Periodic Module Rejuvenation Provides Early Market Entry for Circular All-Perovskite Tandem Photovoltaic Technologies

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With explicit consideration of the short lifetime and high module degradation of perovskite tandem photovoltaics (PVs), we propose a replacement strategy for perovskite tandem PVs by implementing the developed recycling methods periodically to enable their long-term operation to match the multi-decade lifetime of incumbent PV systems. Here we show that market entry of emerging perovskite tandems and their contributions to decarbonization can be accelerated through periodic module replacement to counteract the module degradation and compete effectively with established PVs despite their relatively shorter lifetime in their initial development phases. This study shows that module replacement is a viable technological lever to boost market entry of the emerging all-perovskite tandems and provides insights into sustainable and circular PV technologies. The analyses unmask how periodic module replacement enables emerging PV technologies with short lifetimes to compete effectively with established PVs. The results inform an effective path to accelerate the widespread deployment of perovskite tandems and their contribution to the decarbonization of the energy sector, guiding the real-world implementation of periodic module replacement strategies and relevant regulatory policy-making. The results and implications from this study can be extrapolated to other emerging PV technologies by modifying module-specific dimensions.

Periodic Module Rejuvenation Provides Early Market Entry for Circular All-Perovskite Tandem Photovoltaic Technologies

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Summary

Emerging tandem photovoltaics (PVs) show cell power conversion efficiencies beyond incumbent PV systems but do not yet match their multi-decade lifetimes, impeding widespread deployment. Here we propose periodic module recycling as a strategy to resolve resource scarcity associated with halide perovskite tandem PV and relax initial stability requirements, thus accelerating their commercialization. By way of example, we fabricate high-performance all-perovskite tandems and experimentally show that indium tin oxide-coated substrates can be re-used multiple times without significant device performance loss. We show that for all-perovskite tandems with degradation rates as high as 10% per year, periodic module recycling gives reductions in greenhouse gas (GHG) emission factor and ameliorates energy return on investment (EROI). Our analytical findings provide quantitative and rigorous guidance on how to implement recycling in practice. Our results inform the optimal recycling frequency and the possibility of outcompeting the benchmark silicon PV in each phase of materials development over the commercialization trajectory of perovskite tandems. Being recycled every 10 years with 10% per year degradation results in GHG emission factor of 4.64 g CO_2-eq/MJ, lower than that of silicon PV (4.73 g CO_2-eq/MJ). As the all-perovskite tandem develops toward a lower degradation rate of 7% per year, periodic module recycling every 10 years results in EROI of 15.0, already outcompeting the silicon single-junction benchmark (14.8). Periodic module rejuvenation provides early market entry for all-perovskite tandems to outcompete silicon PVs on key sustainability metrics, while this same strategy is not viable for perovskite-silicon tandems for the current module efficiencies unless a degradation rate lower than 1.4% per year can be achieved from the outset. Our findings of module rejuvenation are economically viable as supported by levelized cost of electricity (LCOE) calculations with sensitivity analyses. This work unveils new strategies to bring online next-generation sustainable PV technologies earlier that can meaningfully contribute to decarbonization goals.

Keywords: periodic module rejuvenation, circular solar economy, perovskite tandem, module degradation, end-of-life recycling, commercialization, sustainability, life cycle assessment, levelized cost of electricity

Introduction

In 2021, solar photovoltaic (PV) technologies contributed to 3.7% of electricity generation globally, making PV the third-largest renewable electricity generation technology behind hydropower and onshore wind.[1] The United States (US) Department of Energy forecasts that solar PV technologies have the potential for driving deep decarbonization of the national grid by powering 40% of the grid by 2035,[2] with a 25% - 30% annual growth rate for new PV installations.[3] Widespread installation of PV, including the adoption of new thin-film technologies, could mitigate environmental issues associated with energy generation[4] but will increase the pressure on certain resource supplies.[5] For example, precious metals, such as indium, are scarce, and indium reserves will only last for two decades at current demand.[6] Another issue is the centralized production of silicon wafers,[7] which can lead to supply chain problems and shortages in countries that do not produce significant amounts themselves, like the US or Europe. On the other hand, the number of end-of-life (EOL) PV modules will increase substantially over the next decades as a result of projected 30-year lifetimes of silicon modules.[8, 9] The cumulative volume of decommissioned PV modules is estimated to reach 80 Mt worldwide by 2050.[4, 9] PV waste constitutes a severe environmental challenge but also offers an unprecedented value-creation opportunity. More extensive research is undertaken to explore a more sustainable EOL management for PVs,[10] typically including recycling,[11, 12] refurbishment, and re-use of retired or failed modules.[10] Different from merely downcycling the modules into land-filling materials of less value for silicon PVs or other lower-value applications, recycling enables the direct use of a fraction of the retired modules without depreciating the recovered components.

The options for EOL management of PVs are region- or country-specific and should be adapted appropriately before implementation. Since 2012, PV-specific waste regulations have come into place in the European Union (EU). EU regulations mandate 85% collection and 80% recycling of the materials used in PV panels through the Waste Electrical and Electronic Equipment (WEEE) Directive.[8, 13, 14] By contrast, no federal regulations exist in the US associated with PV recycling, and therefore PV waste is treated as per general waste regulations. There is currently a lack of recyclability of the mature PV technologies, such as silicon PVs, because the panels are not designed in a way to be deconstructed easily, making crushing of the panels, after removing the junction box, copper wire and aluminum frame,[15] the most viable waste management method. In addition, the recovery of components through crude processing, such as metals, is insufficient to ensure the economic viability of the whole recycling process without relevant incentives. Rich opportunities exist for emerging PV technologies to develop recyclable modules from the outset.

One of the most promising emerging PV technologies is metal halide perovskite solar cells (PSCs). Metal halide perovskites are ideal light-harvesting materials for single-junction and multi-junction solar cells due to their excellent optoelectronic properties, in addition to using earth-abundant materials,[16] low embodied energy,[17] and high-throughput manufacturing compatibility.[18] Single-junction PSCs have already achieved (laboratory-scale) power conversion efficiencies (PCEs) close to silicon.[19] In addition, the tunability of the bandgap through compositional engineering makes halide perovskites ideal candidates for tandem solar cells. In a tandem solar cell, two sub-cells are stacked on top of each other, with each sub-cell harvesting a different part of the solar spectrum. Tandem solar cells harvest a wider range of the solar spectrum more efficiently than single-junction analogs, allowing for higher PCEs. Perovskite-perovskite tandems recently achieved a record-certified PCE of 28%,[20] and devices using established silicon technology as the bottom cell has reached 32.5% PCE.[19] The hybrid perovskite-silicon tandem stack thus yields a higher overall PCE than record silicon cells alone (26.8%),[20] with few additional inputs in energy and materials. However, multi-decade lifetimes are yet to be proven for the emerging perovskite tandems, and it is likely such demonstrations will require multiple years of further testing and development.[21] To accelerate large-scale adoption of these technologies and their contributions to decarbonization, periodic module replacement strategies should be investigated to counteract the degradation of PV modules and enable emerging PV technologies with short lifetimes to compete effectively with established silicon and thin-film PVs. There are extensive research efforts focusing on the recycling of perovskite single junctions. Selective dissolution has been identified as a potential method for reusing glass substrate and electrodes.[22, 23] More encouragingly, a recently proposed mechanochemical approach is promising to recycle encapsulated devices with minimal performance loss.[24] However, there is still a lack of effective recycling approaches for perovskite tandems and multi-junctions.

