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A review of life cycle assessment and sustainability analysis of perovskite/Si tandem solar cells

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Perovskite/silicon (Si) tandem solar cells (TSCs) have emerged as a promising candidate among PV technologies due to their capability to greatly increase power conversion efficiency (PCE) exceeding the Shockley–Queisser limit of single-junction solar cells. Nevertheless, obstacles to the durability of perovskite materials and the environmental consequences of their life cycle present notable barriers to their widespread commercial deployment. The objective of this article is to deliver a review of life cycle assessment (LCA) and sustainability analysis of perovskite/Si TSCs: first, focusing on their working principle, configuration, components and recent progress and then presenting an overview of the LCA and sustainability study performed on perovskite/Si TSCs. Finally, this review highlights important directions for future LCA and sustainability studies required for the successful development of this remarkable perovskite/Si TSC PV technology.

Sustainability spotlight

Photovoltaics play a key role because of their lack of greenhouse gas emissions during operation. However, considering the entire life cycle, PV modules can still have significant environmental impacts depending on the technology being used. Enhancing the power conversion efficiency of solar cells/modules is essential for improving both the environmental sustainability and energy performance of PV systems. The emerging perovskite/silicon tandem solar cells provide an opportunity to upgrade the present market-dominating single-crystal silicon (c-Si) technology. This review aims to present the life cycle assessment and sustainability of perovskite/silicon tandem solar cells while focusing on their criticality. Aligned with UN SDG 7 for affordable and clean energy, it promotes renewable development for a more sustainable PV technology for the future.

1. Introduction

The transition to renewable energy is driven by the urgent need to tackle environmental issues caused by fossil fuel combustion, where photovoltaics (PVs) are significant because they do not emit greenhouse gases during operation. These substantial and remarkable contributions account for a global PV capacity of 1.6 TW in 2023, a 400 gigawatt (GW) increase from 2022.[1] The rapid and extraordinary increase in PV capacity witnessed over the past decade can be attributed to the rising investments being made in renewable energy sources, driven by the downward trajectory in costs, continual technological advancements, and the implementation of supportive policies that are specifically designed to address the issues surrounding climate change and to reduce the dependency on conventional fossil fuels. The fast-tracked deployment of PV technology, combined with broad electrification efforts, offers the possibility of a 21% decline in worldwide carbon dioxide (CO_2) emissions by 2050,

where PV could account for up to 25% of the global electricity supply.[1,2] To achieve this objective, the global solar energy capacity needs to exceed multiple terawatts, propelled by diminishing expenses, technological advancements, and augmented financial backing, notably in the Asian region.[3] Although electricity generation by PV systems is free of greenhouse gas emissions, considering the entire life cycle, PV modules can still have significant environmental impacts depending on the technology being used. Enhancing the power conversion efficiency (PCE) of solar cells/modules is essential for improving both the environmental sustainability and the energy performance of PV systems.[4]

Tandem solar cells (TSCs), or multi-junction solar cells, represent an advanced approach in PV technology. Unlike single-junction solar cells, which utilize a single pn junction to absorb sunlight and convert it into electricity, TSCs are stacked with two junctions of semiconductors with different bandgaps.[5] Each junction is customized to absorb a specific segment of the solar spectrum, thereby reducing energy losses resulting from the thermalization/relaxation process and spectrum losses from below-bandgap photons, which allows TSCs to achieve higher overall efficiencies by harnessing a broader range of wavelengths from sunlight.[6,7] Previously, the most successfully developed TSCs were based on GaInP/GaAs/GaInAs, with

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a champion efficiency of 39.5%.[8] III–V-based TSCs have already been commercialized and are mainly used for space applications. In recent years, perovskite/silicon tandem solar cells (perovskite/Si TSCs) have made a breakthrough in the PV community, impressed by the rocket-like rise of their efficiency to 34.6% reported by LONGI.[9] Moreover, a perovskite/Si TSC provides an opportunity to upgrade the present market-dominating single-crystal silicon (c-Si) solar cell, which is limited by its theoretical efficiency to around 29%, and thus it acts as a collaborator instead of a competitor for the existing c-Si PVs, making it easily accepted from the point of view of commercialization. Perovskite/Si TSCs are designed to capture a wide range of the solar spectrum, with the absorption of high energy photons by the perovskite top cell and low energy photons by the c-Si bottom cell. Here, wide-bandgap metal halide perovskites (MHPs) are used as the top cell. MHPs are characterized as a material family possessing the perovskite structure, wherein the inorganic framework consists of a bivalent metal, typically lead (Pb) or tin (Sn), or a combination of both, along with halides, while the organic molecules are situated between the octahedra of the inorganic framework.[10–12]

Life cycle assessment (LCA) is a structured, comprehensive method for quantifying material- and energy-flows and their associated emissions caused in the life cycle of goods and services. The LCA of perovskite/Si TSCs reveals a lower carbon footprint and shorter energy payback time (EPBT) compared to conventional single-junction solar cells due to their higher efficiency, despite challenges related to material stability and toxicity that necessitate ongoing research and development for optimal sustainability.[13,14] As perovskite/Si TSCs advance technologically, studies to estimate the environmental impact of commercially manufactured devices have become necessary, which have relied primarily on process data from laboratories and test sites.[15,16] There are differences in assumptions regarding the configuration of industrial manufacturing processes and how long perovskite/Si TSC modules will last under certain conditions. These factors make it difficult to predict outcomes based solely on controlled experimental designs, leading to high variability between studies. This review aims to present an overview of recent studies on the LCA and sustainability of perovskite/Si TSCs while focusing on their criticality. We first introduce perovskite/Si TSCs in terms of structure, operation principles, and materials and then look at recent enhancements and compare the efficiency among single-junction solar cells. The goal and scope are discussed, following which the inventory analysis, impact assessment, and interpretation of the LCA are presented for solar cell technologies. The review is about environmental considerations, such as using raw materials, production process, energy consumption, emissions, and disposal of the solar cell. Sustainability in the assessment includes consideration of various economic, social and environmental aspects, and benefits and risks of the organization in the long run. Finally, policy and markets should be considered to improve the overall sustainability analysis of perovskite/Si TSCs.

2. Overview of perovskite/Si tandem solar cells

The perovskite/Si TSC is a perfect example of mixing two different types of solar cells to take advantage of the best in both and achieve better PCE. As shown in Fig. 1(a), the main architecture of perovskite/Si TSCs predominantly comprises a perovskite solar cell situated on top of a c-Si bottom cell. The c-Si bottom cell can be in a structure of tunnel oxide passivated contact (TOPCon), heterojunction (HJT), or back contact (BC), which can be either double-side texturing or front-side polishing, depending upon the particular fabrication methodology employed. As such, the perovskite top cell is made with a p–i–n or an n–i–p structure. The perovskite absorbing layer has a wide bandgap to effectively absorb and convert high-energy photons (short wavelength light), while the underlying c-Si bottom cell captures and converts low-energy photons (longer wavelength light) (Fig. 1(b)). Usually, the bandgap for the top cell made from perovskite materials varies between 1.7 and 1.8 eV, which is ideal for absorbing the blue and green parts of sunlight, and also can be tuned.[19,20] On the other hand, c-Si in the bottom cell with an approximate band gap of 1.12 eV absorbs the red as well as near-infrared regions of sunlight most efficiently.[21,22] The perovskite top cell and c-Si bottom cell are interconnected either in series, necessitating current matching between the two sub-cells, or in parallel, which facilitates the independent functioning of the sub-cells. In this way, it reduces thermalization losses and maximizes the utilization of the solar spectrum to enhance the overall PCE. The recombination interface between the perovskite top cell and the c-Si bottom cell is a critical link that demands careful engineering to ensure efficient charge carrier extraction and minimal recombination losses. The architecture remains an area of interest for many researchers for the commercialization.

In practice, p–i–n type perovskite top cells are widely used in industry due to their good stability and high efficiency. As shown in Fig. 1(c), a p–i–n perovskite top cell consists of a hole transport layer (HTL), a perovskite light absorbing layer, an electron transport layer (ETL), a transparent conductive oxide (TCO) layer, and a metal grid. The record-efficiency perovskite/Si TSCs are constructed on TOPCon or HJT c-Si solar cells that possess textured surfaces characterized by pyramid heights typically ranging from 1 to 3 μm, which offer advantages in terms of improved light absorption.[23] This textured structure design leads to a significant enhancement in the overall performance, ultimately resulting in increased PCE.[24] However, the presence of this surface texture poses a challenge in the fabrication of highly conformal wide-band gap perovskite top cells through a traditional solution method with good performance.[25] Recently, the evaporation method and hybrid solution/evaporation method have been, therefore, proposed for the fabrication of perovskite top cells for high-efficiency perovskite/Si TSCs.

