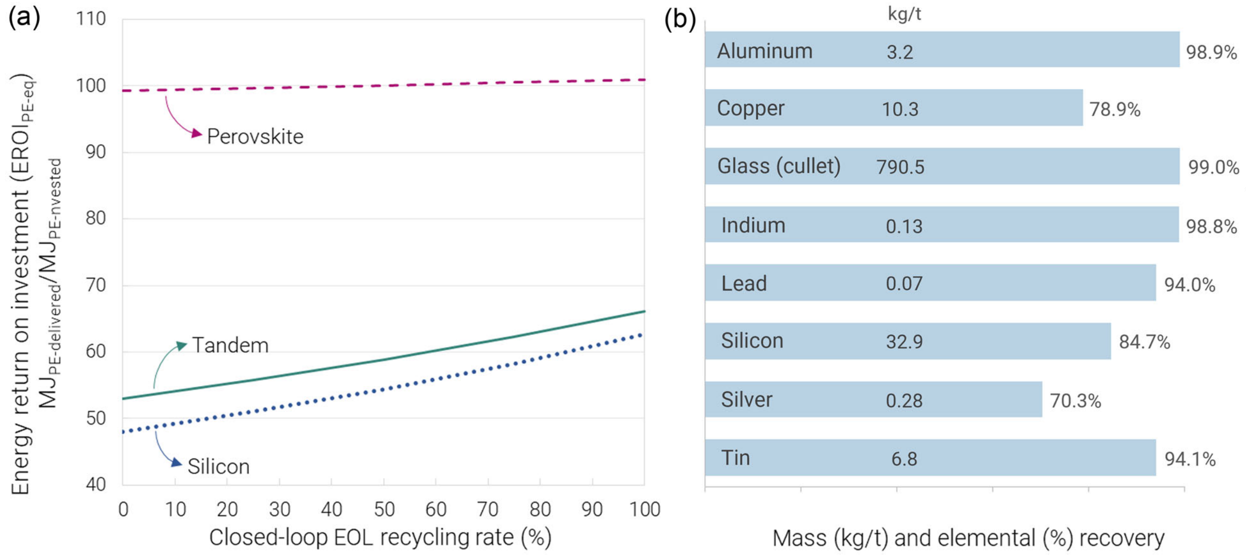

FIGURE 3 (a) Energy return on energy investment ($EROI_{PE\text{-}eq}$) and (b) end-of-life (EoL) recoveries as a percentage of the element entering the recycling process and as the mass recovered per tonne of EoL modules recycled. Note that the recoveries are independent of the recycling rate. Tin and lead are recovered as oxides. $EROI_{PE\text{-}eq}$ values have been normalized to an average irradiation of 1700 kWh/(m^2 year), PR of 0.75, lifetime of 30 years, and a grid efficiency (η_{grid}) of 0.30. Underlying data can be found in Supporting Information S2.

Direct comparisons of PV systems are notoriously challenging because of the number of cell material combinations and configurations, deposition methods, electricity inventories, different system boundaries, and methodological assumptions, among others. Nonetheless, we have normalized results as described in each case to enable valid comparisons. All comparisons are based on our results at a closed-loop recycling rate of zero to ensure alignment with other studies.

3.1 | Resource efficiency

3.1.1 | Energy generated for energy invested

Figure 3a depicts $EROI_{PE\text{-}eq}$ as a function of circularity. We calculate 48, 53, and 99 for the silicon, tandem, and perovskite systems without recycling, respectively. Fthenakis and Leccisi (2021) recently reported a monocrystalline-Si $EROI_{PE\text{-}eq}$ of 38 (34 normalized), and Jia et al. (2021) 32 (39 normalized). Our more optimistic 48 is expected because of a higher PCE (21.7% vs. 20.5% and 20.2%) and potentially lower estimate of energy consumption in the background system. Variations of perovskite cell configurations are many (Li et al., 2021), which adds to the difficulty of like-for-like comparisons. We exclude BoS components to align with other studies and calculate an $EROI_{PE\text{-}eq}$ of 187, which lies between the normalized 223 and 155 calculated from Ibn-Mohammed et al. (2017) and Tian et al. (2021) who modeled fairly similar modules. We have not found published $EROI_{PE\text{-}eq}$ values for four-terminal tandems.

Between not recycling at all and recycling all recovered Si, $EROI_{PE\text{-}eq}$ increases by 30% and 25% for the silicon and tandem systems, respectively. The amount of Si returned to the life cycle strongly influences power consumption in the silicon and tandem life cycles. When returned at solar grade, both the metallurgical- and solar-grade Si production processes are bypassed (see Figure 2). In bypassing these processes, their high energy consumptions are avoided. This is not relevant in the perovskite system, as it does not make use of Si. However, a 1.6% increase in $EROI_{PE\text{-}eq}$ is still observed. This is attributed to the generation of electricity from heat recovered during recycling, which reduces net power consumption. Regardless of recycling rate, the perovskite system's return is considerably higher than that of both the tandem and silicon systems, because its production energy investment is at least 60% lower based on our calculations.