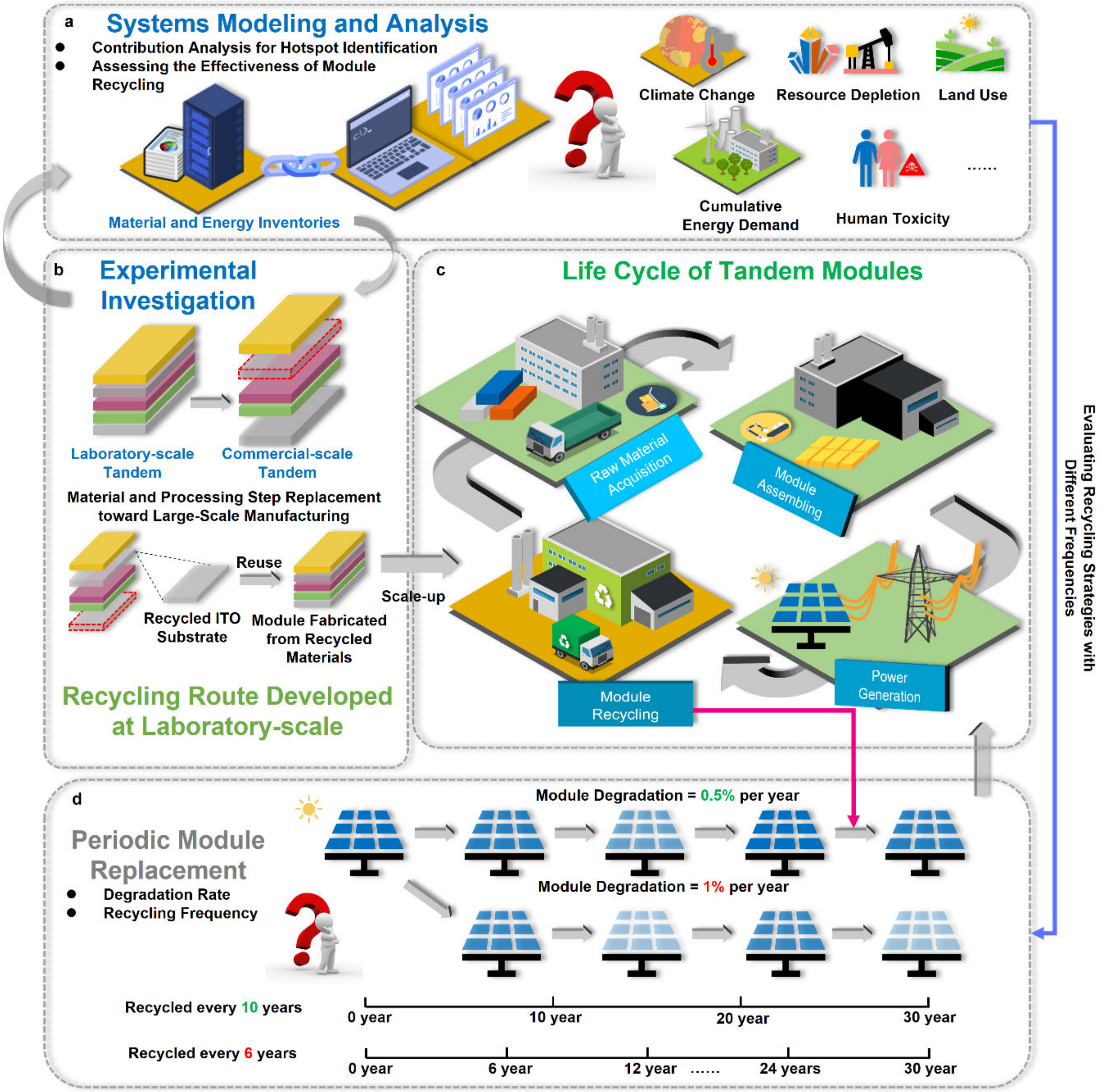


Figure 1. Overview of the integrated approach coordinating systems modeling and analysis with experimental investigation. Key components of experimental investigation and life cycle assessment (LCA) modeling as an integrated tool. The experimental investigation regarding perovskite tandem recycling at the laboratory scale helps in generating life cycle inventory (LCI) datasets and provides a technological lever for periodic module replacement through scale-up. The LCA results facilitate the identification of energy and environmental hotspots and evaluate the performance of module replacement at different frequencies. **a,** Systems modeling and analysis via LCA. **b,** Schematic of experimental investigation on recycling route. **c,** Life cycle of perovskite

tandem modules. **d,** Proposed periodic module replacement strategy.

Toward a quantitative understanding of periodic module rejuvenation's impact on the sustainability implications of circular all-perovskite and perovskite-silicon tandems, we first hypothesize that the periodic module replacement strategy can ameliorate the long-term efficiency of perovskite tandem PV systems in primary energy resource exploitation and conversion while mitigating environmental impacts regarding manufacturing, installation, and maintenance so that these emerging PV technologies can outcompete the market-leading silicon PV despite their relatively shorter lifetime. The proposed schematic of the integrated approach coordinating systems modeling and analysis with the experimental investigation is shown in **Figure 1**. To test the hypothesis, we first fabricate high-performance perovskite-perovskite tandems (>23%) and experimentally show that they can be recycled by mechanically removing the active layers and re-using the indium tin oxide (ITO)-coated substrate at least four times (**Figure 2a**). The recycled substrates do not show any signs of residual material after cleaning (**Figure S8**), or significant changes in resistivity or transmittance (**Figures S5** and **S7**). Perovskite deposited on top of the recycled substrates does not show any changes in optoelectronic properties when compared with perovskite deposited on fresh substrates (**Figure S9**). Perovskite-perovskite tandem devices made with recycled substrates do not suffer a loss in device PCE (**Figure 2c,d**) and long-term stability is unaffected (**Figure S10**). For perovskite-silicon tandems, we consider the possibility of removing the perovskite sub-cell, followed by the deposition of a fresh sub-cell on top of the silicon sub-cell, as shown in **Figure 2b**.

With explicit consideration of the short lifetime and high module degradation of perovskite tandem PVs, we propose a replacement strategy for perovskite tandem PVs by implementing the developed recycling methods periodically to enable their long-term operation to match the multi-decade lifetime of incumbent PV systems. We assume that manufacturing capacity will naturally grow over time globally, with approximately 1 TW installed annually to reach net zero targets.[25, 26] Even upon the market entry, it would probably not progress with the first panels being immediately recycled, which are more experimental. The implementation of recycling strategy is a relatively slow phase requiring the optimization of the process. However, in the steady state, the initial production would solely be new modules, until the first modules are returned for recycling. In the field, these recycled modules will immediately be replaced by new modules. The first set of recycled modules can then facilitate the recycling of further modules, creating a sustainable cycle of reuse and enhancing the material circularity. Here we show that market entry of emerging perovskite tandems and their contributions to decarbonization can be accelerated through periodic module replacement to counteract the module degradation and compete effectively with established PVs despite their relatively shorter lifetime in their initial development phases. We assume that the perovskite-perovskite tandem (including balance of system, BOS) reaches its EOL after 30 years.[27] We find that for perovskite-perovskite tandems with 10% degradation per year, recycling results in an improved EROI, while being recycled every 7.5 years results in the lowest GHG emission factor of 4.45 g CO_2-eq/MJ, lower than 4.73 g CO_2-eq/MJ for silicon PV. If a lower degradation rate of 7% per year can be attained, being recycled every 10 years corresponds to EROIs of 15.0, outcompeting 14.8 for silicon PV. The threshold degradation rates of perovskite-perovskite tandems, where intermediate recycling is no longer needed, are 3.0% and 2.0% per year in terms of EROI and GHG emission factor, respectively. However, recycling the EOL tandem module at the end of the 30-year lifetime is still needed to secure components, such as lead, and re-use valuable components. By contrast, a more stringent target of degradation rate lower than 3.0% per year from the outset should be attained for perovskite-silicon tandem with current PCEs to outperform silicon PVs on GHG emission factor, while a 1.4% per year degradation rate should be attained if EROI is considered. Our research is not focused on determining the incremental carbon budgets that perovskite PV might generate. Instead, we aim to quantify how perovskite tandems could contribute to further decarbonization of an already green technology.

This study shows that module replacement is a viable technological lever to boost market entry of the emerging all-perovskite tandems and provides insights into sustainable and circular PV technologies. The analyses unmask how periodic module replacement enables emerging PV technologies with short lifetimes to compete effectively with established PVs. The results inform an effective path to accelerate the widespread deployment of perovskite tandems and their contribution to the decarbonization of the energy sector, guiding the real-world implementation of periodic module replacement strategies and relevant regulatory policy-making. The results and implications from this study can be extrapolated to other emerging PV technologies by modifying module-specific dimensions.

Results

Investigated perovskite tandem architectures

We investigate two representative perovskite tandem architectures that have the potential to be implemented on a commercial scale. The tandem architectures are selected following three steps:

(1) Identify cutting-edge tandem architectures from recently published literature as the prototypes (laboratory-scale) with particular attention to their PCE and stability.

(2) Identify the energy and environmental hotspots from the prototypical tandem architectures through contribution analysis in terms of materials and processing steps.

(3) Consider replacement of laboratory-scale materials and processing steps that contribute substantial environmental impacts and energy use with industry-relevant counterparts.

In this study, two types of two-terminal (2T) perovskite tandem architectures are considered: a perovskite-perovskite tandem[28-30] and a perovskite-silicon tandem, with PCE of 23.8% and 29.3%, respectively. This is extrapolated from today's record laboratory-scale perovskite-perovskite and perovskite-silicon tandem solar cells, assuming 0.8% absolute loss per order of magnitude.[16, 20] The laboratory-scale cell typically has an area ranging from 0.049 to 1 cm^2 depending on the architecture,[29, 31] while the scaled-up module is assumed to have an area of 1 m^2. The tandem architectures and the corresponding recycling processes are shown in **Figure 2**. Detailed mass and energy balances for fabricating the two tandem solar cells are presented in **Tables S1-4**.