The choice of materials in perovskite/Si TSCs is not only highly selective to optimize performance, stability, and scalability, but also has an impact on the sustainability and the LCA
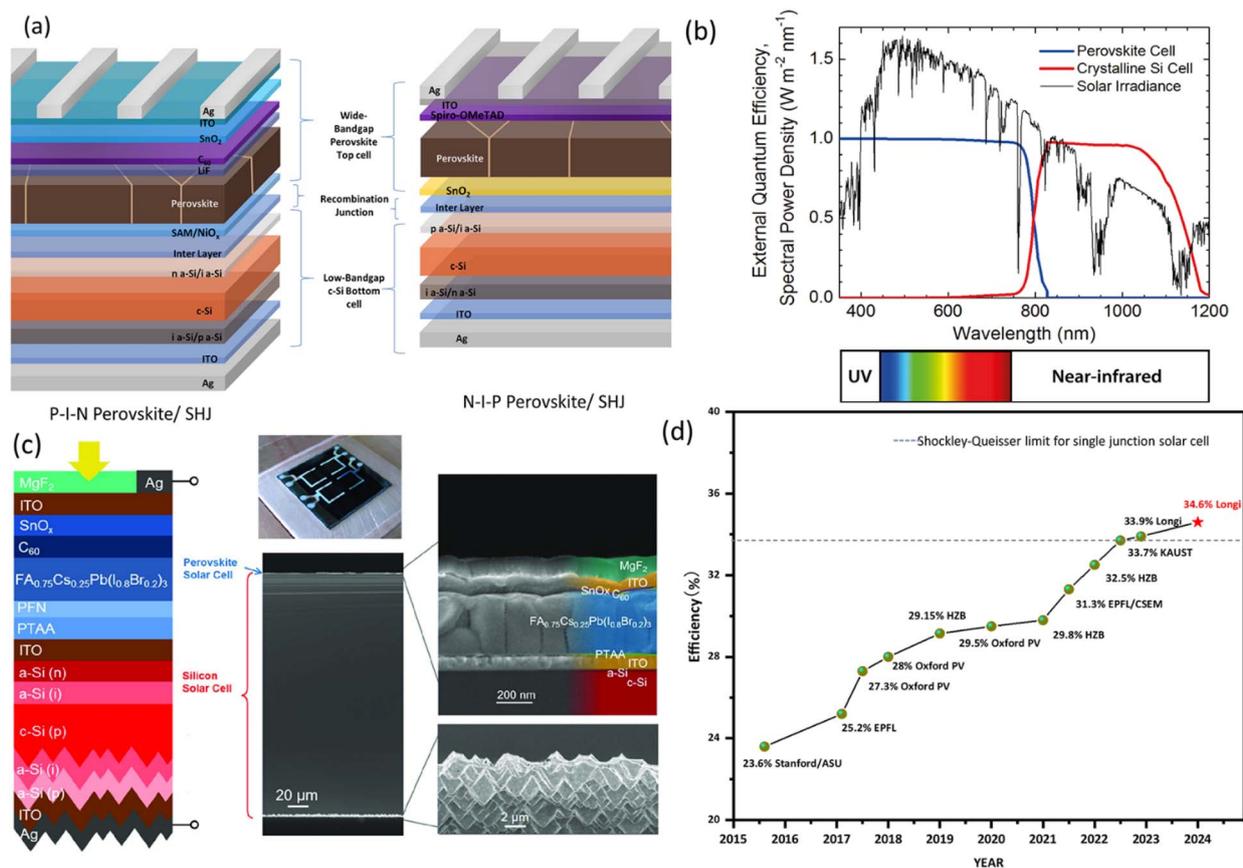
Fig. 1 (a) Schematic of N–I–P type and P–I–N type perovskite/Si TSCs. (b) Spectral response of perovskite top cell and c-Si bottom cell. Light enters through the perovskite cell, where mostly the visible part of the solar spectrum is absorbed. Near-infrared light is transmitted to the silicon cell where it is absorbed. Reprinted from Web https://www.epfl.ch/labs/pvlab/research/page-124775-en-html/.[17] (c) Schematic structure of monolithic perovskite silicon tandem solar cells combining a high bandgap perovskite top cell with a silicon heterojunction bottom cell. Photograph of a tandem substrate with four active cell areas. SEM image (cross section) of a tandem device with zooms of the perovskite top cell and the silicon rear-side texture, respectively. Reprinted with permission from Solar RRL copyright@Wiley library 2020.[18] (d) Efficiency evolution of perovskite/Si TSCs.

analysis. The use of various mixed cations (methylammonium (MA), formamidinium (FA), and cesium (Cs)) and mixed halides (iodine (I), bromine (Br), and chloride (Cl)) in perovskites has been investigated for wide bandgap perovskites.[26–28] For HTLs, NiO_x[29,30] or self-assembled monolayers (SAMs)[31,32] are often used in practice to facilitate hole extraction and transport. The interface between perovskite and silicon is critical to minimize recombination losses and extract charge. For ETLs, SnO_2 (ref. 33) and C_{60}/SnO_2 are commonly used as they have good electron mobility and band alignment with perovskites. Meanwhile, ITO is used for the front TCO as well as the tunnel recombination layer. In the case of an HJT c-Si cell, its materials are mainly single crystalline silicon and amorphous silicon, along with back contact metals like silver (Ag) or copper (Cu). Advanced encapsulation techniques using polyisobutylene (PIB),[34] ethylene-vinyl acetate (EVA),[35] glass or polymer barriers are used to ensure the long-term stability and durability of the tandem modules.[36]

In recent years, tremendous progress has been made in perovskite/Si TSCs with innovative material compositions and fabrication methods. In 2018, Sahli and his coworkers reported a perovskite/Si TSC using a pyramid textured commercial c-Si bottom cell and achieved a J_{sc} of 20.56 mA cm^{-2}.[37] In 2021, Roß et al. used co-evaporation and chemical bonding and achieved a PCE of 24.6%.[38] In 2023, Zhang et al. combined evaporation and spin-coating with buried-interface engineering and achieved a PCE of 28.4%.[39] Tockhorn et al. demonstrated the benefit of nanotextures, increased fabrication yield and efficiency and achieved a PCE of 29.8%.[40] Aydin et al. achieved a PCE of 32.5% by spin-coating perovskite layers on textured silicon cells.[41] Also, LONGI reported that they achieved a highest PCE of 34.6%.[9] These results show the fast progress and high potential of perovskite/silicon TSCs (Fig. 1(d)).

3. Life cycle assessment and environmental impact of perovskite/Si tandem solar cells

LCA is an organized process for examining the ecological effects associated with every step of a product's life span, starting with the gathering of natural resources and concluding with its

disposal, usually called a 'cradle-to-grave' assessment. The LCA methodology comprises four principal stages: goal and scope definition, which defines the aims and parameters of the evaluation; life cycle inventory analysis (LCI), wherein data regarding energy, material inputs, and environmental emissions are aggregated; life cycle impact assessment (LCIA), which analyzes the potential environmental impacts predicated on the inventory data; and interpretation, during which findings are examined to guide decision-making and pinpoint prospects for enhancement. Fundamental components analyzed in an LCA encompass material utilization, greenhouse gas emissions, water consumption, and waste production, yielding insights into resource efficiency and sustainability dilemmas.[42,43] It determines the resulting environmental emissions and assesses the total potential impacts on the environment to improve the environmental performance of the product. The procedures for executing LCA, which are widely acknowledged, are defined within the 14 000 series of environmental management standards established by the International Organization for Standardization (ISO), specifically in ISO 14040 and ISO 14044. ISO 14040 articulates the 'principles and framework' of the standard, whereas ISO 14044 outlines the 'requirements and guidelines'. In general, ISO 14040 was formulated for an audience engaged in administrative functions, whereas ISO 14044 was made for practitioners in the field.[44] The main purpose is to develop a detailed reference framework for adequately comparing a wide range of environmental impact indicators, including but not limited to carbon footprints, toxicity, water usage, land use, and resource depletion.[45,46] However, for LCA of PVs, the frameworks of ISO 14040 and ISO 14044 leave the individual expert with a range of choices that can affect the results and thus the conclusions of an LCA study. Additional standards and guidelines have later been published such as ISO 21930 (Environmental Product Declaration on Construction Products, International Organization for Standardization (ISO) 2017) and the Product Environmental Footprint Category Rules (PEFCR) for PV electricity (TS PEF Pilot PV 2018). In addition, IEA PVPS guidelines have been developed to offer guidance for consistency, balance, and quality to enhance the credibility of the findings from LCAs on photovoltaic (PV) electricity generation systems.

Previously, lots of studies have been carried out on the possible environmental impacts of various types of solar cells.[47] The International Energy Agency (IEA), for example, publishes every year the 'Life Cycle Inventories and Life Cycle Assessments of Photovoltaic Systems' through the Photovoltaic Power Systems Programme. This document contains important data about the inventories and impacts of commercial c-Si solar modules. There have also been studies on the LCA of perovskite single-junction solar cells.[48,49] At present, the investigation of the ecological consequences and LCA of perovskite/Si TSCs constitutes a crucial area of study that explores the sustainability of these advanced PV technologies. Nevertheless, the life cycle of perovskite/Si TSCs from the extraction of raw materials and manufacturing to installation, operation, and eventual disposal poses notable environmental hurdles that demand thorough examination and mitigation.