3.1.2 | Recovery of valuable and hazardous materials

Figure 3b shows the recoveries of key elements, that is, the maximum amount of each element that can be returned to the same or similar life cycle in a closed loop, or sold for use in a different application. Note that recovery rates are independent of recycling rate as they are expressed per tonne

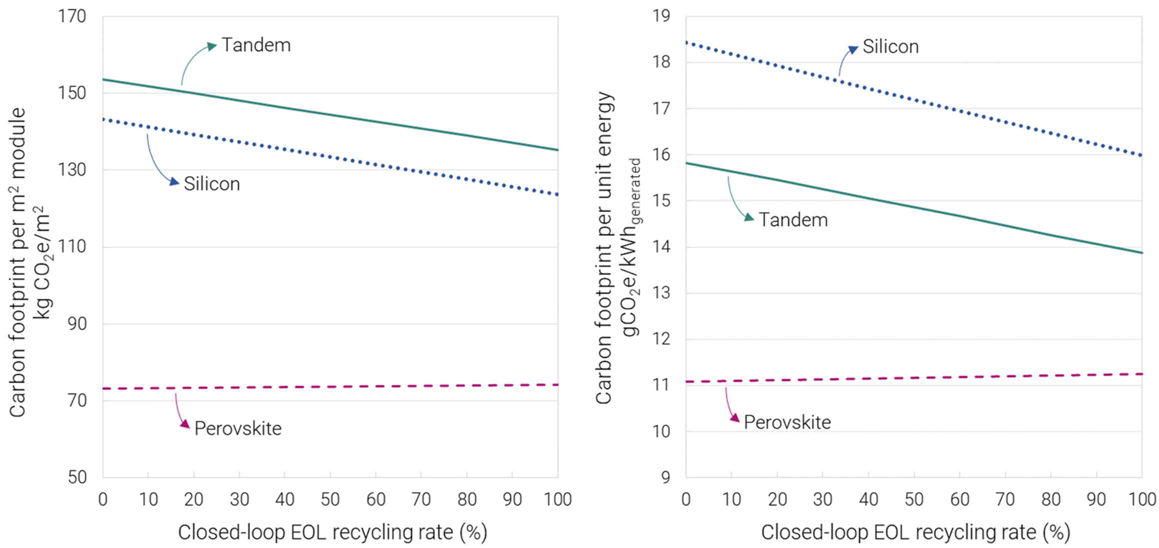

FIGURE 4 Carbon footprints (a) per m^2 module produced, and (b) per lifetime energy generated, normalized to an average irradiation of 1700 kWh/(m^2 year), PR of 0.75, and a lifetime of 30 years. Breakdowns of Scope 1, 2, and 3 emissions can be found in Supporting Information S1 (Section S3, Figure S2) and all underlying data in Supporting Information S2.

of modules recycled. Even at a closed-loop Si recycling rate of 100%, 84.7% of the Si entering the recycling process is returned, while the remainder is lost. Thus, even with total circularity, the Si material loop cannot be completely "closed."

3.2 | Carbon footprint

Life cycle carbon footprints are depicted in Figure 4 for the two functional units described in Section 2.3.

We estimate 143, 73, and 153 $kgCO_2e/m^2$, and 18.4, 11.4, and 15.8 gCO_2e/kWh for the silicon, perovskite, and tandem systems without closed-loop recycling, respectively. Our 18.4 gCO_2e/kWh for Si is in line with the normalized 21.3 and 16.7 gCO_2e/kWh calculated from Jia et al. (2021) and Lunardi et al. (2018), respectively. Fthenakis and Leccisi (2021) reported a higher 23 gCO_2e/kWh (normalized to 26) due to their larger system boundary. We also find reasonable agreement with previous perovskite studies. Again excluding BoS components, we calculate a footprint of 3.1 gCO_2e/kWh, compared with 2.0 (Ibn-Mohammad et al., 2017), 4.9 (Gong et al., 2015), and 4.9 reported by Tian et al. (2021). We have not found published footprints for four-terminal tandems.

Because the four-terminal tandem is a straight-forward combination of perovskite and silicon modules, its higher manufacturing footprint (Figure 4a) is expected. Including lifetime performance (Figure 4b), the tandem's higher PCE compensates for the increased manufacturing emissions to lower its footprint to below that of the silicon reference. Compared to silicon, the same amount of energy will be delivered by a smaller tandem system in a given time period, reducing resource consumption and impacts. The silicon and tandem system footprints decrease with increasing circularity, again because of avoiding two Si production steps and the associated Scope 2 emissions. Contrary to Tian et al. (2021), we find the perovskite system's footprint to worsen with increased circularity, which comes down to the choice of recycling process. The incineration of encapsulation and backsheet materials in our process causes a net increase in emissions—while power generated from recovered heat reduces net Scope 2 emissions, it is not enough to compensate for the increase in direct CO_2 emissions from incineration. Tian et al. (2021) assumed selective layer dissolution to recover substrates and other components but did not specify the recycling treatment applied for de-encapsulation, as it was likely not the focus of their study.