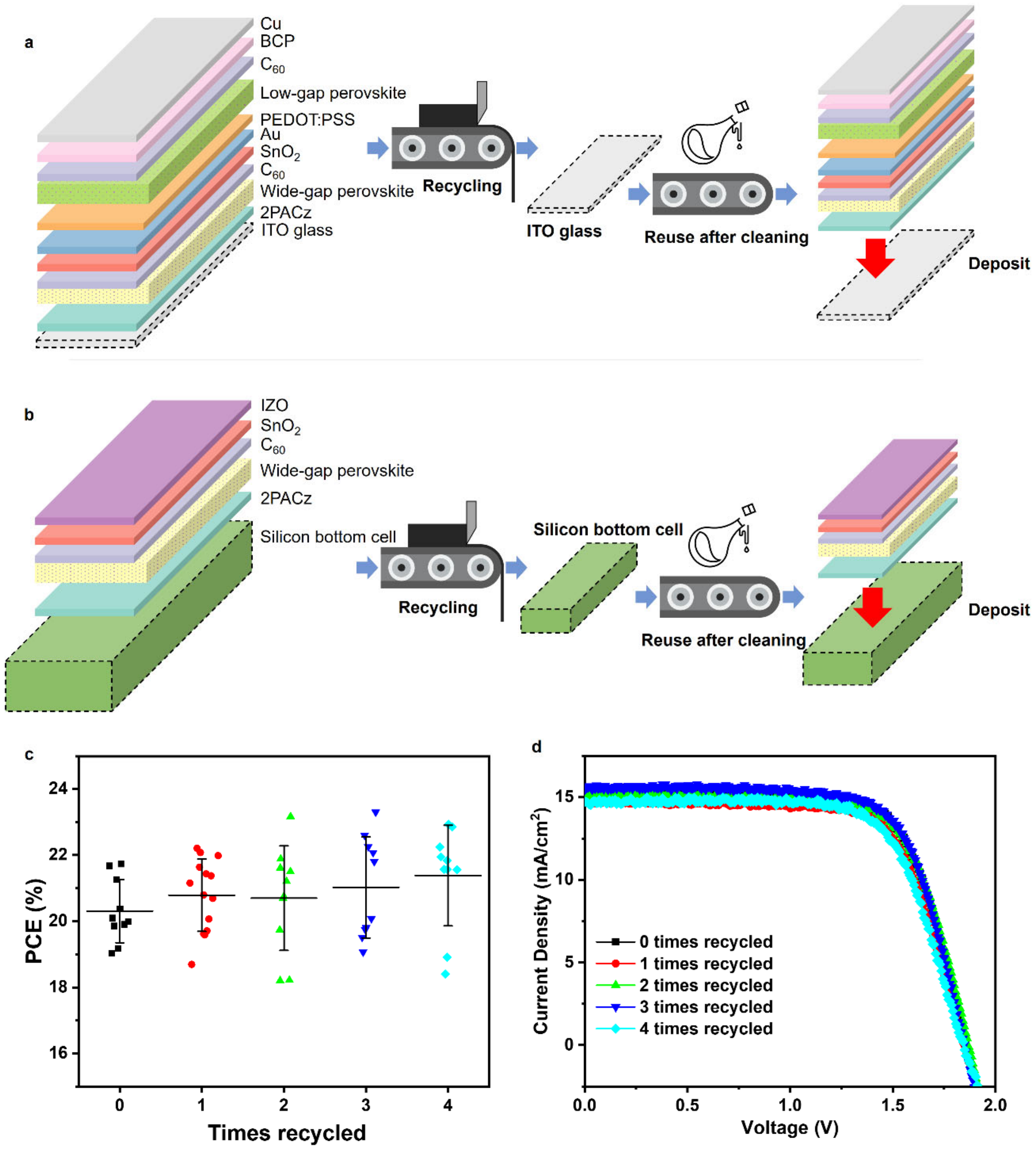


Figure 2. Schematics of perovskite tandem solar cells, deposition method from recycled materials, and key experimental results. a, Structure of the perovskite-perovskite tandem and recycling route by re-using ITO-coated substrate. **b,** Structure of the perovskite-silicon tandem and recycling route by re-using silicon bottom cell. **c,** PCE of perovskite-perovskite tandem fabricated from recycled materials. **d,** J-V curves of the perovskite-perovskite tandem solar cell recycled zero to four times.

Identification of recycled components in perovskite tandems

To explore the potential of widespread deployment of perovskite tandem solar cells, considering scalable fabrication in the context of the terawatt era, we assess the sustainability of such innovative PV technology.[27] To this end, silver is replaced with the more common metal copper.[32] Based on the results of the "cradle-to-grave" LCA, the atomic layer deposition (ALD) of the SnO_2 layer is identified as the most energy-intensive processing step. Although the industry-relevant spatial atmospheric ALD is two times faster than the laboratory-scale counterpart thus consuming less energy,[33] it is still energy-intensive in terms of absolute energy consumption. Therefore, we adopt sputtering for the deposition of the SnO_2 layer in the commercial-scale perovskite tandem fabrication. We note that the damage caused by sputtering could be minimized by reducing the sputtering power according to the method proposed by Härtel et al.[34] We do not consider any material and processing step replacement for the fabrication of silicon sub-cell since its fabrication method is relatively mature and well documented in the existing literature. Moreover, organic solvents would induce substantial environmental impacts if they are not recycled at the EOL or merely incinerated as hazardous waste.[35] To this end, for commercial-scale processes, organic solvents, such as acetone and isopropanol, are considered to be effectively recovered and re-used with an efficiency of approximately 90%, according to the existing literature.[36, 37] The environmental profiles for the prototypical laboratory-scale and adapted commercial-scale perovskite tandems, considering replacement of material and processing steps and solvent recovery based on the discussion above, are shown in **Figures S1-4**.

Effect of recycling frequencies on the global warming potential (GWP) and cumulative energy demand (CED)

To explore the potential of periodic module replacement in reducing the GWP and CED throughout the life cycle of perovskite tandems, we compare the emerging perovskite tandem PVs with the benchmark silicon PVs that typically demonstrate a much longer lifetime. Specifically, we assume the perovskite tandem module is recycled periodically over a total operation time of 30 years,[38] corresponding to the operating lifetime of silicon PV modules.[27] We explicitly consider re-using substrates (ITO-coated substrate and silicon bottom cell), organic solvent recovery,[36, 37] and lead recycling by precipitation as lead (II) iodide (PbI_2) for re-use.[39] **Figure 3a** and **b** compare the carbon footprint and CED for the two investigated perovskite tandems, considering that the EOL modules are recycled up to four times throughout the total system lifetime. The BOS (non-module components, such as inverter, cable, transformer) lifetime is considered to be the same as the system lifetime of 30 years.[27] As shown in the first column (left) of **Figure 3a** and **b**, the perovskite-perovskite and perovskite-silicon tandems correspond to 103.0 and 292.3 kg CO_2-eq without recycling at their EOL (landfill), compared to 318.9 kg CO_2-eq for the silicon PV. For the perovskite-perovskite tandem, we note that the BOS accounts for about 52.3% of the total carbon footprint and 44.3% of CED as the primary contributor, whereas the fraction of non-module components are below 20% for both perovskite-silicon tandem and benchmark silicon PV. This difference can be explained by the lower processing temperatures and embodied energy for the silicon-free tandem modules.

We fabricated perovskite-perovskite tandem solar cells with PCE of champion devices over 23% using an ITO / 2PACz / $Cs_{0.25}FA_{0.75}PbI_{2.1}Br_{0.9}$ / C_{60} / SnO_2 / Au / PEDOT:PSS / $Cs_{0.15}FA_{0.85}Pb_{0.5}Sn_{0.5}I_3$ / C_{60} / BCP / Cu architecture. The active layers were removed from the ITO substrates using a blade, and the substrates were re-used after cleaning (see Methods). We subsequently make further tandem batches on the recycled ITO substrates and show that there is no significant loss in device performance even after re-using the substrates four times (**Figure 2c-d),** indicating that the recycling process does not damage the ITO substrate and fully removes residue.

Periodic module rejuvenation could enable the near-term market entry of PSCs, especially in the tandem architecture, without the need for a multi-decade lifetime. A periodic module replacement strategy is proposed based on the recycling procedure depicted in the Methods section to counteract the decrease of PCE due to the degradation of active materials, the schematic of which is shown in **Figure 1d**. Here, we explore the impact of recycling frequency on the total carbon footprint and CED throughout the system lifetime and compare the results to the scenario without module replacement. Additional recycling process over the system lifetime would induce 32.7% and 9.6% more carbon footprint, and 43.3% and 12.6% more CED for perovskite-perovskite and perovskite-silicon tandems, respectively, relative to the scenario where module replacement is not considered. The all-perovskite tandem can be recycled up to three times before it exceeds the CED of silicon PV without recycling, while the GWP will not attain the level of silicon PV even after being recycled four times. The perovskite-silicon tandem has GWP and CED comparable to silicon PV without recycling, but recycling can still make sense if energy yield potential is taken into account.