Fig. 2 shows the schematic illustration of a closed loop recycling process of perovskite/Si TSCs. Raw material extraction stands out as a notable contributor to the environmental impact of perovskite/Si TSCs, given that silicon constitutes the base cell requiring energy-intensive purification processes. The manufacturing of high-purity silicon involves the energy-demanding carbothermic reduction of silica, followed by purification techniques, which are highly energy-intensive and substantially contribute to carbon emissions.[50] In a recent study, the HJT c-Si bottom cell results in an extremely intensive primary energy consumption of $\sim$3534 MJ m^{-2} per module, which is fundamentally due to the substantial inputs in materials and energy during the fabrication of the bottom cell, especially the energy-intensive purification process of silicon,[51] while this value is 188 MJ m^{-2} for the wide bandgap perovskite top cell. Despite the lower temperatures necessary for depositing perovskite layers, the complete manufacturing process still entails substantial energy consumption, particularly in electricity, which may contribute to the carbon footprint if derived from non-renewable sources.[52]

Throughout the operational phase, the perovskite/Si TSC helps convert sunlight into electricity, thereby harvesting renewable solar energy. While traditional fossil fuel energy relies on burning carbon-rich fuels and produces large amounts of greenhouse gas emissions, the perovskite/silicon TSC generates electricity without emitting greenhouse gases, making a significant contribution to reducing global emissions. The heightened efficiency of TSCs enables them to produce more electricity during their lifespan, thereby amplifying their environmental advantages. Maka et al.[53] focused on identifying the contribution of solar energy, especially PV systems and concentrated solar power (CSP), to sustainable development goals (SDGs). Some of the findings included increased deployment of solar technologies because of their declining costs, ability to offer clean energy, lower emissions of greenhouse gases and potential as a source of employment. The study established that the level of installed solar PV capacity increased from 40 334 MW in 2010 to 709 674 MW in 2020 and that the solar energy sector now provides employment opportunities for over 3 million people. The approach adopted incorporates techno-economic and environmental perspectives examining at the role played by solar energy in the sustainability of energy security, climate change and economic development. The research used global treaties such as the Paris Climate Accord and applied the International Organization for Standardization (ISO) 14040-44 guidelines to LCAs. From the study, it is evident that as technology in the utilization of solar energy continues to be developed, solar energy will be central in meeting the energy needs of the world and supporting development.

Hallam et al.[54] investigated the requisite demand for polysilicon to facilitate extensive electrification through photovoltaics (PVs) by the year 2050. The analysis anticipates that the accomplishment of this objective will require an escalating demand for polysilicon ranging from 46 to 87 million tons, with annual requirements potentially peaking at 7 million tons by 2050. Furthermore, the research elucidates the considerable energy demands associated with silicon wafer fabrication,
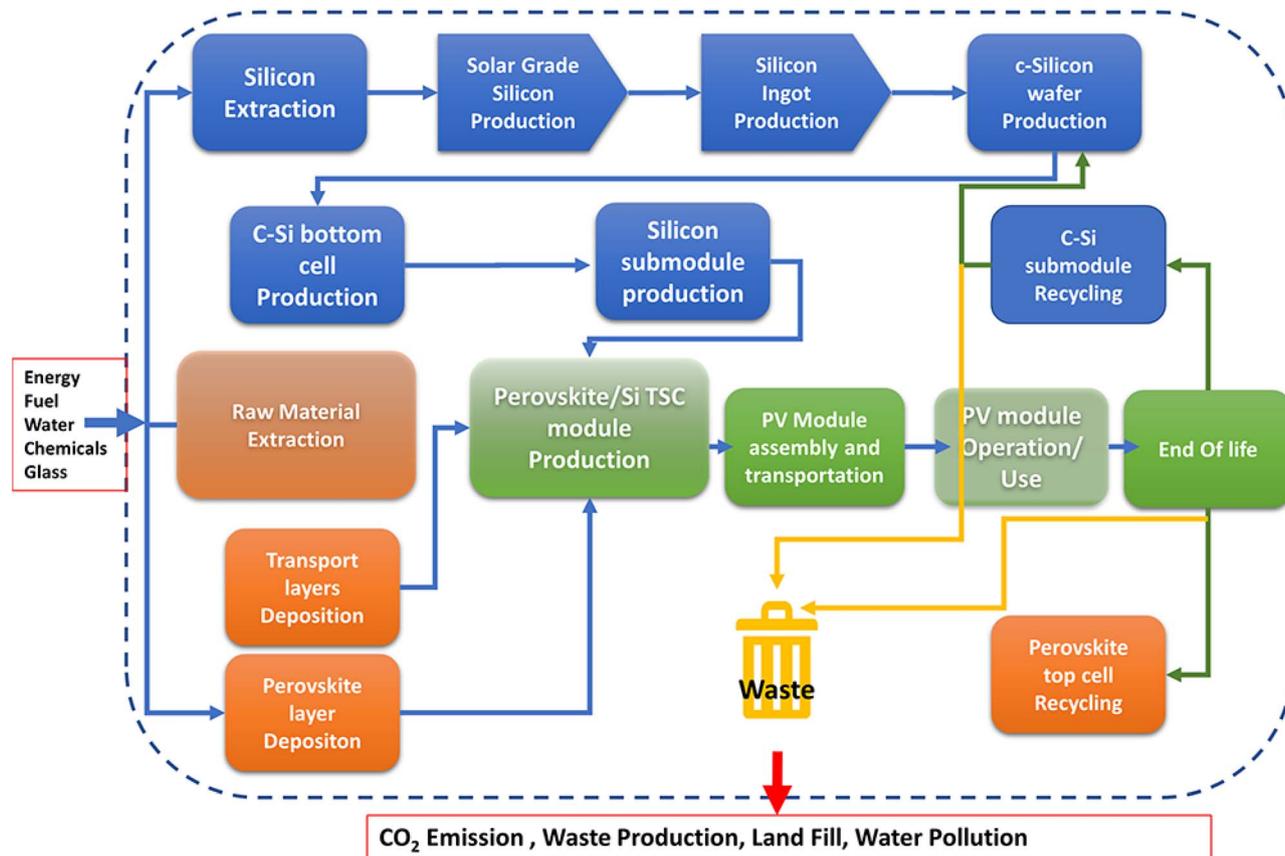
Fig. 2 Schematic illustration of a closes loop recycling process of perovskite/Si TSCs.

positing a cumulative carbon footprint of 16.4 to 58.8 gigatons of CO_2-equivalent emissions by 2050. In order to lessen these ecological impacts, the study underscores the necessity for enhanced efficiencies, the utilization of thinner wafers, diminished kerf loss, and the exploration of alternative purification techniques. The methodology employed incorporates an LCA framework to scrutinize the environmental ramifications linked with polysilicon production and its application within PV systems, conforming to ISO standards for thorough evaluation. Collectively, their study highlights the demanding requirement for innovations in polysilicon production methodologies and materials management strategies to foster sustainable advancement within the solar energy domain. Perovskite materials commonly include lead halides and organic halides, which also present environmental challenges such as lead mining-linked habitat destruction, soil and water pollution, and health hazards due to lead toxicity.[55–57] The apprehensions about lead in perovskite materials have prompted extensive research efforts, leading to the exploration of alternative lead-free perovskites that can achieve comparable efficiencies minus the associated health and environmental risks.[58–60]

The application of diverse chemicals in the manufacturing of perovskite top cells raises concerns about environmental and health risks if not managed adequately. The production of waste, including defective cells and off-spec materials, further compounds the ecological impact.[61,62] Exposure to chemicals

during the manufacturing process also poses health risks to workers, thereby requiring strict safety measures and the exploration of safer material alternatives. The supply of important raw materials such as lead, silicon, indium and silver poses a challenge to the sustainability of tandem solar cells. The high demand for these resources is likely to put pressure on existing supplies, highlighting the importance of sustainable sourcing and recycling practices. Moreover, the energy consumption during production, particularly if derived from non-renewable sources, exacerbates concerns regarding resource depletion.[63,64] Therefore, it is imperative to devise strategies that minimize material usage and optimize resource efficiency to ensure the enduring sustainability of PV technologies. Monteiro Lunardi et al.[65] conducted an evaluation of the ecological repercussions associated with perovskite/Si TSCs in comparison with conventional c-Si cells, using data from the literature. Their research complies with the guidelines of ISO 14040 and ISO 14044 concerning LCA, emphasizing environmental aspects like GWP, human toxicity, freshwater eutrophication, freshwater ecotoxicity, and the exhaustion of abiotic resources. The principal conclusions revealed that perovskite/silicon tandems exhibit reduced EPBT relative to c-Si solar cells, with perovskite/Si tandems utilizing aluminum contacts yielding the most favorable environmental results. The research highlights critical environmental hotspots, including the utilization of precious metals such as gold and the Spiro-OMeTAD

layer, advocating for the substitution of these materials to mitigate ecological consequences. Furthermore, enhancing the longevity and stability of the perovskite layer is imperative to diminishing overall environmental repercussions and bolstering the sustainability of these technological advancements. Martin Roffeis *et al.*[66] conducted a comprehensive LCA of industrially manufactured perovskite/Si TSC modules (Oxford PV) using the ReCiPe 2016 v1.1 method, following the internationally recognized LCA guidelines provided in ISO 14040 and ISO 14044 standards. Data of key environmental impacts such as global warming potential (GWP), terrestrial ecotoxicity potential (TEP), freshwater consumption (FWC), and fossil and metal depletion potential (FDP and MDP) have been estimated. The study identified a GWP of 434 kg CO_2 equivalent per module, with silicon wafer production being the primary contributor (Fig. 3(a)). Additionally, materials such as copper, aluminum, and float glass were found to significantly influence the metal depletion potential (MDP) and terrestrial ecotoxicity potential (TETP). Despite the higher environmental impacts per piece compared to conventional c-Si solar modules, perovskite/silicon TSC modules demonstrated superior efficiency, resulting in lower environmental impacts per kW h produced

(Fig. 3(b)). These findings underscore the necessity of optimizing production processes and material usage to enhance the sustainability of perovskite/silicon TSC modules.