Figure 5 depicts the sensitivity of carbon footprint to system lifetime and PCE for the three systems. The labeled datapoints represent the footprints at the respective reference PCE and lifetime. At constant PCE, the perovskite and tandem footprints remain lower than the silicon system's 18.4 gCO_2e/kWh at lifetimes down to 18.1 and 25.5 years, respectively. With a 30-year lifetime, the tandem system footprint is less than that of the silicon reference at PCEs down to 23.4%. The minimum perovskite PCE would be less than the 16% lower limit shown.

Considering the constant CO_2e emission contours in Figure 5, the area above any given contour is greatest in the perovskite system, followed by the silicon and tandem systems. Qualitatively, this could be interpreted to mean that it would be least challenging to achieve lower footprints in the perovskite system and most challenging in the tandem system if PCE and lifetime are the only factors considered. Important, however, is that the full ranges of lifetime and PCE are not available in all three systems. For instance, if the recent 21.1% perovskite PCE (Liu et al., 2022), and the 22.2% and 28% expected for monocrystalline Si and tandems, respectively (VDMA, 2021), are taken as upper limits (the dashed vertical lines), it is

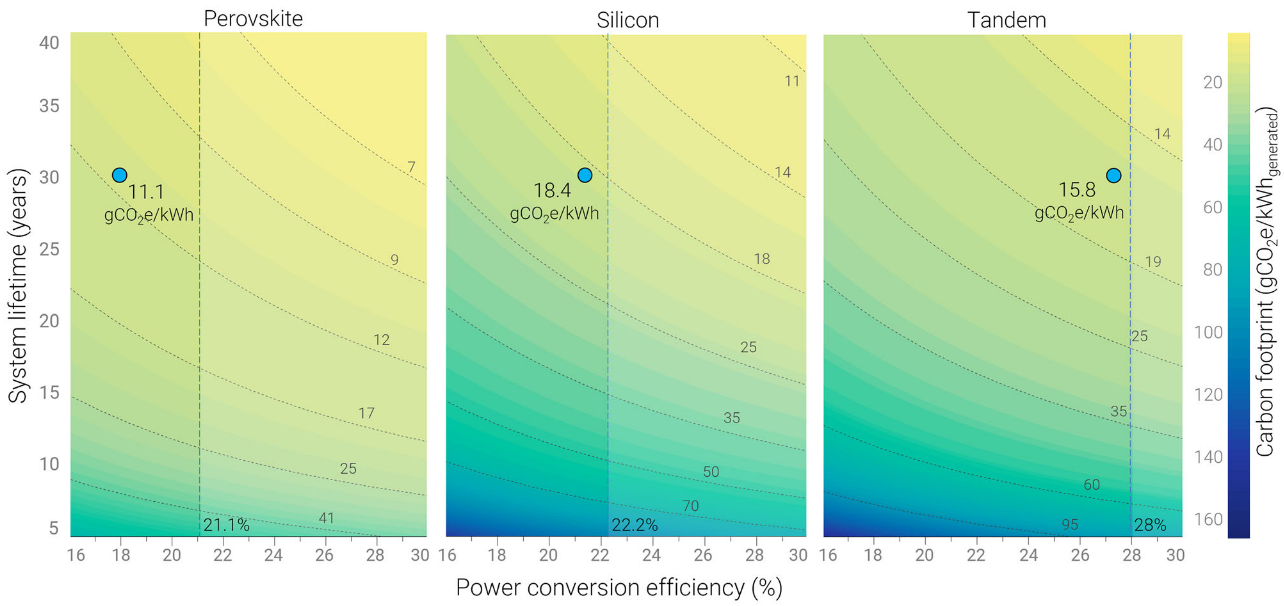

FIGURE 5 The sensitivity of carbon footprint to system lifetime and power conversion efficiency (PCE) for the case with no recycling. The labeled datapoints show the footprint for each system at its reference PCE (18%, 21.7%, and 27.3% for perovskite, silicon, and tandem systems, respectively) with a 30-year lifetime, annual irradiation of 1700 kWh/(m^2 year), performance ratio of 0.75, and an annual relative degradation rate of 0.5%. The dashed vertical lines indicate current perovskite (21.1%) (Liu et al., 2022) and projected Si (22.2%) and tandem (28%) efficiencies (VDMA, 2021). Underlying data can be found in Supporting Information S2.

clear that footprints lower than the reference can be achieved in both the perovskite and tandem systems. Although lifetime limits may currently exist, we assume that continued development would result in perovskite lifetimes similar to that of current commercial technologies.

3.3 | Techno-economic assessment and interaction with carbon footprint

Figure 6 depicts the sensitivity of LCOE to lifetime and PCE.