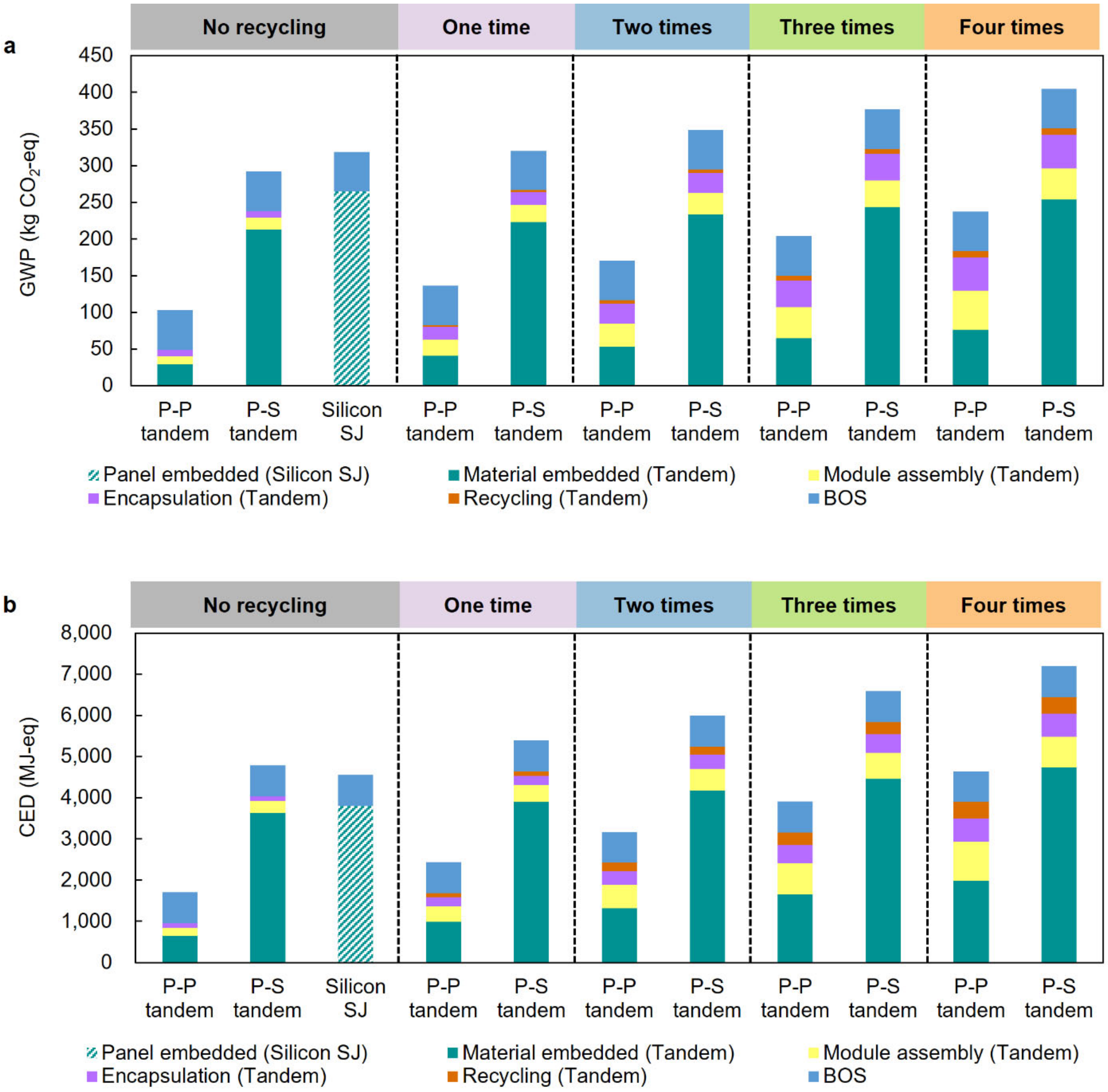


Figure 3. GWP and CED for perovskite-perovskite and perovskite-silicon tandems, compared to the benchmark silicon single junction (SJ). Comparison of the GWP and CED of perovskite-perovskite (P-P) and perovskite-silicon (P-S) tandems during their first life (module fabricated from pristine materials) and second life (module fabricated from recycled materials) and afterward as well as the total GWP and CED over the system lifetime considering different recycling frequencies. The sustainability metrics for perovskite tandems are assessed in terms of material embedded, module assembly, encapsulation, recycling, and BOS (including structure BOS for racking, and electrical BOS), while that of silicon PV is only assessed in terms of panel embedded impacts and BOS embedded impacts. Data for estimating the panel embedded GWP

and CED are retrieved from the literature,[40] shown as the shaded columns. **a,** GWP of perovskite-perovskite and perovskite-silicon tandems with different recycling frequencies, compared to the silicon PVs without recycling. **b,** CED of perovskite-perovskite and perovskite-silicon tandems with different recycling frequencies, compared to the silicon PVs without recycling.

Effect of degradation rates on the energy yield potential and levelized cost of electricity

One barrier to the widespread deployment of perovskite tandem PV technologies is the high degradation rate, varying widely with different active materials, tandem architectures, encapsulation approaches, and operating conditions. The short lifetime of the perovskite sub-cell bottlenecks the lifetime of the overall perovskite-silicon tandem, as well as the perovskite-perovskite tandem. For PSCs, a wide range of degradation rates have been reported, the lowest of which is around 2% (relative) per year.[41] It is noted that these values are measured based on laboratory-scale experiments with controlled conditions. Therefore, the degradation rates for emerging perovskite tandem PV technologies are subject to uncertainty. Historically, many PV technologies have achieved degradation rates of well below 2% per year and approaching the value of high-quality crystalline silicon PV.[42] Typically, monocrystalline silicon presents a degradation rate of 0.36% per year, polycrystalline silicon presents a degradation rate of 0.64% per year, amorphous silicon presents a degradation rate of 0.87% per year, cadmium telluride (CdTe) presents a degradation rate of 0.4% per year, copper indium gallium selenide (CIGS) presents a degradation rate of 0.96% per year.[42]

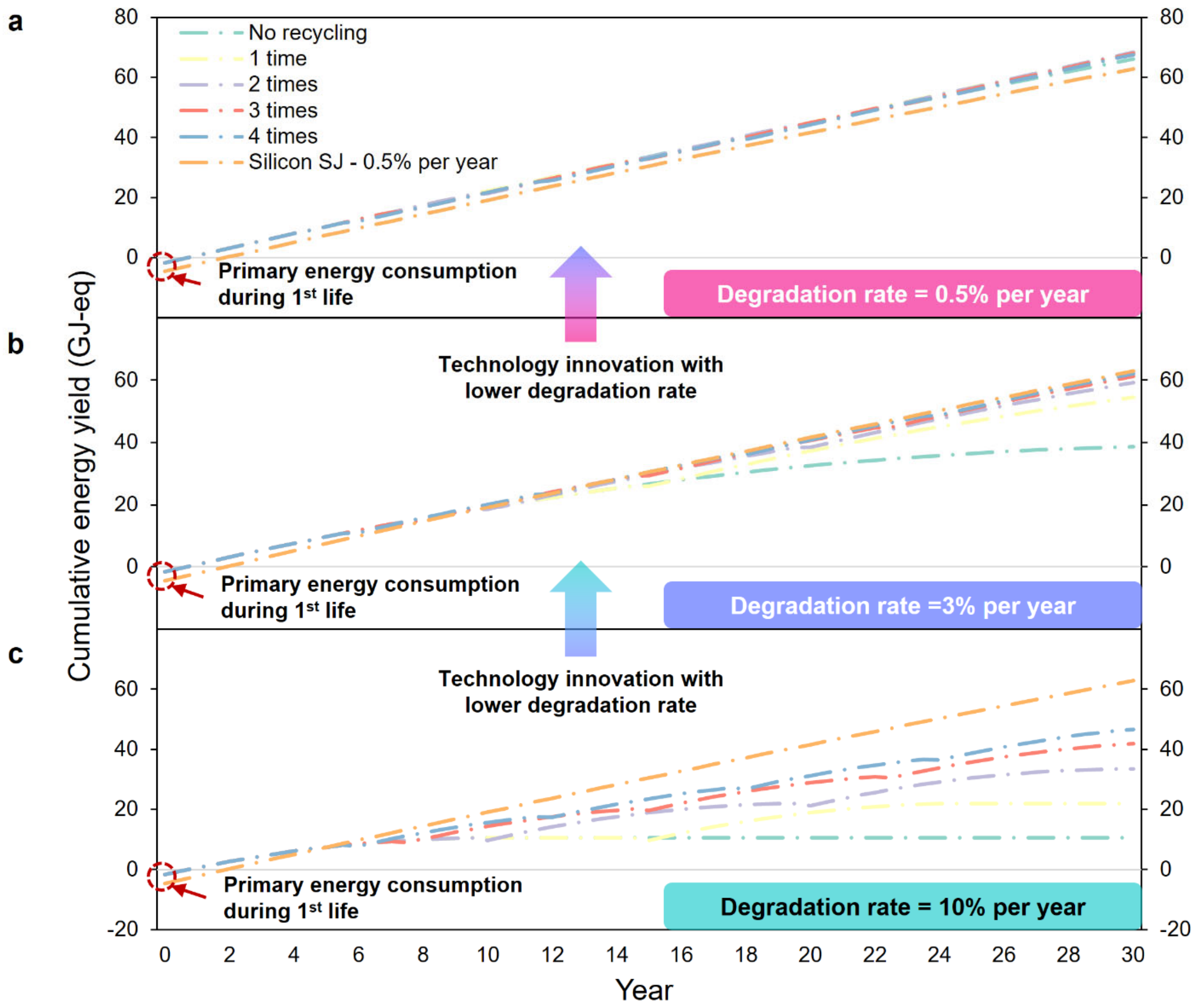


Figure 4. Cumulative energy yield for perovskite-perovskite tandems considering degradation rates of 0.5%, 3%, and 10% per year. a, Cumulative energy yield for perovskite-perovskite tandems with different recycling frequencies (from no recycling to four times over the system lifetime) under 0.5% per year degradation, and compared to the silicon PVs (0.5% per year degradation). **b**, Cumulative energy yield for perovskite-perovskite tandems with different recycling frequencies (from no recycling to four times over the system lifetime) under 3% per year degradation and compared to the silicon PVs (0.5% per year degradation). **c**, Cumulative energy yield for perovskite-perovskite tandems with different recycling frequencies (from no recycling to four times over the system lifetime) under 10% per year degradation, and compared to the silicon PVs (0.5% per year degradation). The cumulative energy yields initiate as negative values (year zero), which are equal to the initial primary energy consumption.

In this study, we assume a range of degradation rates, with an upper bound of 10% per year,[43] indicating the PCE drops to zero after 10 years, and the typical degradation rate of 0.5% per year for the crystalline silicon PVs serves as the lower bound. We consider a high degradation rate of 10% per year because 1) For perovskite solar cells, a wide range of degradation rates have been reported, the lowest of which is around 2% (relative) per year, 10% would be an extreme upper-bound that captures the degradation rate of perovskite-based solar cells that stand a chance to be widely deployed; 2) To capture what the threshold degradation rate would be to at least match silicon PV in terms of energy use and environmental impacts without recycling, it is worthwhile to explore quite a wide range of degradation rates to see where the critical thresholds are. To capture the general long-term degradation processes, we have chosen a linear decay rate, consistent with many perovskite tandems in literature that do not show a 'burn-in' effect.[28, 44, 45] We note that the overall PCE of the perovskite-silicon tandem is not the same as the starting PCE in each recycling cycle because of the irreversible performance loss of the silicon bottom cell (0.27% degradation per year).[46] As there is insufficient data on degradation rates of Si bottom cells in tandems, we assume degradation rates are identical to Si single junction cells. **Figure 4** shows the 30-year cumulative energy yield for the perovskite-perovskite tandem when the PV modules are recycled from zero to four times over the system lifetime, compared with a reference scenario where the silicon PVs degrade at a rate of 0.5% per year over a 30-year system lifetime for a fair comparison. With an initial module PCE of 23.8%, the 30-year cumulative energy yield reaches 67.8 GJ for the perovskite-perovskite tandem under the degradation rate of 0.5% per year if recycling is not implemented. The cumulative energy yield increases by 4.1%, 5.4%, 6.1%, and 6.5% when the PV modules are recycled one to four times, respectively. After being recycled four times, the perovskite tandem-based system does not present a substantial increase in energy yield potential. To this end, we stop recycling in the fourth cycle because the perovskite tandem-based system does not present a substantial increase in energy yield potential, showing perovskite tandems no longer outperform the silicon PVs in terms of EROI and GHG emission factors being recycled more than four times. It is also noted that achieving such a low degradation rate comparable to silicon PVs would be a long-term goal, which serves as the upper bound for estimating the energy yield potential of perovskite-perovskite tandem PVs. Higher degradation rates generally have lower energy yield potentials (**Figure 4b-c**). At 3% per year degradation, perovskite-perovskite tandem PV could generate up to 62.1 GJ over 30-year operations when recycled four times, while perovskite-silicon tandem has the potential to yield 72.2 GJ with the same recycling frequency (**Figure S12**), compared to 62.8 GJ for silicon PV (0.5% per year degradation, no recycling). If a 3% degradation can be achieved for perovskite-silicon tandem, recycling results in a cumulative energy yield higher than that of silicon PV.