The operational stage is marked by minimal emissions, positioning PV technology as a fundamental component of sustainable energy frameworks. Nevertheless, the enduring stability and performance of perovskite materials are ongoing subjects of research, with continuous endeavors to enhance their resilience in real-world scenarios. Ensuring the sustained high efficiency and stability of these solar cells across numerous years of operation is vital for optimizing their environmental merits and is advantageous for the distribution of solar cell materials and products.[68,69] A sensitivity analysis of two distinct categories of perovskite tandem solar cells (perovskite/Si and perovskite/perovskite) based on the Monte Carlo simulation method revealed that lifetime was the dominant factor influencing the GHG emission factor for both tandem modules (Fig. 3(c)). The perovskite–silicon tandem cannot achieve the GHG emission factor as low as the benchmark by merely enhancing the PCE but it could do so by prolonging the 15 year lifetime to 28 to 29 years while retaining the current PCE, which is hard to realize in the short run.[51]
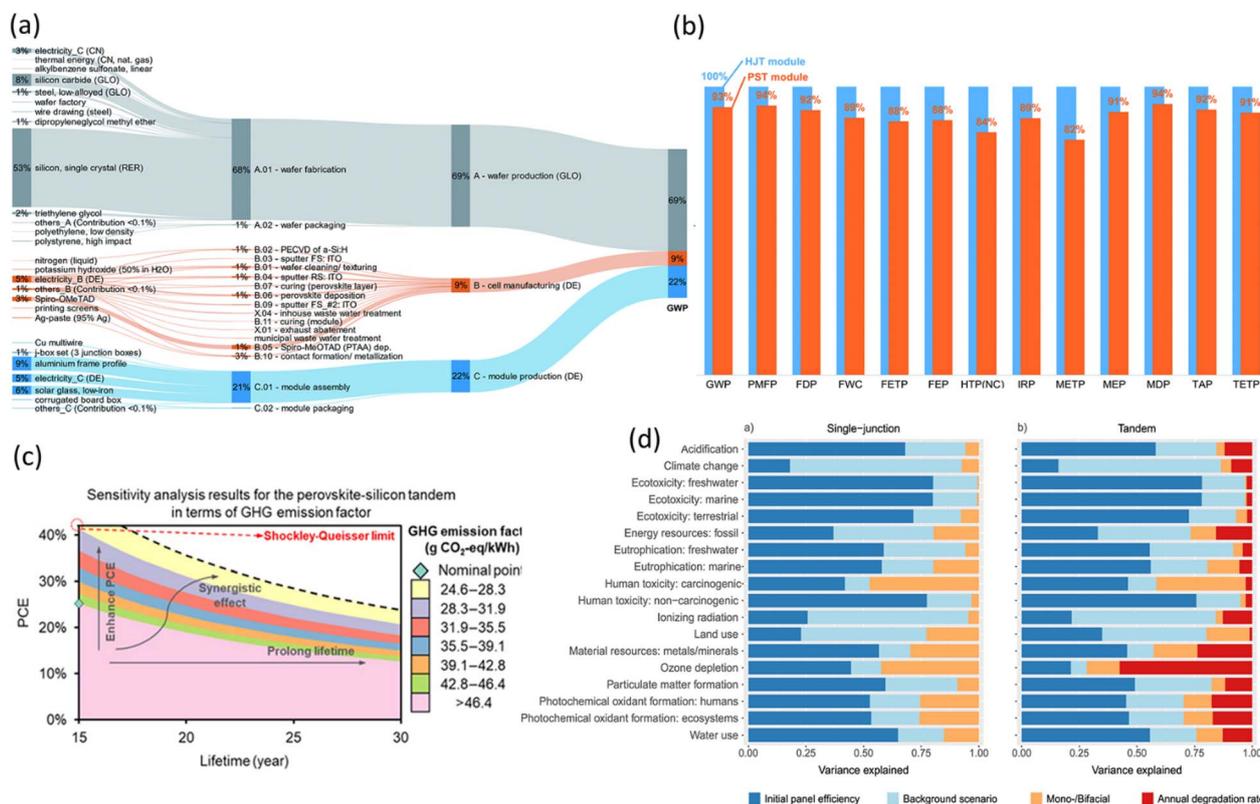
Fig. 3 (a) Analysis of the contributions to the global warming potential (GWP) linked with the supply of a PST module to a hypothetical market in Germany. Identification of inventory components with contributions exceeding 0.1% to the overall outcome. (b) Estimated environmental impacts per kW h for a SHJ and perovskite/Si TSC module operating over 25 years in Germany, with perovskite/Si TSC impacts shown as a percentage relative to those of the SHJ module across each impact category. Reprinted from Copyright@RSC Sustainable Energy Fuels, 2022.[66] (c) Effects of lifetime and PCE on the GHG emission factor of a perovskite−silicon tandem solar cell. Reprinted with permission from Copyright © 2020 Science Advances.[51] (d) Variance decomposition analysis was conducted for midpoint impact categories of (a) the single-junction panel and (b) the tandem panel utilizing the hierarchist (H) perspective of the ReCiPe 2016 LCIA method. Adopted from Copyright © 2024 ACS Sustain. Chem. Eng., 2024.[67]

The solar panel design and initial panel efficiency have an impact on the environment. Mitchell K. van der Hulst et al.[67] compared the full life cycle of monofacial and bifacial perovskite/Si TSCs with single-junction c-Si solar cells produced up to 2050 with a comprehensive prospective LCA. They analyzed the variance decomposition of footprints calculated with the ReCiPe 2016 (H) method with the impact categories recommended by the IEA PVPS. As shown in Fig. 3(d), the initial panel efficiency explains more than half of the variance in 12 of the 18 midpoint impact categories for the solar panel. This is to be expected since the efficiency has a strong influence on the lifetime electric output, with higher outputs resulting in lower impacts per functional unit (i.e., per kW h provided to the ENTSO-E grid). Balance-of-system (BOS) and recycling contributed substantially to impact categories. It is also suggested that annual degradation rates should not exceed 1% for monofacial or 3% for bifacial tandems to provide environmental benefits for tandem panels, and refurbishment of panels with advanced degradation is crucial.

Effective management of end-of-life (EOL) solar cells is important for reducing their environmental impact. Recovering important components and reducing waste is possible through the efficient recycling of silicon and perovskite materials. Progress in chemical recycling techniques and the establishment of closed-loop systems can improve the sustainability of perovskite/Si TSC technology. Mercy Jelagat Kipyator et al.[70] conducted an evaluation of diverse recycling methodologies for perovskite/Si TSCs predicated on their ecological ramifications. The investigation employed the ISO 14040 and ISO 14044 benchmarks, concentrating on four recycling strategies across monolithic and mechanically stacked perovskite/Si TSCs, thereby contrasting their environmental efficacy through the GWP metric. Principal findings indicate that the recycling strategy, where both perovskite and c-Si solar cells are recycled and replaced, exerts the greatest environmental impact, characterized by the periodic replacement of both perovskite and silicon cells, whereas the recycling strategy demonstrates the minimal impact, wherein solely the perovskite top cell is extracted while the c-Si bottom cell remains functional. Their results highlighted the environmental impacts of the recycling strategies, with the most sustainable waste management option identified as the one with the least environmental impact. Lu Wang and colleagues[71] also studied the environmental impacts tied to recycling practices for perovskite/Si TSCs. The investigation employs six end-of-life (EoL) modeling frameworks that are widely recognized in LCA, adhering to the stipulations outlined in ISO 14040 and ISO 14044 standards. Their study revealed that the selection of EoL modeling substantially influences the resultant environmental impact assessments. They used LCA results for a 1 m^2 perovskite/Si TSC using six different EoL modeling approaches. The research further emphasizes that the cumulative energy demand (CED) along with various environmental metrics, including abiotic depletion potential and ecotoxicity, exhibit comparable sensitivity to the recycling framework implemented. The study concludes that the circular footprint formula (CFF) represents the most holistic and representative methodology owing to its capacity to incorporate market realities and material degradation inherent in recycling processes.