Our reference LCOEs are 3.92, 4.82, and 4.48 c/kWh for the perovskite, silicon, and tandem systems, respectively. The associated MSPs are 0.20, 0.31, and 0.38 $/Watt, respectively. As expected, our values agree with the 0.21, 0.32, and 0.36 reported by Liu et al. (2020) and Sofia et al. (2020) as our assumptions are closely aligned with those studies. Perovskite LCOE remains below that of the silicon reference if its lifetime exceeds 17.6 years compared to 18.1 years for a lower carbon footprint. A perovskite lifetime greater than 18.1 years would, therefore, give it both environmental and economic advantage over the silicon system. The same applies in the tandem system at the reference PCEs—a lifetime greater than 25.5 years is needed, and carbon footprint is the deciding factor (cf. 23.7 years for a lower LCOE). Alternatively, if 30-year lifetimes can be guaranteed, tandem LCOE will remain below that of the silicon system down to PCEs of 24.8% (compared to 23.4% for carbon footprint). Here, LCOE is the deciding factor—at PCEs above 24.8%, the tandem outperforms the silicon system in both the economic and environmental dimensions. As before, the minimum perovskite PCE would be less than the 16% lower limit shown.

As mentioned, the full ranges of lifetime and PCE are not technically achievable in all systems. There may be additional thresholds beyond which a system would not be considered a sound investment or environmentally acceptable. The sustainability of a particular system can only be maximized where the feasible operating windows bounded by these thresholds overlap, that is, within the bounds of all technical, environmental, and techno-economic limits.

Figures 7a and 7b, respectively, show the variation of LCOE and carbon footprint with circularity.

The increase in recycled content brought about by closed-loop recycling lowers silicon system LCOE (Bartie et al., 2021b). In the perovskite and tandem systems, on the other hand, increased circularity increases LCOE. As explained, the perovskite system does not benefit from the direct displacement of an energy-intensive and expensive raw material that would contribute significantly to lowering power consumption and cost, such as high-grade Si. In essence, the four-terminal tandem system represents the net effect. Viewed in isolation from a profit-only perspective, increased circularity seems unfavorable in the perovskite and tandem systems. From a sustainability perspective, however, Figures 7a and 7b together show that, in the tandem system, a trade-off exists between LCOE and carbon footprint with respect to circularity—increased circularity brings about lower CO_2e emissions, while the delivered energy becomes more expensive. Therefore, all other things being equal, an optimum level of Si circularity exists that minimizes both cost and environmental impact.

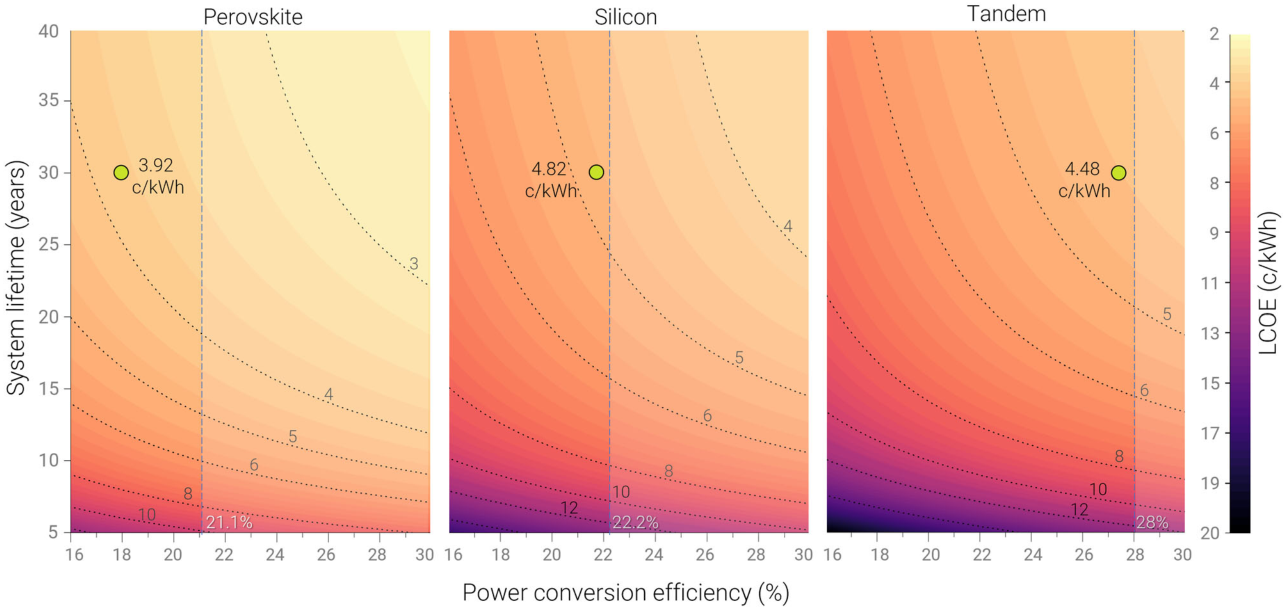

FIGURE 6 The sensitivity of levelized costs of electricity (LCOE) to system lifetime and power conversion efficiency (PCE) with no-recycling case. The labeled datapoints indicate the LCOE for each system at its reference PCE (18%, 21.7%, and 27.3% for the perovskite, silicon, and tandem systems, respectively) with a 30-year lifetime, annual irradiation of 1700 kWh/(m^2 year), performance ratio of 0.75, and an annual relative degradation rate of 0.5%. The dashed vertical lines show current perovskite (21%) (Liu et al., 2022) and projected Si (22.2%) and tandem (28%) efficiencies (VDMA, 2021). An assessment of the sensitivity of LCOE to overall recycling cost and discount rate can be found in Supporting Information S1 (Section S2 and Figure S1) and all underlying data in S2.