Our results show that increased recycling frequency would be an effective technological lever to boost the energy yield potential of perovskite tandem PV technologies and counteract degradation. Increasing recycling frequency helps shrink the gap among different degradation scenarios. It indicates a less stringent requirement for the module lifetime, thus accelerating the market entry in terms of energy yield potential. The energy yield potential of a particular PV system is essential and worth investigating since it substantially affects economic performance. For a fair comparison of the feasibility and competitiveness of emerging PV technologies against their established counterparts, levelized cost of electricity (LCOE),[47, 48] the ratio of the total lifetime cost to the 30-year lifetime electricity production, is assessed for different module rejuvenation scenarios given varying module degradation rates and recycling frequencies. We assume a total of 36 modules with an individual area of 1.77 m^2 for residential systems and a constant area of 0.6 km^2 for utility-scale systems, following the assumption of existing studies.[49, 50] The total cost of the module, which includes the expenses related to utilities, materials, labor, depreciation, and maintenance, is estimated from the data, methodologies, and assumptions found within existing literature.[50-53] The techno-economic analysis for the recovery process is detailed in the Supplemental Information. The equipment cost, electricity usage, process throughput, and associated labor cost used for the techno-economic analysis are extracted from existing literature.[54, 55] The maintenance costs for the facilities are assumed to be 20% of the annual equipment depreciation.[53] Material costs are estimated based on recycling process of lead and transparent conductors from perovskite solar modules by Chen et al.[39] Module and BOS (including structure BOS for racking, and electrical BOS) costs are assumed to be 30% lower than in 2020.[50, 56] Module rejuvenation is assumed to incur labor costs for uninstallation and re-deployment that are twice as high as the initial installation expenses.

Different optimal recycling frequencies are identified corresponding to varying degradation rates. As shown in **Figure 5**, in cases where the perovskite tandems degrade at a higher rate of 10% annually, adopting a strategy of recycling three or four times throughout the system's 30-year lifetime results in the minimum LCOE for this degradation rate. However, as technological advancements lead to a reduction in the degradation rate of perovskite tandems to 4% annually, a less frequent recycling schedule, such as one or two times during the system's lifespan, becomes more desirable when considering LCOE. Further improvements in stability that reduce the degradation rate of the perovskite tandems to 0.5-1% per year render intermediate recycling unnecessary since higher LCOE is induced than without recycling. This is due to the fact that the supplementary electricity generated from module rejuvenation does not offer an effective return on the investment required for recycling, consequently leading to a higher LCOE. This critical threshold might be sensitive to the recycling cost, and we thus explore the impacts of varying input parameters (±20%) on LCOE, including the costs associated with energy consumption, labor, materials, and equipment during the module rejuvenation, represented by the error bars in **Figure 5**. In the short term, the proposed strategy of module rejuvenation displays potential for bringing online the emerging perovskite tandems earlier with lower LCOE, given their initial instability. We report LCOE of 14.1 (residential perovskite-perovskite tandem PV, 3% annual degradation) and 13.5 (residential perovskite-silicon tandem PV, 3% annual degradation) cents per kWh of electricity generated through one-time recycling, compared to the LCOE for current residential PV indicated at 11.7-28.2 per kWh.[57] However, in the long term, what truly drives the reduction of LCOE for perovskite tandem PV again comes back to improvements in module stability. Our results indicate that, with these advancements, the LCOE of utility-scale perovskite-perovskite tandem PV could decline to 3.6 cents per kWh of electricity generation in the long term where recycling during lifetime is no longer viable. More frequent module replacement would also introduce more intensive energy and material inputs, which should be balanced with the energy yield and is detailed in terms of EROI in the next section.

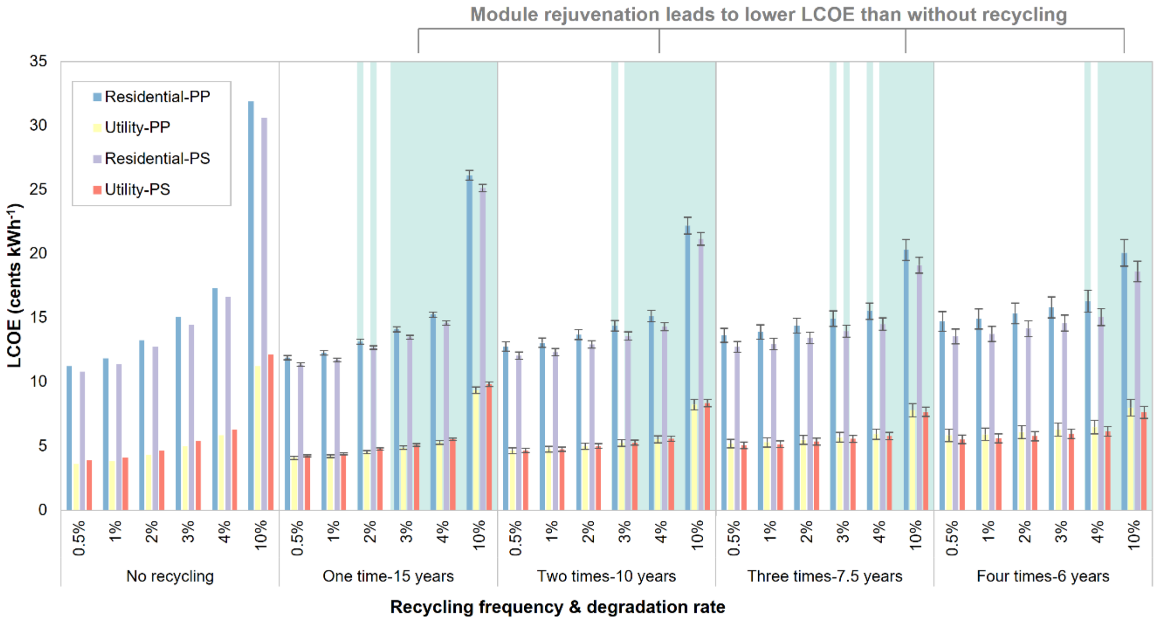


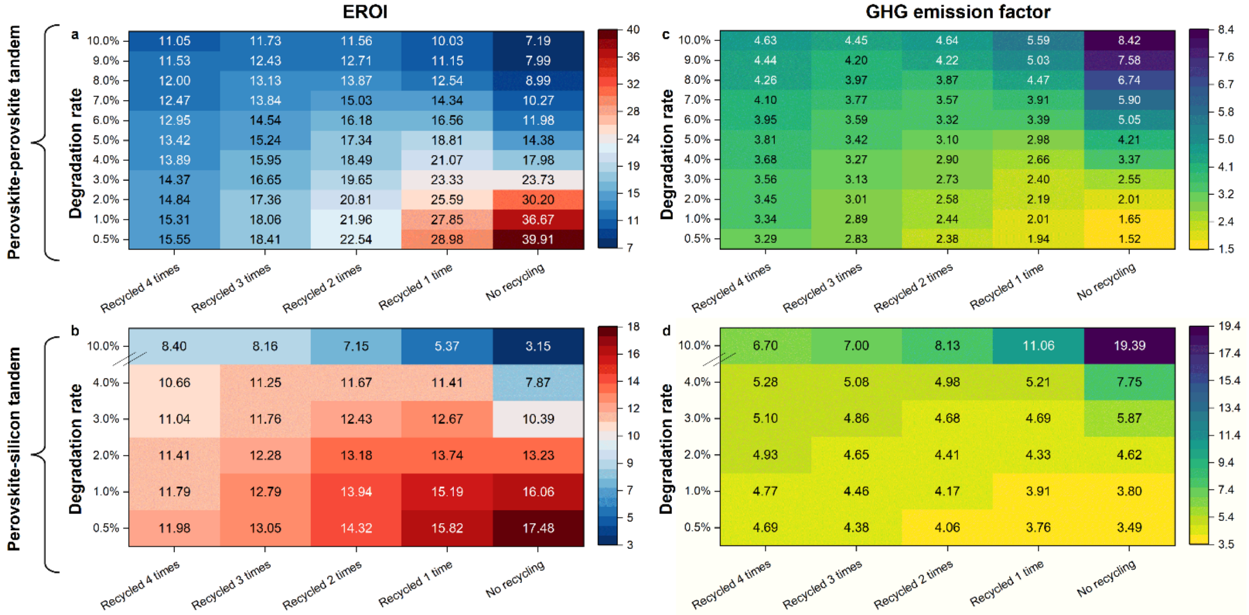
Figure 5. Levelized cost of electricity for residential and utility-scale perovskite-perovskite (PP) and perovskite-silicon (PS) tandem PV systems. This metric is assessed under varying module degradation rates from 10% to 0.5% and module rejuvenation frequencies, namely one to four times over the 30-year system lifetime, compared to the operations without recycling. The error bars reflect the impacts of varying input parameters (±20%), including the costs associated with energy consumption, labor, materials, and equipment during the module rejuvenation. The region in cyan-blue indicates where module rejuvenation leads to lower LCOE than with no recycling.