The integration of strategies to promote a circular economy in the solar industry can further enhance sustainability. Enhancing recycling methods for both perovskite and silicon components is vital for waste reduction and the recovery of valuable materials. Yang et al.[72] conducted a comprehensive investigation into the feasibility of reusing c-Si bottom cells extracted from perovskite/Si TSCs. As shown in Fig. 4, it demonstrates that by employing techniques like thermal delamination and chemical cleaning, it is viable to recycle c-Si bottom cells. The optoelectronic properties of the regenerated c-Si bottom cells do not exhibit notable deterioration, thereby enabling the achievement of the same efficiency in tandem cells as if they were constructed using new perovskite top cells. Furthermore, this strategy has the added benefit of reducing the cost of solar energy production and enhancing the sustainability of TSCs by prolonging the lifespan of the silicon components. On the other hand, compared to silicon recycling technology, perovskite recycling technologies are currently less advanced. Lead in perovskite cells creates significant difficulties for recycling and safe disposal.[73,74] The existence of lead in perovskite materials raises significant concerns due to its harmful nature. Therefore, it is imperative to ensure the safe management, utilization, and disposal of materials containing lead to address and reduce these risks.[75] Tight EOL standards and improvements in recycling techniques are necessary since improper disposal of lead-containing objects can contaminate soil and water. Strong recycling regulations, reusing and infrastructure can greatly reduce the environmental concerns associated with solar cell disposal.[76–78] To tackle the environmental issues linked to perovskite–silicon tandem solar cells, various mitigation approaches can be put into place. The development of lead-free perovskite materials shows promise in reducing risks associated with toxicity.[79–81]

LCA of perovskite/Si TSCs is also influenced by transportation activities, which contribute to the carbon footprint by moving raw materials, components, and final products. The distances covered and the transportation modes utilized, such as shipping and trucking, are pivotal in determining the overall emissions associated with transportation. Effective management of the supply chain can help alleviate these impacts, with the utilization of low-emission transport options playing a crucial role as well, highlighting the significance of enhancing logistics to decrease carbon emissions by minimizing travel distances. The phase of installation encompasses the placement of solar panels in various environments, such as rooftops or solar fields. The consideration of land usage is crucial, especially for extensive installations that may necessitate land clearance. Conversely, rooftop installations utilize pre-existing structures, resulting in a reduced additional land usage impact. R. McKenna et al.[82] conducted a comprehensive investigation into the environmental and technical trade-offs associated with the implementation of onshore wind and PV technologies throughout Great Britain. This research adheres to ISO standards related to LCA (ISO 14040-44) and supports for enhanced coherence between energy and planning policies to facilitate the deployment of variable renewable energy technologies in scenic and
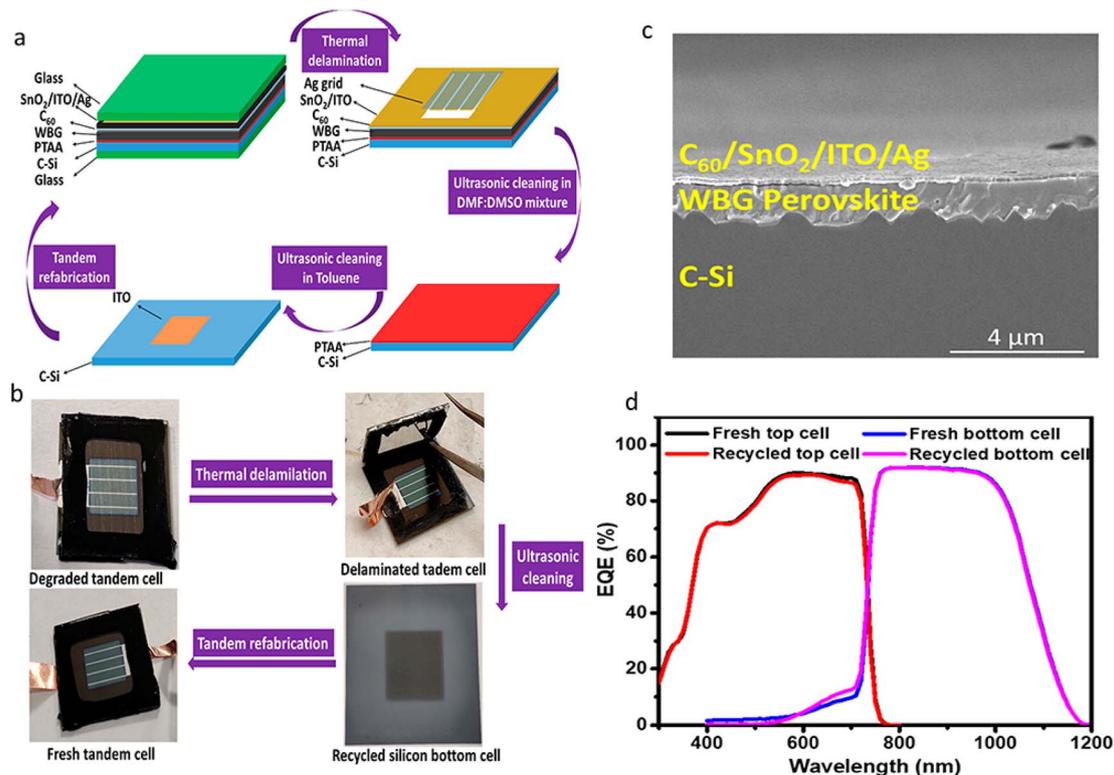
Fig. 4 (a) Schematic recycling process of silicon bottom cells from degraded, encapsulated perovskite silicon tandems. Step 1: thermal delamination: removal of the encapsulant and cover glass from encapsulated tandems by a thermal process. Step 2: ultrasonic cleaning: the PTAA/WBG/C_{60} films were removed sequentially by ultrasonication in DMF, DMSO, and toluene. Step 3: tandem remanufacturing from recycled silicon bottom cells. (b) Schematic illustration of tandem solar cell recycling and remanufacturing processes. (c) Cross-sectional SEM image of the refabricated tandem device. (d) J−V curves and EQE spectra of fresh and refabricated tandem devices. @Copyright © 2023, American Chemical Society.[72]

high-potential regions.[83] Their research employed a synergistic approach, utilizing both techno-economic and geospatial analyses, while also incorporating crowd-sourced information regarding scenic landscapes to evaluate the visual ramifications of renewable energy installations. Principal findings indicated that, although there existed substantial potential for ground-mounted PV (up to 7093 TW h), these opportunities were significantly impeded by competing land use and the intrinsic value of scenic environments. Rooftop PV presented a promising potential, with an estimated energy yield of 153 TW h. The study introduced an innovative top-down/bottom-up methodology for the assessment of rooftop PV, yielding spatially disaggregated outcomes. The choice of materials for electrical components and structural support, which may be made of plastic or metal, also affects how the installation phase affects the environment. During installation, using sustainable materials and designs can help to lessen the impact on the environment.