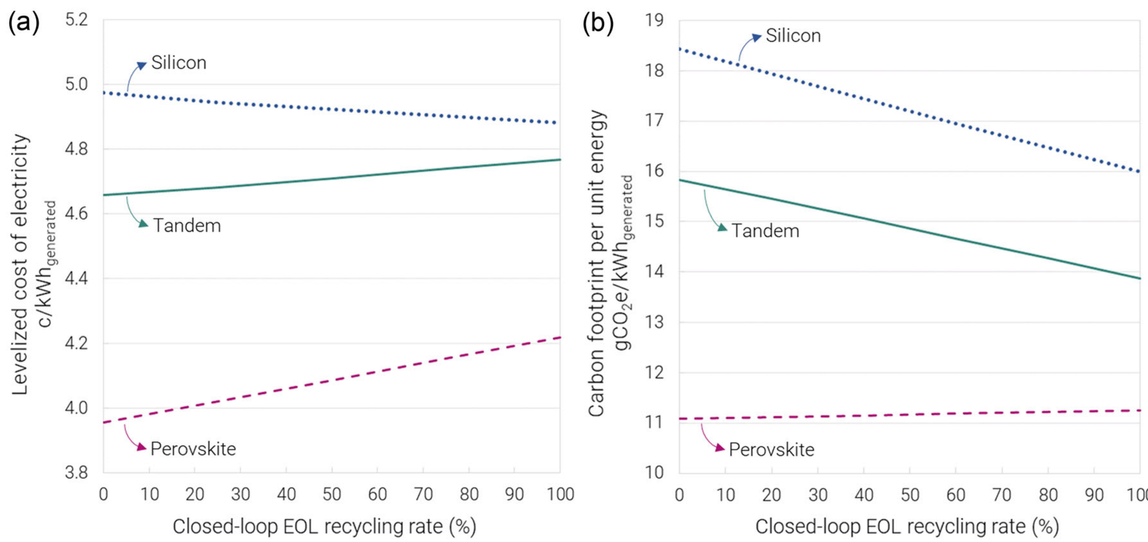

FIGURE 7 The variation of (a) levelized costs of electricity and (b) CO_2e emissions with end-of-life recycling rate. Figure 4b is repeated here as 7b for convenience. Underlying data can be found in Supporting Information S2.

3.3.1 | Carbon tax

Carbon taxation is generally considered an effective policy measure for stimulating emission reductions. Our approach facilitates estimation of the effects such a tax might have if imposed on the energy sector. Figure 8a shows the effects of hypothetical taxes (between 25 and 400 \$/tCO_2e emitted) and circularity on tandem LCOE. The public revenue stream generated by the tax increases LCOE, as expected. Increased circularity at the low tax has little effect—the \$25 line is almost parallel to the zero-tax line. As the tax increases, line slopes become less positive, and eventually negative, above the breakeven tax rate. Below the breakeven rate, increased circularity always increases cost, but as the tax rate approaches the breakeven rate, the cost increase becomes less pronounced, that is, increased circularity softens the tax's cost-increasing effect. Above the breakeven rate,