EROI and GHG emission factor

As a principal metric for evaluating the performance of energy generation systems, EROI is calculated as the ratio of electricity generation (in terms of primary energy) to the primary energy invested throughout the life cycle of PVs. Unlike the conventional definition of EROI, we tailor the analysis horizon from one life cycle of PVs to multiple ones by incorporating material circularity consideration and periodic module replacement strategy. The EROI is estimated to evaluate the net energy return over the system lifetime, which may contain multiple life cycles of the investigated tandem PVs and compared to the benchmark silicon PVs. Aligned with the previous discussion on the energy yield potential, we set the system lifetime as 30 years. Another widely investigated sustainability metric, the GHG emission factor, is estimated by dividing the life cycle GHG emissions by the total energy yield during the lifetime of the given PV system. The insolation of 1,700 kWh m^{-2} per year is assumed consistently throughout the calculation of the selected sustainability metric, including GHG emission factor, CED, EROI, and LCOE, following the assumption in a recent study for moderate insolation conditions.[40] The GHG emission factor and EROI corresponding to the reference scenario of silicon PV are estimated to be 4.73 g CO_2-eq/MJ and 14.8, respectively. **Figure 6** informs the optimal recycling frequency and possibility to outcompete the benchmark silicon PV for the perovskite tandems under different degradation scenarios. The highest EROIs for the perovskite-perovskite (39.9, no recycling) and perovskite-silicon tandems (17.5, no recycling) are observed under the lower bound degradation rate of 0.5% per year. For perovskite-perovskite tandems, even with 10% degradation per year, being recycled every 7.5 years results in a promising EROI (11.7), while being recycled every 7.5 years results in the lowest GHG emission factor (4.45 g CO_2-eq/MJ), lower than that of silicon PV. If 7% per year degradation can be attained, recycling twice over 30 years corresponds to EROI of 15.0, outcompeting silicon PV (14.8). To identify where the periodic module rejuvenation first becomes viable, i.e., to capture the threshold degradation rate, more scenarios are considered between 3.0% and 4.0%, with a 0.1% increment (**Figure S21**). The sensitivity analysis results show intermediate recycling is no longer needed as the perovskite-perovskite tandem develops toward lower degradation rate of 3.0%. However, recycling the EOL tandem module at the end of the 30-year is still needed to secure the toxic lead and re-use valuable components. The trends observed in the LCOE results varying with degradation rates consistently align with GHG emission factors and EROIs, despite slight variations in the optimal recycling frequencies under different degradation rates.

By contrast, for the perovskite-silicon tandem, a 1.4% degradation per year should be achieved before EROI becomes comparable to that of silicon PV, which is close to 1.5% per year degradation where intermediate recycling is no longer needed, indicating periodic module replacement is not viable for perovskite-silicon tandem on these metrics and module efficiency considered. The degradation rate of the tandem module is dominated by the least stable sub-cell, so in the case of the perovskite-silicon tandem, the degradation rate is dominated by the perovskite sub-cell. The reason it is not beneficial to recycle the perovskite-silicon tandem is that the impact of the tandem would be higher than for a silicon single junction, which would defeat using a tandem as a more efficient way of harvesting energy. Additionally, the perovskite-silicon tandems need much more energy and material inputs than the perovskite-perovskite tandem during its first-life deposition before modules are replaced, according to **Figure 3**. Drastic reduction in the CED and GWP during device fabrication is the fundamental factor and potential pathway to improve the sustainability of perovskite-silicon tandems. It is likely that the perovskite-perovskite tandem will degrade at a higher rate than perovskite-silicon tandem in the early stages. However, the EROIs of perovskite-perovskite tandem at 5-8% per year degradation are still higher than that of perovskite-silicon tandem at 4% per year degradation with the same recycling frequency. Therefore, the perovskite-perovskite tandem with periodic module replacement could be a promising option to outcompete silicon PV, due to its higher tolerance to the degradation rate than the perovskite-silicon tandem.

The EROI results would be further ameliorated if more materials involved in the perovskite tandems are recovered and re-used. With the same degradation rate, increasing the PCE or lowering the energy demand during the fabrication of perovskite tandems from the pristine materials and the recycling process after their first life would result in higher EROIs, enhancing the promise of perovskite tandems to outcompete the silicon PVs. Our estimates of energy inputs during the recycling process (based on a roll-to-roll blade coater with 10-second operations for active layer removal)[58] serve as upper bound for the real-world application as we are not recuperating much of the EOL tandem modules (merely re-using ITO-coated glass), and energy consumption is scaled based on the “best available” data obtained at laboratory scale. Reducing the energy inputs further during the recycling process of perovskite tandems may alter the trend of EROI varying with the recycling frequency and re-using more of the components, i.e., metals, enables further decrease in energy inputs and thus a higher EROI.



a EROI, Perovskite-perovskite tandem

Degradation rate	Recycled 4 times	Recycled 3 times	Recycled 2 times	Recycled 1 time	No recycling
10.0%	11.05	11.73	11.56	10.03	7.19
9.0%	11.53	12.43	12.71	11.15	7.99
8.0%	12.00	13.13	13.87	12.54	8.99
7.0%	12.47	13.84	15.03	14.34	10.27
6.0%	12.95	14.54	16.18	16.56	11.98
5.0%	13.42	15.24	17.34	18.81	14.38
4.0%	13.89	15.95	18.49	21.07	17.98
3.0%	14.37	16.65	19.65	23.33	23.73
2.0%	14.84	17.36	20.81	25.59	30.20
1.0%	15.31	18.06	21.96	27.85	36.67
0.5%	15.55	18.41	22.54	28.98	39.91

c GHG emission factor, Perovskite-perovskite tandem

Degradation rate	Recycled 4 times	Recycled 3 times	Recycled 2 times	Recycled 1 time	No recycling
10.0%	4.63	4.45	4.64	5.59	8.42
9.0%	4.44	4.20	4.22	5.03	7.58
8.0%	4.26	3.97	3.87	4.47	6.74
7.0%	4.10	3.77	3.57	3.91	5.90
6.0%	3.95	3.59	3.32	3.39	5.05
5.0%	3.81	3.42	3.10	2.98	4.21
4.0%	3.68	3.27	2.90	2.66	3.37
3.0%	3.56	3.13	2.73	2.40	2.55
2.0%	3.45	3.01	2.58	2.19	2.01
1.0%	3.34	2.89	2.44	2.01	1.65
0.5%	3.29	2.83	2.38	1.94	1.52

b EROI, Perovskite-silicon tandem

Degradation rate	Recycled 4 times	Recycled 3 times	Recycled 2 times	Recycled 1 time	No recycling
10.0%	8.40	8.16	7.15	5.37	3.15
4.0%	10.66	11.25	11.67	11.41	7.87
3.0%	11.04	11.76	12.43	12.67	10.39
2.0%	11.41	12.28	13.18	13.74	13.23
1.0%	11.79	12.79	13.94	15.19	16.06
0.5%	11.98	13.05	14.32	15.82	17.48

d GHG emission factor, Perovskite-silicon tandem

Degradation rate	Recycled 4 times	Recycled 3 times	Recycled 2 times	Recycled 1 time	No recycling
10.0%	6.70	7.00	8.13	11.06	19.39
4.0%	5.28	5.08	4.98	5.21	7.75
3.0%	5.10	4.86	4.68	4.69	5.87
2.0%	4.93	4.65	4.41	4.33	4.62
1.0%	4.77	4.46	4.17	3.91	3.80
0.5%	4.69	4.38	4.06	3.76	3.49

Figure 6. GHG emission factor (g CO_2-eq/MJ) and EROI for the perovskite-perovskite and perovskite-silicon tandems with periodic module replacement under different module degradation scenarios and recycling frequencies. The GHG emission factor and EROIs for the two investigated perovskite tandems are estimated under an insolation level of 1,700 kWh m^{-2} per year over a total system lifetime of 30 years. Five scenarios varying recycling frequencies are simulated, considering the perovskite tandem modules are recycled zero to four times over a total system lifetime of 30 years. **a**, Heatmap for EROI of the perovskite-perovskite tandems. **b**, Heatmap for EROI of the perovskite-silicon tandems. **c**, Heatmap for GHG emission factor of the perovskite-perovskite tandems. **d**, Heatmap for GHG emission factor of the perovskite-silicon tandems. The GHG emission factor and EROI of the silicon single junction at 0.5% per year degradation are estimated to be 4.73 g CO_2-eq/MJ and 14.8, respectively.

Typically, commercial PV modules are considered to be replaced when their power conversion efficiency drops to 80% of the initial value.[42] To account for this critical threshold, we examine its impacts on the two sustainability metrics under investigation. It is important to note that an 80% lower limit for the initial power conversion efficiency during the operation of perovskite tandem solar modules might be overly stringent, given their weaker stability upon early market entry. For instance, if an 80% threshold is adopted as a replacement indicator in the short term, tandem modules with a 10% annual degradation rate would require replacement every two years, which may not be the most cost-effective choice. To address this concern, we assume a starting threshold of 70% for this emerging technology, slightly below the standard 80%, and study the impacts of varying thresholds that eventually converge to the 80% level with continued research and development, as shown in **Figure S22** in the Supplemental Information.