4. Sustainability analysis of perovskite/Si tandem solar cells

4.1 Cost analysis of perovskite/Si tandem solar cells

At present, it is difficult to accurately evaluate the actual manufacturing cost of the perovskite/Si TSC modules due to the multiple uncertain factors such as raw materials, fabrication processing, including technological variability, material costs, and the emerging stage of commercial production.[84–86] It is worth noting that the cost model relies on many assumptions and can display a variation of the levelized cost of electricity (LCOE). The LCOE is often used to evaluate technoeconomic competitiveness, which is affected by the efficiency of the device and its initial cost.[87] For example, it was found that commercial systems made from perovskite/Si TSCs would be competitive if their efficiency surpassed 26% and if the module fabrication costs stayed between US$90 and US$150 per m^2, assuming that the stability of perovskite/Si TSC matches that of c-Si cells.[88] C. Messmer et al.[89] compared the total module cost per watt peak (W_p) of traditional PERC single-junction c-Si cells with perovskite/Si TSCs (Fig. 5(a)). They found that when considering the impact of improving the efficiency of tandem solar cells, all tandem solar cells showed lower costs compared to single-junction c-Si cells. Li et al. conducted a detailed cost analysis of two types of perovskite-based tandem modules (perovskite/Si and perovskite/perovskite tandems) with standard c-Si solar cells and single-junction perovskite solar cells. They found that if the lifetime of the module is comparable to that of c-Si solar cells, tandem cells were competitive in the LCOE. Compared to the single-junction c-Si solar cell (5.50 US cents per kW h), the perovskite/Si tandem module shows a lower LCOE (5.22 US
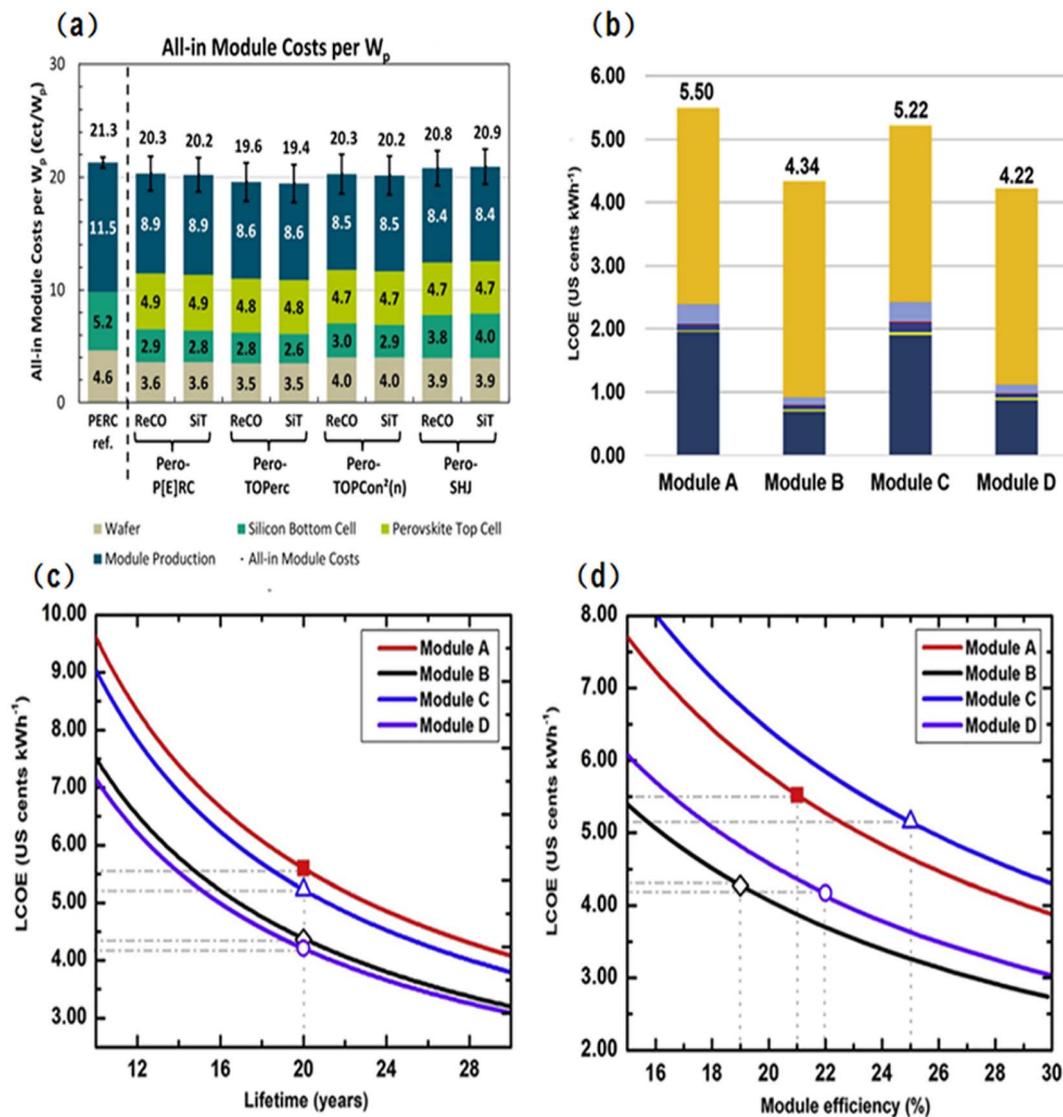
Fig. 5 (a) All-in module costs per W_p for all Pero−Si tandem concepts of Fig. 4 in comparison with a conventional PERC single junction reference (left side). Reprinted with permission. Copyright Wiley Library 2020.[89] (b) The comparison of LCOE. Module A is composed of traditional silicon cells, module B is composed of planar perovskite cells, module C is composed of silicon/perovskite tandem cells and module D is composed of perovskite/perovskite tandem cells. The sensitivity analysis regarding (c) module lifetime and (d) efficiency. (b−d) Reprinted with permission. Copyright 2018, Elsevier Inc.[90]

cents per kW h), indicating that the tandem configuration has potential for LCOE reduction in the future (Fig. 5(b)).[90] Moreover, lifetime and PCEs are two primary factors that significantly reduce the LCOE value (Fig. 5(c) and (d)). For example, in the case of the single-junction perovskite solar cell as an example, the LCOE decreases from 4.9 to 3.9 US cents per kW h if the module efficiency increases from 17% to 22%.

However, some technoeconomic studies show that excellent stability is more critical than efficiency increases or cost reduction to achieve competitive LCOE. The analysis of degradation rates showed that an increase in the rate of power loss from 1% to 3% per year would require at least a 10% absolute higher efficiency to obtain the same LCOE. If the efficiency cannot be increased, the module manufacturing costs would need to be reduced by more than 60%.[91] For a utility scale

system, if degradation rates increase by 1% per year, a technology needs to be either 3–5% more efficient or 9–39 US cents per W cheaper for equivalent economic performance. Perovskite/Si TSCs are expected to have higher costs than single-junction solar cells, and the degradation rate must be better than this value of 4%. When taking stability into consideration, the LCOE of perovskite/silicon TSCs also showed different changes.

In real applications, due to the cost of the top cell, perovskite/Si TSCs are about $3.7 per m^2 more expensive than standard silicon modules. Therefore, considering a lifespan of 25 years, an equivalent LCOE requires a series efficiency of 23%.[92] However, if the lifespan of TSCs is relatively short, at 20 years, the efficiency needs to be around 27% to be competitive. Due to the higher efficiency of perovskite/Si TSCs, they can

tolerate a lifespan reduction of 2–5 years compared to silicon modules. Duan *et al.* established a generalized model to assess the impact of the degradation rate, TSC efficiency, and TSC manufacturing cost and concluded that compared to single-junction c-Si solar cells, TSCs that degrade over 3% annually must be delivered at zero additional cost, while achieving over 30% efficiency with the same LCOE.[93]

Overall, although there are still some obstacles in terms of commercial feasibility, research on perovskite/Si TSCs has made astonishing progress, and after only 10 years of development, it has been proven that the efficiency of these batteries exceeds 34%. At present, the field is refocusing on solving stability issues, and some solutions have been demonstrated.[93] The key research work in the future will alleviate these stability issues and make perovskite/Si TSCs a major contributor to the ground photovoltaic market.

4.2 Toxicity analysis and management

The use of toxic materials, especially lead, should require compliance with existing regulatory frameworks. The European Union's Directive on the Restriction of Hazardous Substances (RoHS) and the Resource Conservation and Recycling Act (RCRA) set limits on electronic and electrical equipment, with a maximum concentration of lead in homogeneous materials of 0.1% per weight.[94] According to previous reports, perovskite solar cells (PSCs) with a typical perovskite layer thickness of 400 nm (*e.g.*, MAPbI$_3$ or FAPbI$_3$) would contain ≈0.4 g lead per square meter. When converted to a mass fraction, the lead concentration was estimated to be 344 ± 4 mg k^{-1} g for PSCs fabricated on rigid glass substrates, lower than 0.1% (Fig. 6(a)).[95] However, the RoHS directive does not apply to the United States. According to the regulations of the US Environmental Protection Agency (EPA), photovoltaic modules are classified as solid waste and require waste identification before disposal. The EPA has established a Toxicity Characteristic Leaching Procedure (TCLP) test (Method 1311) under RCRA for waste characterization. This procedure simulates leaching from landfills to determine whether harmful elements will leach from waste. There are currently eight TCLP restrictions for heavy metals, and the lead concentrations should be below 5 mg mL^{-1}.[95] Moody *et al.* performed the TCLP on two perovskite
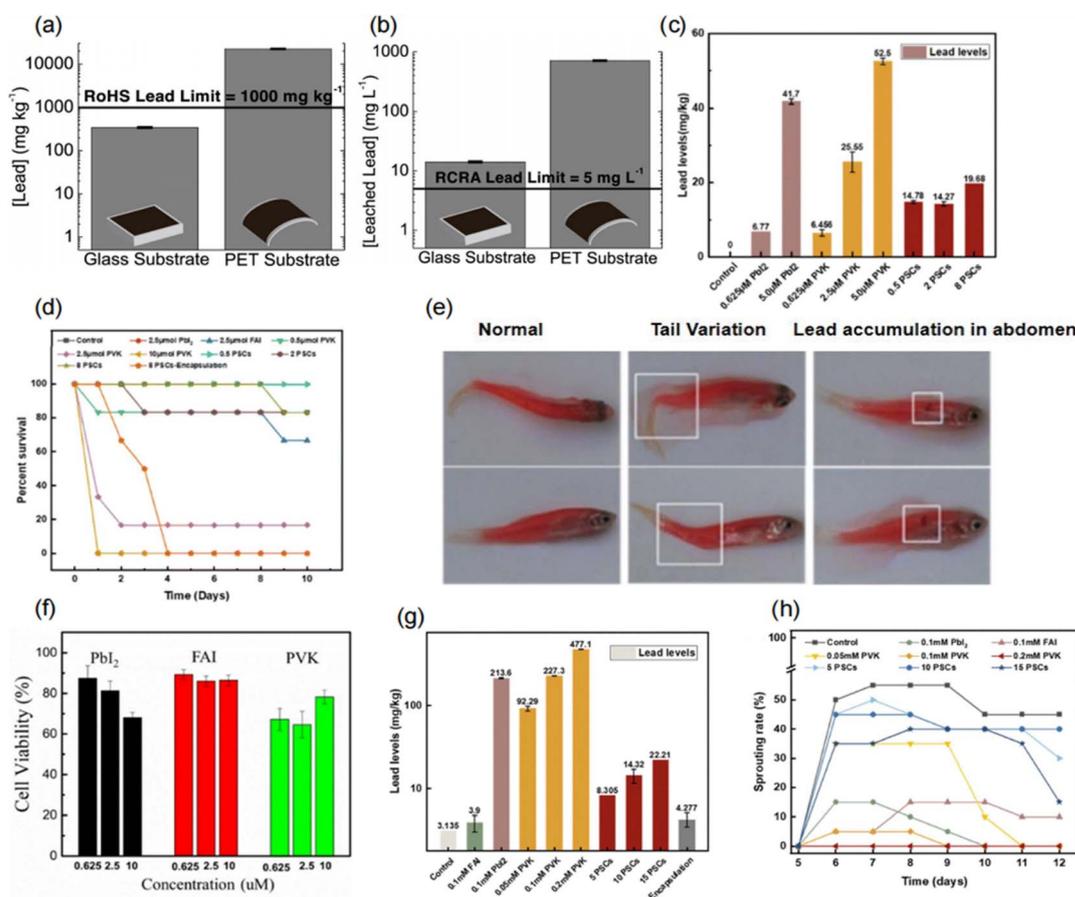