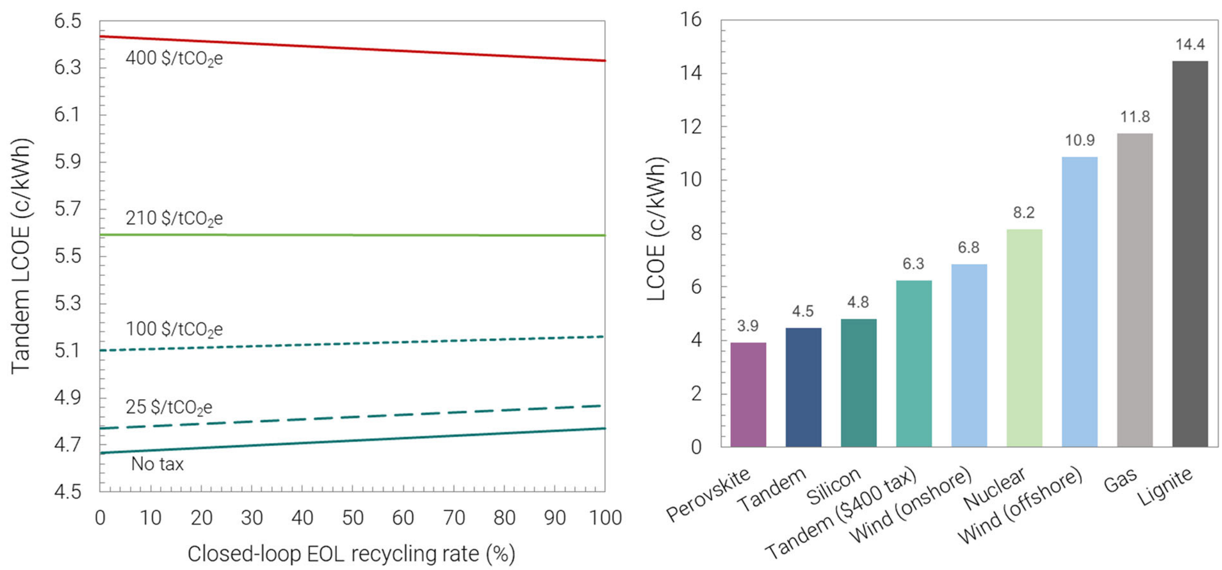

FIGURE 8 (a) The combined effects of recycling and carbon tax on levelized costs of electricity (LCOE) in the tandem system, and (b) a comparison of photovoltaic LCOE with that of other electricity sources (Fraunhofer-ISE, 2021; EIA, 2022 for nuclear). Underlying data can be found in Supporting Information S2.

increased circularity always reduces cost, and this effect becomes stronger the higher the tax. Based on our assumptions, the breakeven tax is $210/t$CO_2$e. With this value significantly above any current, for example, Sweden's $134/t$CO_2$e (Sweden Ministry of Finance, 2021) or predicted tax rates (Jaumotte et al., 2021), it is highly unlikely that closing materials loops alone would be enough to reverse tax-induced cost increases. Despite these findings, Figure 8b shows that, even at $400/t$CO_2$e, PV LCOEs remain below that of other electricity sources with no taxes applied.

3.3.2 | The contribution of transport to carbon footprint and MSP

With PV supply chains being globally distributed, it is worth investigating the impacts and costs of material and product movement across the globe. We estimated the contribution of transport to carbon footprint and MSP for various combinations of manufacturing and recycling location for the tandem system. Results are presented in Supporting Information S1 (Section S4 and Figure S3).

4 | DISCUSSION

An important aspect of this work is the method by which the inventory data were generated and the amount of detail included. Contrary to methods that account for material streams as if they are flowing through a system as pure elements, process simulation accounts for the flows of the compounds and solutions actually present, also including their thermochemical properties. The rigorous mass and energy balances calculated as a result allow each flow in the system to be quantified in terms of mass and energy (for enthalpy and entropy) units per unit of time, so that all flows and thermodynamic losses from the system can be quantified in the same units as the energy delivered by the energy carrier, in this case PV electricity.

Our results have shown that the perovskite system is the best performer in terms of our indicators for resource efficiency ($EROI_{PE\text{-}eq}$), environmental impact (CO_2e emissions), and techno-economic performance (LCOE), with the tandem in second place, and the silicon system in third, regardless of the degree of circularity. The caveat is that the perovskite and tandem systems achieve the same long lifetimes current commercial technologies do. The perovskite and tandem lifetimes must exceed 18.1 and 25.5 years, respectively, to outperform the silicon system both environmentally and techno-economically.

Compared to the Si system, the tandem will be more effective at enhancing the sustainability of whichever life cycle system consumes the energy it delivers, because both the embodied carbon footprint and the levelized cost of that energy will be lower, and a system 20% smaller in physical size would deliver a given amount of energy within a fixed period. While the perovskite system's footprint and LCOE are considerably lower, a 21% larger system would be needed.

Although recycling increases *direct* (Scope 1) emissions in all systems, the increased consumption of high-quality recycled Si considerably reduces electricity consumption in the silicon and tandem systems. The reduction in associated *energy-related* (Scope 2) emissions compensates for the direct emissions added through recycling several times over. Importantly, this is conditional upon the further development and commercialization of recycling processes that recover Si at solar grade. There is no similar benefit in the perovskite system because of the absence of large quantities of input materials as energy intensive as Si. While the recovery and recycling of intact glass substrates appear to be a promising option for perovskite recycling (instead of downcycling glass into cullet), a recent analysis of 13 potential approaches revealed that all but one are environmentally *more* detrimental than using virgin coated glass, mainly because of the solvents needed for delamination (Rodriguez-Garcia et al., 2021). However, the perovskite system's footprint is already 30%–40% lower than that of the silicon, and 19%–30% lower than that of the tandem system.