Discussion

A rapidly increasing amount of EOL PV panels provide a tremendous opportunity to accelerate the circular solar economy due to the embedded values of waste glass, semiconductors, and metals, which account for most of the PV panels by weight. For instance, typical crystalline silicon PVs comprise about 76% glass, 8% aluminum, 5% silicon, 1% copper, and ~0.1% silver.[59] Moreover, McDonald and Pearce have reported a theoretical negative recycling cost of -$21.38/module for the CIGS PV modules, showing that it could be profitable to recycle EOL PV panels.[60] However, only around 10% of EOL PVs are currently recycled worldwide,[61] because disassembly of EOL modules is not straightforward, and recovery of components through non-specialized processing is insufficient to ensure the economic viability of the whole recycling process without political and techno-economic interventions, including stronger regulatory policies to limit the landfill of EOL PVs, higher landfill cost, and financial incentives for PV recycling. This situation would change if the EOL panels are effectively rejuvenated as we propose here so that perovskite tandems could become viable in both energy consumption and environmental sustainability terms beyond just reducing landfill. There is an opportunity for the emerging perovskite tandems to incorporate potential recycling considerations from the outset. Furthermore, it is likely that the PCEs of perovskite tandems continue to be significantly improved over the next decade. These advances can be used to upgrade the tandems during recycling, making the proposed periodic module replacement strategy even more attractive. A preliminary techno-economic analysis (**Figure S16**) along with sensitivity analysis (**Figure S17**) for the recycling process shows that the costs would be dominated by labor (31.6%) and material costs (55.8%), and the total recycling cost is $8.3/m^2 of the module, representing a small fraction given PV module costs across different technologies are expected to reach $50/m^2.[62, 63] Enhancements in stability, which lower the degradation rate of perovskite tandems to a range of 0.5-1% annually, eliminate the need for intermediate recycling, since such recycling tends to increase the LCOE, rather than reducing it as intended. In the long term where recycling during the system's lifetime is no longer viable, the utility-scale perovskite-perovskite tandem PV is estimated to reach 3.6

cents per kWh of electricity generated. The trends reflected in these LCOE results, which fluctuate with different degradation rates, are consistently in line with both GHG emission factors and EROIs. However, it is worth noting that optimal recycling frequencies exhibit minor variations for the investigated environmental and economic sustainability metrics when accounting for different degradation rates. Further in-depth techno-economic analyses will need to be performed to further examine the economic feasibility of the periodic module replacement strategy and factor in costs for transportation, dismounting and remounting the arrays as more commercial-scale data becomes available in future. Combining the techno-economic analysis results with the LCA results obtained in this work would enable a more judicious selection of viable periodic module replacement schemes, and refinement of the recycling processes.

As per EU regulations, 85% collection and 80% recycling of the materials used in PV panels is required under the WEEE Directive.[8, 13, 14] More components need to be collected and recycled for perovskite-perovskite tandems to ensure compliance with the regulations. So far, we have only considered the re-use of ITO-coated substrate, but other materials involved in the perovskite tandems could potentially be recovered and re-used as well. For instance, the metal electrode could be simply peeled off from the EOL PVs and re-used after cleaning; other materials could be separated by solubility in a particular solvent and re-used after purification. Experimental investigation into the effective methods to re-use the aforementioned components would further ameliorate the circularity of perovskite tandem PVs and further reduce the carbon footprint and CED of circular perovskite tandem PVs if the proposed recycling processes are properly incorporated into systems modeling.[39, 64, 65] The systems modeling and analysis should be closely woven with the relevant experimental investigation to explore potential recycling routes and promote the circularity of emerging PVs. The interactions between systems analyses and materials developments are needed at the earlier laboratory-scale development stage of tandem solar cells to identify hotspots for new materials or layers before they are developed further. LCA studies in the early stage could foster the development of laboratory-scale tandem solar cells in a more systematic way. Moreover, LCA modeling proposes a need for more industry-relevant data input, and it is impractical to acquire all the data needed for roll-to-roll manufacturing in most academic laboratories, which will need industry or national laboratories feeding these data in.

Conclusions

This study explored the potential of energy-saving and emission mitigation by recycling perovskite tandem PVs and reusing the critical constituents within. The technical viability of the recycling method was validated through experimental investigation. The knowledge was further advanced by exploring periodic module replacement as a viable pathway to bring online next-generation sustainable PV technologies earlier that can meaningfully contribute to decarbonization goals. In this work, an integrated approach coordinating LCA modeling and experimental device work was used to investigate the potential of implementing periodic module replacement for two types of perovskite tandem modules. Experimental inputs bridge the data gap in developing the LCI for the tandem recycling process and disclose the effectiveness of the proposed recycling method by characterizing the recycled materials. Energy and environmental hotspots were identified through a contribution analysis, and the results provided guidance to the development of the periodic module replacement strategy for perovskite tandems and evaluated its performance. Based on the proposed recycling strategy, increased recycling frequency helps shrink the gap among different degradation scenarios and indicates a less stringent requirement for the module lifetime, thus accelerating the market entry in terms of energy yield potential. The results show that for perovskite-perovskite tandems with 10% degradation per year, recycling results in an improved EROI (up to 11.7) compared to the no-recycling scenario, while being recycled every 7.5 years results in the lowest GHG emission factor (4.45 g CO_2-eq/MJ), lower than that of silicon PV (4.73 g CO_2-eq/MJ). If 7% per year degradation can be reached, recycling twice over 30 years corresponds to EROIs over 15, outcompeting silicon PV (14.8). Intermediate recycling is no longer needed beyond this as the perovskite-perovskite tandem develops toward even lower degradation rates, around 3%. By contrast, periodic module recycling cannot enable similar early market entry for perovskite-silicon tandems on these sustainability metrics for current considered module efficiencies unless a degradation rate lower than 1.4% per year from the outset is achieved. Deploying perovskite-perovskite tandems with periodic module replacement could be a promising route to bring perovskite tandems to market and outcompete silicon PV in the shorter term due to its high tolerance to the degradation rate. We also note that perovskite tandem technologies have the promise to attain higher module PCEs than the current values toward >30% as they mature in the longer term. Extremely high PCE makes perovskite-silicon tandem with periodic module recycling another potential pathway for large-scale adoption of perovskite tandem PV technologies. Future work should indicate the threshold PCE where periodic module recycling becomes viable with perovskite-silicon tandem.

In this work, we mainly focus on evaluating the sustainability and feasibility of early market entry through periodic module replacement for the selected tandem modules with fixed configuration based on the state-of-the-art device architecture. Further optimizing the power conversion efficiency and stability is beyond the scope of this work and is the subject of ongoing work in the field.[28, 45, 66, 67] In any case, the analysis could be extrapolated to other device architectures by substituting the material inputs and processing steps in both experimental investigation and LCA modeling.

Experimental Procedures

Resource availability

Data and code availability: All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplemental Information. Additional data is available from authors upon request.

Overview

This study combines LCA and experimental data as an integrated approach to study the implementation of periodic replacement strategy based on the proposed recycling process and explore the potential of energy-saving and pollution mitigation of circular perovskite tandem PV technologies, where LCA and experiments are integrated and guide each other, as shown in **Figure 1a, b**. We generate experimental data on materials and energy used in the fabrication of perovskite tandem PV and identify environmental hotspots.[53, 68] The overview, LCA modeling details, techno-economic analysis details for LCOE examination, and experimental details are presented in this section to describe the integrated approach.

As a preliminary step, a contribution analysis is conducted to identify the energy and environmental hotspots throughout the life cycle of perovskite-silicon and perovskite-perovskite tandems. The results of the contribution analysis with a "cradle-to-grave" system boundary inform the components worth recycling and guide the experimental investigation on the recycling process at the laboratory scale. The "cradle-to-grave" system boundary comprises four life cycle stages, including raw material acquisition, device fabrication, electricity generation, and EOL disposal. Landfill of used PV modules is selected as the EOL scenario method because 90% of decommissioned US solar panels end up in landfill based on estimation by Recycle PV Solar company. Consistent with previous studies,[35, 69] the contribution analysis results unmask the full-spectrum environmental profiles for the two investigated perovskite tandems using the Product Environmental Footprint (PEF) method.[70] We note that acetone could be easily recycled through distillation, while isopropanol could be recovered by multiple approaches, such as air stripping (over 90% recovery rate).[36, 37] We also note that non-precious metals, such as copper, are promising candidates for replacing silver embedded in the tandem stacks.[32]

Next, experimental investigations are implemented to explore feasible recycling routes of the hotspot materials. For instance, we experimentally show the ITO-coated substrate can be re-used after mechanically removing all other layers in the perovskite-perovskite tandem stack. The experimental investigation on the recycling route provides laboratory-scale data, including the amount of cleaning solvents (including acetone and isopropanol), the consumption of active materials and organic solvent to form the solution, energy consumption by the major processing steps, including sonication, UV/ozone treatment, heating, spin-coating, evaporation, etc. Here, the properly designed experiments enable data generation to bridge the data gaps when developing the complete mass and energy balances for the recycling process at the EOL of perovskite tandems, which have not been thoroughly investigated and documented in the existing data sources.

LCA modeling

In this LCA study, we systematically evaluate and compare the CED, carbon footprint, and full-spectrum environmental impacts according to the PEF method.[70, 71] In the system boundary, raw material acquisition, module manufacturing, and module recycling with material recycling are considered. The functional unit of this study is defined in terms of the module area – 1 m^2 of the module. The selection of the functional unit follows the existing literature in the field of PV life cycle assessment and does not imply the actual size of the commercial module.[72]

A complete LCI data set constitutes detailed material and energy balances throughout all the life cycle stages of the investigated perovskite tandem modules. The material and energy consumption are estimated based on the experiments of perovskite-perovskite tandem fabrication and recycling. We note that all the energy consumed is converted to electricity, following the existing literature.[73] Energy consumption for scaled-up processes and the corresponding material utilization rates are retrieved from the literature.[74]

In the life cycle impact assessment (LCIA) phase, the LCI results are interpreted into the midpoint impact categories based on the selected LCIA method. We focus on the CED,[75] GWP,[76] and the PEF environmental scores at the midpoint level.[70] The PEF approach is selected to unmask the full spectrum environmental profiles of the investigated perovskite tandem modules.[71] There is a total of 17 midpoint indicators, including climate change; ozone depletion; human toxicity, cancer effects; human toxicity, non-cancer effects; particulate matter/respiratory effects; ionizing radiation, human health; photochemical ozone formation; acidification; eutrophication, terrestrial; eutrophication, fresh water; eutrophication, marine; ecotoxicity, freshwater; land use; resource depletion, water; resource depletion, mineral, fossil, renewable; CED, renewable; and CED, non-renewable.