Fig. 6 (a) Total lead concentration of perovskite thin films on glass and PET substrates; (b) TCLP leached lead concentration of perovskite thin films on glass and PET substrates; reprinted with permission. Copyright 2020, Elsevier Inc.[95] (c) The Pb content in zebrafish after 10 days of breeding; (d) the survival of zebrafish under different growth conditions over a 10 day period; (e) the growth state of zebrafish with and without FAPbI$_3$; (f) the specific mortality data of chondrocytes under different perovskite material concentrations; (g) the Pb concentration of the radish sample under different conditions; (h) the sprouting rate of *Arabidopsis* with different times. Reprinted with permission. Copyright 2024@Royal Society of Chemistry.[96]

solar cells on glass and PET substrates and measured the leached lead concentrations to be 14.2 ± 0.2 and 713 ± 5 mg L^{-1}, respectively, both exceeding the limit of RCRA (Fig. 6(b)).[95] Thus, these materials require specific hazardous waste disposal rather than direct landfilling. Zhu *et al.* systematically investigated the toxicity potential of Pb-containing compounds that could be released accidentally from perovskite PV devices. They conducted ecological toxicity bioassays and cell toxicity assays, based on three different species and mouse chondrocytes, to study the harmful effects of PSCs on the environment and human health.[96] With the increase of environmental lead content, the lead content in zebrafish increases (Fig. 6(c)), leading to a gradual decrease in the survival rate (Fig. 6(d)). It is worth noting that zebrafish exhibited adverse reactions such as tail deformities and significant lead accumulation in the abdomen with the addition of 2.5 mmol FAPbI$_3$ solution (Fig. 6(e)). Although FAI is not toxic, the acidity of ammonium salts increases the acidity of the environment, which also leads to a gradual decrease in the population of zebrafish. The viability of mouse chondrocytes under different conditions is shown in Fig. 6(f), which is consistent with that of zebrafish. This indicates that lead content may lead to an increase in species mortality and growth inhibition, as well as a significant decrease in biological cell viability, highlighting the significant potential danger of PSCs and their degradation products that cannot be ignored. In addition, they also studied the ability of plants to absorb heavy metals from the ground in perovskite containing media, namely the bioavailability of lead. They found that radish and *Arabidopsis* absorbed much more lead from perovskite contaminated soil than from natural soil or soil contaminated with only PbI$_2$ (Fig. 6(g)), leading to varying degrees of *Arabidopsis* death (Fig. 6(h)). This situation occurred simultaneously in mouse chondrocytes and zebrafish experimental groups. This is due to the pH changes of organic cations in perovskite. Therefore, lead in halide perovskites is more dangerous than lead from other sources. Despite the lower content of lead-based perovskites, their harmful effects on animal and human health, as well as on plant growth and the entire ecosystem, cannot be ignored, which has become an important obstacle to commercialization.

Therefore, the ideal solution would be to replace lead while maintaining the considerable efficiency of perovskite/Si TSCs. Sn is the most favorable element among the candidates.[97,98] However, compared to lead based PSCs, Sn based PSCs exhibit lower PCE. Moreover, although tin is less toxic than lead, it is also a threat to the environment and human health. Short-term Sn ingestion can cause ataxia, muscle weakness, and irritation of the gastrointestinal mucosa, which may lead to vomiting and diarrhea. Chronic exposure to Sn can induce a mineral imbalance and dysfunction of the kidney and liver.[99] It is noteworthy that in the LCA category, Sn-based PSCs may be more hazardous because of their lower efficiency, suggesting that considerably more solar modules are required for the power supply, which can cause severe pollution.[100] Also, the reproductive toxicity, neurotoxicity, and carcinogenic potential of tin are still under debate. Thus, ion substitution of Pb is still an immature solution currently. Considering that lead is still critical in achieving

high efficiency and superior stability in PSCs, alternative solutions to reduce environmental pollution should reduce the exposed lead. There are mainly two strategies to solve the problem: (I) preventing lead leakage from the working modules and (II) recycling lead from the decommissioned modules. Regarding the reduction of lead leakage, Jiang *et al.*[101] demonstrated that using an epoxy resin for encapsulation can reduce the Pb leakage rate by a factor of 375 compared with using a glass cover with a UV-cured resin. Li *et al.*[102] developed a Scotch-tape-like design of an EVA film and a pre-laminated *P,P'*-di(2-ethylhexyl) methane diphosphonic acid layer that can capture over 99.9% of Pb leakage from damaged PSCs by applying it on both sides of the cells. Chen *et al.*[103] used a cation-exchange resin-based method that can prevent lead leakage from damaged perovskite solar modules. Meanwhile, the same group[104] integrated a mesoporous sulfonic acid-based lead-adsorbing resin into perovskites, demonstrating more effectiveness in preventing lead leakage than the configuration with the coating on the glass surface. On the other hand, Chen *et al.*[105] reported a lead management method for perovskite solar modules with a recycling efficiency of 99.2%, in which lead is first separated by weakly acidic cation exchange resin and is released as soluble Pb(NO$_3$)$_2$, followed by precipitation as PbI$_2$. Park *et al.*[106] reported a new adsorbent, iron-incorporated hydroxyapatite, for both separation and recovery of Pb from PSCs, which guarantees the recycling of 99.97% of Pb ions by forming lead iodide.

In addition to lead in perovskite/Si TSCs, the toxic solvents used in perovskite top cell manufacturing also raise community concerns. Generally, the perovskite precursor is dissolved in a polar non-protonic solvent and deposited to form polycrystalline films by thermal annealing.[107–109] However, most of the solvents used to prepare PSCs are poisonous to humans. Vidal *et al.* implemented a full LCA to analyze the impact during the whole procedure of perovskite solar cell manufacturing including industrial solvent production, use, removal, and end-of-life (EOL) treatment (Fig. 7(a)).[110] For EOL, four possible solutions are shown: (1) direct emission of solvent into the air; (2) incineration of condensed solvent with energy production; (3) recycling condensed solvents without further processing; (4) distillation recovers the solvent and incinerates the unrecovered portion. Mid-term processes such as solvent removal may require energy input, which could further raise concerns about global warming and fine particulate matter. Thus, Ding *et al.* conducted an analysis of ten commonly used solvents regarding their toxicity to the ecosystem, workers, and the general population. Based on device performance, the solvents were classified into four categories: most widely used, high industry potential, low industry potential, and high toxicity. Fig. 6(b) presents the results of this analysis. Notably, dimethyl sulfoxide (DMSO), ethanol, and isopropyl alcohol (IPA) are shown to have the least impact on the environment and human health while maintaining the desired device performance (Fig. 7(b)).[111] Though the application of green solvents may slightly restrict the PCE, efforts to reduce the toxicity of the preparation solvents are ongoing, and a compromise between efficiency and toxicity can be achieved through process advances in the foreseeable future.
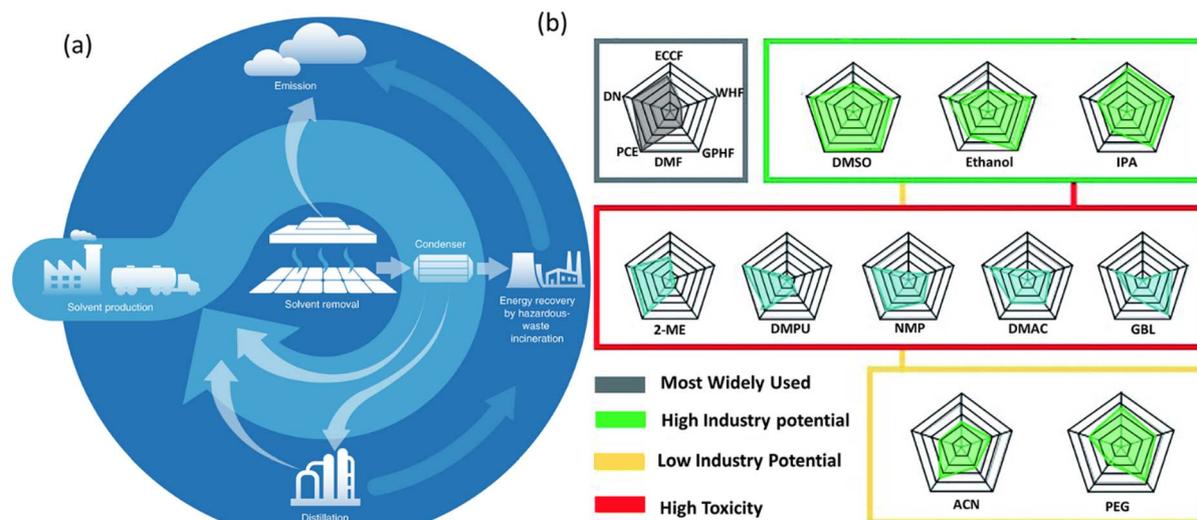
Fig. 7 (a) LCA system boundary schematic showing possible pathways for perovskite PV production. Reprinted with permission copyright@Nature Sustainability, 2021.[110] (b) Solvent five-dimensional property map, in which fuller means higher commercialization potential. The solvents are N,N-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), g-butyrolactone (GBL), N,N-dimethylacetamide (DMAC), tetrahydro-1,3-dimethyl-1H-pyrimidin-2-one (DMPU), poly(oxy-1,2-ethanediyl), a-hydro-u-hydroxy-ethane-1,2-diol, ethoxylated (PEG), acetonitrile (ACN), isopropyl alcohol (IPA), 2-methoxy ethanol (2-ME), ethanol, and 1-methyl-2-pyrrolidone (NMP). Adopted from Journal of Material Chemistry Copyright@Royal Society of Chemistry 2022.[111]