Increased circularity in the silicon system is beneficial in terms of both carbon footprint and LCOE, while the opposite is true for the perovskite system. In the tandem system, the trade-off that emerges indicates that an optimum level of circularity exists at which both cost and environmental impact will be minimized, but with compromises in both dimensions. The implication is that total circularity does *not* deliver the most sustainable outcome in this case, highlighting the importance of the assertion that circularity does not automatically come with an overall sustainability guarantee (Korhonen et al., 2018). The advantage of our approach is the use of fully aligned inventory data to calculate the environmental and techno-economic indicators as a function of circularity, so avoiding the introduction of additional uncertainty resulting from potential data and system boundary inconsistencies.

However, to gain a more complete picture of interactions within the system, future work should investigate further options and constraints that may influence cost and footprint. For instance, while we found the perovskite footprint to increase with circularity, Tian et al. (2021) reported the opposite. In this example, recycling process design and its associated costs and impacts play key roles in creating, modifying, or undoing any sustainability trade-offs. Results from studies such as that by Rodriguez-Garcia et al. (2021) mentioned earlier should be incorporated in future life cycle simulations to analyze the system-wide effects of different production and recycling approaches. Another example is the potential revenue from recycled products. All processes have to operate within economic, societal, and environmental impact constraints to be viable. Also, depending on the supply and demand for recycling and the products from recycling, this may involve increased focus on the recovery of certain elements at the expense of others.

The linked resource and economic models also allowed us to quantify the potential cost effects of carbon taxation, while investigating the role of closed-loop recycling in modifying these effects at the same time. We found that, besides clear environmental benefits in the tandem system, recycling dampens the tax's cost impact. Therefore, increasing the recycled content in PV modules alone would not be expected to fully compensate for the cost impact of taxation. Additional measures upstream in the supply chain and higher up in the CE material hierarchy (e.g., by reducing consumption, re-using, and refurbishing) are needed before recycling becomes inevitable. If this occurs with overall sustainability, rather than merely cost reduction in mind, the tax will have the intended effect. It could, of course, also be counteracted with measures that reduce cost but not emissions, such as cross-border carbon leakage for which other mechanisms like border tax adjustments need to be implemented. From a business perspective, however, the lower perovskite and tandem LCOEs provide more room to move in terms of margins and investment in emission reduction and/or energy storage technologies relative to the silicon reference.

There are limitations associated with the obtained results. Power consumption has a significant impact on carbon footprint as Scope 2 emissions and is therefore sensitive to location-specific grid compositions. Although PV supply chains are typically globally distributed, all results presented in this paper are based on the German electricity mix for the sake of simplicity. Results are based on static simulations—we have not considered potential evolution of the electricity mix or innovations in manufacturing technologies and efficiencies over time. Although standard methods have been used for recycling cost estimates, they should be seen as preliminary, as none of the recycling processes exists commercially. While we only quantified carbon footprint and considered the risk of burden shifting to be low, future assessments should include other impact categories to examine whether other trade-offs exist. We considered two of the sustainability dimensions in this study. To gain a comprehensive understanding of overall sustainability, this approach will be expanded to include the effects of societal impacts.

5 | CONCLUSION AND OUTLOOK

PV has a key role to play in the decarbonization of future energy systems, but its full potential will only be realized once PV life cycles achieve sustainable circularity. To confirm whether this is occurring, reliable methods are needed to assess sustainability and how it is influenced by circularity. We presented a novel approach that uses process simulation to generate physics-based inventory data that comply with the laws of mass conservation, and the first and second laws of thermodynamics, as opposed to elemental flows based on linear input–output transformations. By linking simulations with bottom-up cost models, we evaluated and compared the resource efficiencies, carbon footprints, and LCOEs of three contemporary PV technologies. Direct evaluation of the simultaneous, system-wide effects of circularity, PCE, system lifetime, and carbon taxation on the selected sustainability indicators are a further novelty, which are facilitated by NN-based surrogate functions that serve as proxies for simulation results. Assessments of resource efficiency, and environmental and techno-economic performance, as well as the effects of policy measures can, therefore, all be conducted within the same framework from a consistent foundation of physics-based inventory data. As a result, potential trade-offs among the sustainability dimensions and in relation to CE strategies can be identified and quantified with a view to maximizing overall sustainability.

With rapid technological development in the PV and other industries that make use of complex material combinations to achieve required functionalities and efficiencies, agile approaches that ensure data consistency and adherence to the laws of conservation and thermodynamics—such as that presented in this paper—are needed. This rigorous quantification of life cycle mass and energy flows (including thermodynamic losses) provides the true starting points and performance assessments along development paths aimed at increasing overall sustainability.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are openly available in Zenodo at https://doi.org/10.5281/zenodo.7102103, reference number 7102103, and in the supporting information (S1 and S2) of this article.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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Annexure E: Additional publications

Table E-1 lists additional publication related to this work, but not included as part of the dissertation.