The LCIA results of the two tandem modules gain important insights regarding the contributions of materials and processing steps to different impact indicators. The primary impact contributors are identified as the energy and environmental hotspots through contribution analysis. Given a set of recycling frequencies, holistic scenario analyses are conducted to compare and evaluate energy yield potential, EROI, and GHG emission factor of perovskite tandem-based systems with a 30-year lifetime under different degradation scenarios. The results show how the two sustainability metrics vary with recycling frequencies (each corresponding to a specific requirement for module lifetime) under the explored degradation rates. Based on these results, more insightful suggestions are made toward the sustainable deployment and periodic replacement of tandem solar modules to accelerate their near-term market entry.

Levelized cost of electricity

The LCOE is defined as the ratio of the total lifetime cost to the lifetime electricity production as follows:

$$LCOE = \frac{CI + \sum_{t=0}^{N} \frac{OM(t,d)}{(1+r)^t}}{\sum_{t=0}^{N} \frac{E(t,d)}{(1+r)^t}}$$

where *CI* represents the initial investment required for the installation of the PV system, which includes costs associated with PV modules and installation, including BOS, inverters, labor, and permits among other factors. *OM* denotes the annual operation and maintenance and module rejuvenation costs in year *t*, while *E* stands for the annual electric power generated by the system in year *t*. *N* refers to the lifetime of the PV system, *d* signifies the annual module degradation rate, and *r* represents the discount rate. Following established literature,[49] we break down the total installation costs into components, including those proportional to the system area, the system power output, and a fixed investment per project.[49] For each module type we examined, we assumed a total of 36 modules with an individual area of 1.77 m^2 for residential systems and a constant area of 0.6 km^2 for utility-scale systems, aligned with existing literature.[49, 50, 56] Furthermore, both residential and utility-scale systems are projected to have a lifespan of 30 years.

Perovskite precursor solution

$Cs_{0.25}FA_{0.75}PbI_{2.1}Br_{0.9}$: A 1.1 M solution of $Cs_{0.25}FA_{0.75}PbI_{2.1}Br_{0.9}$ was prepared by dissolving 0.825 M formamidinium iodide (FAI, Greatcell Solar), 0.275 M cesium iodide (CsI, Sigma-Aldrich), 0.495 M lead bromide (TCI) and 0.616 M lead iodide (PbI_2, TCI) in a 4:1 (vol:vol) mixture of N,N-dimethylformamide (DMF, Sigma-Aldrich) and dimethylsulfoxide (DMSO, Sigma-Aldrich). The solution was stirred at 50 °C for 2 hours and filtered using a 0.22 μm PTFE membrane before use.

$Cs_{0.15}FA_{0.85}Pb_{0.5}Sn_{0.5}I_3$: A 2 M solution of $Cs_{0.15}FA_{0.85}Pb_{0.5}Sn_{0.5}I_3$ was prepared by dissolving 1.7 M FAI, 0.3 M CsI, 1 M PbI_2, 0.9 M SnI_2 and 0.1 M SnF_2 in a 3:1 (vol:vol) mixture of DMF/DMSO. The solution was stirred for 2 hours and filtered using a 0.22 μm PTFE membrane before use.

Perovskite-perovskite tandem fabrication

Patterned ITO glass substrate (KINTEC Company) was cleaned using 15 minutes of sonication in a 2% Hellmanex III (Sigma-Aldrich) solution, followed by 5 minutes in deionized water, 15 minutes in acetone, and 15 minutes in isopropanol. The substrates were dried using a nitrogen stream and subjected to a 15-minute UV/Ozone treatment before being transferred into a nitrogen-filled glovebox. A 1.5 mmol/ml solution of 2PACz in anhydrous ethanol was spin-coated at 3000 rpm (5s ramp) for 30s, followed by annealing for 10 minutes at 100 °C. After cooling down to room temperature, $Cs_{0.25}FA_{0.75}PbI_{2.1}Br_{0.9}$ perovskite was deposited onto the substrates by spin-coating at 2000 rpm for 10s (2s ramp) and 6000 rpm for 40s (4s ramp). Anhydrous chlorobenzene was dripped onto the spinning substrate 20s before the end of the program. The substrates were then annealed for 30 minutes at 100 °C. The substrates were then transferred to thermal evaporation for deposition of 20 nm of C_{60} (Sigma-Aldrich). A 25 nm SnO_2 interlayer was deposited by ALD (Picosun). Tetrakis (dimethylamino) tin (IV) (TDMASn, EpiValence) was used as a precursor and H_2O as a reactant. The precursor bubbler was heated to 75 °C and the chamber to 100 °C, the reactant vessel was kept at room temperature. The pulsing sequence consisted of a

0.6s pulse of TDMASn, 10s purge, 0.1s pulse of H_2O, 10 s purge, resulting in a growth rate of 0.1 nm/cycle. Following ALD, 1 nm of Au was deposited by thermal evaporation. The substrates were removed from the glovebox, and a filtered (0.45 μm membrane) 3:1 solution of methanol (Sigma-Aldrich) and PEDOT:PSS (Clevios Heraeus Al 4083) was subsequently spin-coated on top of the substrates at 4000 rpm (3.5s ramp) for 30s, followed by annealing at 140 °C for 20 minutes. After removing the substrates from the hotplate, they were immediately transferred to a nitrogen-filled glovebox. $Cs_{0.15}FA_{0.85}Pb_{0.5}Sn_{0.5}I_3$ was spin-coated at 5000 rpm (4s ramp) for 50s. Anisole was dripped onto the spinning substrate 25s before the end of the program. The substrate was immediately transferred to a hotplate and annealed at 100 °C for 10 minutes. After cooling down to room temperature, 20 nm of C_{60}, 8 nm of bathocuproine (Sigma-Aldrich), and 120 nm of Cu were deposited by thermal evaporation. Our experiments show that for a laboratory-scale solar cell deposition with 64.5 cm^2 total deposition area, the C_{60} deposition consumes 0.08 kWh for depositing 15 mg C_{60} with merely 0.01419% material utilization rate, while the ALD of the SnO_2 layer consumes up to 0.75 kWh. We note that the experimental data are subject to uncertainty, and it is possible that C_{60} deposition would be a hotspot for industrial-scale processing due to the stringent requirement for vacuum conditions and low material utilization efficiency.

Recycling of ITO substrates

ITO substrates were recycled by removing the active layers from finished devices using a razor blade. Once there was no visible residue, the substrates were cleaned and processed in an identical fashion to pristine ITO substrates. Even though we do not adopt the method of solvent dissolution in our experimental investigation, a supplementary calculation is implemented to compare the environmental profiles of these two methods, as shown in **Figures S14** and **S15**.

Current-voltage characteristics

Current-voltage characteristics were collected using a Rodeostat Plus Potentiostat (IO Rodeo) and an AM1.5 solar simulator (TS-Space Systems Unisim Compact Solar Simulator), calibrated using a KG5 filtered reference cell (ReRa Solutions B.V.). An aperture mask of 9.93 mm^2 was used to define the active area.

Resistivity test of ITO substrates

The resistivity of the ITO substrates was measured using a four-point probe setup, consisting of a 2450 Keithley SourceMeter and a four-point collinear probe.

Transmittance

Transmittance was measured using a Bentham PVE300 system in transformer mode. A dual xenon short-arc lamp and a quartz halogen lamp were utilized as the light source, with a swing-away mirror set to 750 nm. A 10 × 10 mm Si reference diode was used as detector. Transmittance was calculated by dividing the diode response in the presence of a substrate by the diode response without substrate.

Scanning Electron Microscopy

Scanning electron microscopy was performed using a Zeiss LEO 1550 FE-SEM with a field emission source operating at 2 kV acceleration voltage in the InLens mode.

Photoluminescence (PL) intensity and photoluminescence quantum efficiency (PLQE)

PL and PLQE measurements were recorded using an integrating sphere, following the three-measurement approach of de Mello et al.[77] In both PL and PLQE measurements a continuous wave temperature-controlled Thorlabs 520 nm laser was used to photoexcite samples. Excitation intensity was varied with an optical filter wheel. The emission was recorded using an Andor IDus DU420A silicon detector.

Solar Cell Stability Test

A 100-hour maximum power point tracking stability test of fresh and recycled substrates shows that there is no significant change in degradation rate when recycled substrates are used. Each curve is the average of five devices. Devices were encapsulated using glass and UV-curable epoxy glue immediately after fabrication. Stability measurements were carried out under inert atmosphere and AM1.5G illumination generated by a G2V Base-UV Sunbrick. Maximum power point traces were collected using a 32-channel Arkeo setup (Cicci Research).

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Author Contributions

FY and SDS conceived the research. XT developed the LCA models and conducted the simulations. BR and SDS designed and implemented the experiments regarding perovskite tandem fabrication, recycling, and characterization. XT, BR, SDS, and FY captured relevant parameters, including module degradation rate, recycling frequency, etc., defined the scope of scenario analyses, and co-developed the periodic module replacement model. XT and FY analyzed the results. XT, BR, SDS, and FY wrote the manuscript. All authors reviewed the final manuscript.

Declaration of Interests

SDS is a co-founder of Swift Solar, Inc.

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