5. Challenges and opportunities

The shift to sustainable energy sources has made perovskite/Si TSCs new favorites because of their high PCEs. However, further and general incorporation and market penetration of this technology require a comprehensive LCA and sustainability assessment since these technologies are faced with several challenges and opportunities concerning their life cycle processes starting from development to use and disposal. It is crucial to define the research and development directions to control further development of perovskite/Si TSCs. Some of the major challenges comprise increasing perovskite stability and the adaptation of higher stability perovskite, enhancing the interface between perovskite and other layers, developing non-toxic and environmentally friendly perovskite materials, and working on recycling technologies.

The most pressing issue refers to the stability and durability. Perovskite materials easily degrade under the influence of moisture, oxygen, high temperatures, and UV radiation. The degradation herein presented has an impact on the service life and stability of perovskite/Si TSCs. To ensure commercial viability, standardized testing protocols that predict the long-term performance of perovskite-based PV must be established. Adhering to International Electrotechnical Commission (IEC) standards is a baseline requirement, but for true commercial viability, testing sequences with longer cycles and stricter criteria are necessary. Issues need to be addressed, and often, material engineering is the key to improving these facets and the lifetime of such devices, including the development of more stable perovskite compositions and better means of encapsulation. For instance, authors have found that the use of halide additives and a strong encapsulation of the perovskite

layers has been found to enhance the stability of perovskite layers.

There are interfaces between the perovskite layer, the charge transport layer, and the c-Si sub-cell, where losses and mobilities of charge carriers must be fine-tuned. Proper matching of these layers is required to account for problems like lattice mismatch, which leads to the formation of defects that are deleterious to the performance of devices. Thus, the engineering of the interface is remarkably important when it comes to designing perovskite/Si TSCs with high efficiency.[112] Methods such as passivated contacts and the creation of new CTLs that are compatible in terms of chemical structure with perovskite and silicon layers are critically important.[59,62,113,114] The design of perovskite/Si TSCs is important and should take environmental factors into account. The various features of these solar cells include the production process of the solar cells, transportation and use of the solar cells, and their final disposal, which affect the environment and must be managed to enhance the sustainability of the solar cells. The degrees of extraction and refining of the raw materials and the amounts of energy used in production as well as the possible release of dangerous chemicals at the time of disposal are other significant factors which ought to undergo scrutiny. Reducing hazardous solvent usage, environmentally friendly manufacturing processes and effective recycling solutions are the measures that could contribute to perovskite/Si TSCs' non-harmful effect on the environment.[14,66,115]

Another challenge that has been linked to the perovskite top cell is the one arising from the fact that lead is often used in efficient perovskite devices. Lead in any form if not well handled can be a health and environmental hazard. Lead has also been considered replaceable by non-more toxic metals such as tin, although the alternatives present lower efficiency currently.

Moreover, the encapsulation techniques which should avoid lead leaching and the enhancement of the recycling technology for the promotion of the recovery and the correct disposal of the lead are also important in handling this problem.[55,56,60,116]

The procurement of materials is also another important factor that should be taken into consideration in the LCA and sustainability assessment of perovskite/Si TSCs. It is crucial to make sure that the materials used in these solar cells are obtained from sustainable sources which have low impacts on the environment and without any unethical practices. This includes the responsible procurement of raw materials, recycling, and chain of supply management. Supplier relations that entail the use of sustainable strategies and accreditations can further improve the sustainability of perovskite/Si TSCs. For the present limitations of perovskite/Si TSCs to be addressed, further improvements in its materials and processes are necessary. These goals include the continued search for novel materials to be used in solar cells and improvement in their fabrication procedures to lower costs and energy intake as well as improve the general performance of these solar cells. Technological advancements currently include the formation of fresh hybrid perovskite materials, better deposition technologies, and productive technology in mass manufacturing. An important aspect of perovskite/silicon TSCs is the potential for closed-loop recycling, enabling the recovery and reuse of materials, thereby minimizing waste and reducing environmental impact. However, this recycling potential is not limited to perovskite/silicon TSCs and can also be applied to other solar technologies, highlighting the broader opportunities for sustainable end-of-life management across the PV industry.

Evaluating ways through which valuable materials can be recovered from EOL solar cells and then reused in creating new ones can help in cutting the input raw materials and waste levels. This includes coming up with new designs of solar cells with special consideration for the ability to recycle the used cells and integration of materials which can easily be recycled and publicizing known recycling programs within the solar industry. Thus, efforts can be made to achieve a circular economy in the photovoltaic industry. This review has demonstrated that technological innovations are critical in overcoming the difficulties and maximizing the abilities of perovskite/Si TSCs.

Some of the synergistic work between academia, industry, and government can be used to enhance the progress in these fields, as well as facilitate the scale-up of perovskite/Si TSCs. Governmental involvement in the determination of policies and/or regulations can be essential in the growth and implementation of perovskite/Si TSCs. International governments, agencies and regulatory authorities can facilitate progress through the formulation of policies, support of research and development and manufacturing policies, and setting policies and standards for the use of renewable energy technologies and safety and environmental issues. The following policies relate to the utilization of sustainable material, recycling and the support of financial incentives to promote the market sales of advanced solar technology perovskite/Si TSCs. The commercialization and the ability of perovskite/Si TSCs to penetrate the market largely rely on this triad of factors: cost, efficiency and sustainability. Thus, to make such types of cells common in the market, reducing manufacturing costs, increasing the efficiency and stability of production, and strictly following environmentally friendly benchmarks are critical. Consumers' knowledge and willingness to use will also be factors influencing market adoption which can be enhanced through the promotion and display of demonstration projects concerning the function of perovskite/Si TSCs.

6. Conclusions

In conclusion, perovskite/Si TSCs can be considered a huge step forward in the development of PV devices with increased efficiency and prospects for the creation of eco-friendly energy sources. Overall, from the LCA and sustainability analysis points of view, it is evident that although perovskite/Si TSCs have significant potential to realize a better environmental performance, problems persist. Challenges include the stability of perovskite layers, resolving the problem of interface engineering, and the toxicity of some precursors and effects on the environment that need also to be discussed to unleash the potential of perovskite. The quality of these cells also plays an important role; continuing R&D tasks help to improve their service life and their performance that would conform to commercial and legal requirements. When it comes to the present drawbacks, more projects in the field of materials research, advanced manufacturing processes, and life cycle analysis will be crucial. Finally, by fixing the key issues and exerting more efforts on enhancing more factors, perovskite/Si TSCs could be promising devices for serving sustainable and renewable energy systems.

Data availability

No primary research results, software or code have been included and no new data were generated or analyzed as part of this review.

Author contributions

Waseem Akram: conceptualization, writing – original draft and review & editing. Xikang Li: writing – original draft. Shakeel Ahmed: writing – review & editing. Zhengbiao Ouyang: supervision and project administration. Guijun Li: supervision, conceptualization, writing – review & editing, and funding acquisition.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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