Table E-1: Details of articles and other publications related to, but not included in this dissertation

1	Reuter, M. A., van Schaik, A., Gutzmer, J., Bartie, N. J., & Abadías-Llamas, A. (2019). Challenges of the Circular Economy: A Material, Metallurgical, and Product Design Perspective. *Annual Review of Materials Research*, 49(1), 253–274. https://doi.org/10.1146/annurev-matsci-070218-010057	
	Type	Journal article
	Contribution	Conceptualisation, investigation, writing—original draft, review, and editing
2	Abadías Llamas, A., Bartie, N. J., Heibeck, M., Stelter, M., & Reuter, M. A. (2020). Simulation-Based Exergy Analysis of Large Circular Economy Systems: Zinc Production Coupled to CdTe Photovoltaic Module Life Cycle. *Journal of Sustainable Metallurgy*, 6(1), 34–67. https://doi.org/10.1007/s40831-019-00255-5	
	Type	Journal article
	Contribution	Conceptualisation, methodology, validation, investigation, writing—review and editing, visualisation, supervision
3	Bartie, N. J., & Reuter, M. A. (2020). Quantification of the Inconvenient Truths about the Circular Economy (CE): Digital twinning of very large systems. *FIfF-Kommunikation,* 3/2020. Bremen. Forum Informatiker*Innen für Frieden und gesellschaftliche Verantwortung e.V. (FIfF). https://www.fiff.de/publikationen/fiff-kommunikation/fk-2020/fk-2020-3/fk-2020-3-content/fk-3-20-p43.pdf	
	Type	Article in a trade magazine
	Contribution	Conceptualisation, writing—original draft, review and editing, visualisation
4	Bartie, N. J., Heibeck, M., Abadías Llamas, A., Reuter, M. A. (2019, December 8). Metal interactions and symbioses: optimization of quality, quantity, and sustainability. Vienna University of Technology. *5th International Conference on Final Sinks*, Vienna, Austria. http://www.icfs2019.org	
	Type	Conference presentation
	Contribution	Conceptualisation, methodology, validation, investigation, formal analysis, writing—original draft, visualisation

5	Bartie, N. J., & Reuter, M. A. Process metallurgy in circular economy system design: challenges & solutions. In *Proceedings of the 58t Annual Conference of Metallurgists (COM) hosting the 10th International COPPER CONFERENCE 2019*. Canadian Institute of Mining, Metallurgy and Petroleum.	
	Type	Conference paper
	Contribution	Conceptualisation, investigation, writing—original draft, review and editing, visualisation
6	Abadías Llamas, A., Valero, A., Bartie, N. J., Stelter, M., & Reuter, M. A. (2019). Process metallurgy in circular economy system design: the copper and base metal value chain. In *Proceedings of the 58t Annual Conference of Metallurgists (COM) hosting the 10th International COPPER CONFERENCE 2019*. Canadian Institute of Mining, Metallurgy and Petroleum.	
	Type	Conference paper
	Contribution	Writing—review and editing, visualisation
7	Abadías Llamas, A., Bartie, N. J., Heibeck, M., Stelter, M., & Reuter, M. A. (2020). Resource Efficiency Evaluation of Pyrometallurgical Solutions to Minimize Iron-Rich Residues in the Roast-Leach-Electrowinning Process. In A. Siegmund, S. Alam, J. Grogan, U. Kerney, & E. Shibata (Eds.), The Minerals, Metals & Materials Series. PbZn 2020 [electronic resource]: *The 9th International Symposium on Lead and Zinc Processing* / A. Siegmund, S. Alam, J. Grogan, U. Kerney, E. Shibata, editors (pp. 351–364). Springer. https://doi.org/10.1007/978-3-030-37070-1_31	
	Type	Conference paper
	Contribution	Conceptualisation, methodology, validation, investigation, formal analysis, writing—review and editing
8	Bartie, N. J., Cobos-Becerra, L., Fröhling, M., Schlatmann, R. & Reuter, M. A. (2021, June 23). Process simulation for comprehensive sustainability assessment of the silicon photovoltaic life cycle. Minerals Engineering International. *Sustainable Minerals '21*, Online. https://mei.eventsair.com/sustainable-minerals-21/	
	Type	Conference presentation
	Contribution	Conceptualisation, methodology, software, validation, formal analysis, investigation, writing—original draft, review & editing, visualisation

9	Bartie, N. J., Cobos-Becerra, L., Fröhling, M., Reuter, M. A., & Schlatmann, R. (2021, June 20 - 2021, June 25). Process simulation and digitalization for comprehensive life-cycle sustainability assessment of Silicon photovoltaic systems. In *2021 IEEE 48th Photovoltaic Specialists Conference (PVSC)* (pp. 1244–1249). IEEE. https://doi.org/10.1109/PVSC43889.2021.9518984	
	Type	Conference presentation and paper
	Contribution	Conceptualisation, methodology, software, validation, formal analysis, investigation, writing—original draft, review & editing, visualisation
10	Bartie, N. J., & Reuter, M. A. (2021). The link between photovoltaics, sustainability, and the metals industry. In *IMPC2020: XXX International Mineral Processing Congress* (pp. 3733–3746). South African Institute of Mining and Metallurgy.	
	Type	Conference paper
	Contribution	Conceptualisation, methodology, software, validation, formal analysis, investigation, writing—original draft, review & editing, visualisation