Perovskite/silicon tandem photovoltaics: Technological disruption without business disruption ()

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ABSTRACT

Meeting the ambitious challenge of net-zero greenhouse gas emissions by 2050 and holding the average increase in global temperature below 1.5 °C necessitate the upscaling of readily available renewable energy sources, especially solar photovoltaics. Since the window of time to achieve this goal is closing fast, it is of paramount importance that we accelerate the decarbonization of the global energy system by increasing the power output of solar cells through advancing their power conversion efficiencies toward and beyond the Shockley–Queisser limit. In this Perspective, we describe how the integration of perovskites into the well-established silicon production infrastructure to form perovskite/silicon tandem photovoltaics can raise the rate of solar deployment. We present a holistic analysis of the technology from different perspectives, such as materials science, manufacturing, sustainability, and business, which highlights how the pairing of perovskite and silicon is advantageous at many different levels of consideration. Altogether, perovskite/silicon tandems deliver a technological disruption in efficiency while maintaining compatibility with the present photovoltaics industry, making it the fastest route to enhance the silicon market and rapidly address climate change.

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The United Nations Framework Convention on Climate Change (UNFCCC) stated in its January 2020 address that the world was on course for a global temperature rise of 3 °C, which is twice the internationally accepted target established by the 2015 Paris Climate Change Agreement. To have a 66% probability of remaining below the agreed upon 1.5 °C global warming goal, the cumulative global carbon dioxide (CO₂) emissions should not exceed about 200 × 10⁹ additional tonnes (200 GtCO₂) after the start of 2021.^{2,3} Unless the current annual rate of 40 GtCO₂ emissions⁴ is slowed, the remaining CO₂ emissions budget will disappear by 2026. Exceeding the emissions budget before a global carbon-neutral energy infrastructure is established will require actively removing greenhouse gases from the atmosphere through sequestration strategies or risk the dire consequences of global warming.⁵ Bold action is urgently required, and fortunately, many countries committed in 2020 to new or revised climate action plans to reach net-zero emission goals within the next 15-30 years. Achieving this aim can only be accomplished by shifting the global energy infrastructure as quickly as possible from fossil fuel to renewable sources.

The combined sectors of electricity generation, heating, and transportation currently account for about 75% of global greenhouse gas emissions.^{6,7} Therefore, the international community must seize upon any opportunities to accelerate the replacement of existing fossil fuels with renewables while also installing carbon-neutral power sources to meet the growing energy demand in developing nations. The 2021 World Energy Transitions Outlook published by the International Renewable Energy Agency (IRENA) presents a vision for transformational change of the global energy system that leverages mature and available renewable technologies for direct electricity generation as well as either indirect electrification through green fuels or sustainably sourced biomass to power various modes of transportation.8 By 2050, IRENA's "1.5 °C Scenario" plan specifies that 90% of an expected 30 TW of global electricity demand will be supplied by renewables, with solar photovoltaics (PV) and a combined onshore/ offshore wind accounting for about 14 and 8 TW, respectively. While other plans have been put forward by other international agencies such as the World Energy Outlook, there is a clear consensus that restructuring the global energy economy is a very great challenge that

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will require the coordination of many technologies and policies, and that the path forward will rely heavily on utilizing the world's most abundant energy resource: sunlight.

Over decades of development, the combined efforts of research institutions and the solar photovoltaics industry have led to exponential price reductions in PV modules. During the last ten years alone, the cost of PV has decreased by over 85%, 10,11 making solar energy less expensive than fossil fuel sources in most locations worldwide.⁸ This dramatically reduced cost, together with the implementation of forward-looking energy policies in many nations, positions solar PV as the ideal affordable clean energy for both the electricity generation and transportation sectors. However, as of 2020, the cumulative global installed capacity of PV was only 0.7 TW,13 which means that aggressive growth in the production and installation of PV is required to meet IRENA's 2050 solar energy target.8,14 Currently, the principal solar PV technology is based upon crystalline silicon (c-Si), which accounted for 95% of the production volume in 2019. 10,111 While the cost of silicon PV is expected to continue falling in the near term, a practical cell efficiency limit of about 26% in production is expected in the future for c-Si PV, and disruptive innovation will be required to surpass it if further energy cost reductions are to be achieved. ¹² A technologically mature strategy to boost the efficiency of c-Si modules beyond this value is to pair it with another wider bandgap solar material that selectively absorbs a higher energy region of the solar spectrum. By stacking this material above the lower bandgap c-Si, a tandem configuration is achieved, which can overcome the fundamental Shockley-Queisser limit for a single-junction device made with either material alone. This strategy has already been identified in the International Technology Roadmap for Photovoltaics (ITRPV), which forecasts the entry of Si-based tandems into the PV market after 2023. While several options exist for pairing a wide bandgap top cell with c-Si, the most promising candidates are metal halide perovskites, which has been the subject of intense research and development by the scientific community over the past 10 years. At Oxford PV, we are commercializing perovskite/Si tandems and have already practically demonstrated the potential for this technology with our recently certified 29.52% device, which has surpassed both the efficiency record of all non-concentrated single-junction solar technologies ¹⁵ and the theoretical limit 16,17 of silicon alone. We believe that the pairing of perovskite and c-Si in a tandem device is ideal at many different key levels of consideration, and that it embodies the quickest route for the successful entry of tandem PV into the terrestrial energy market. In this Perspective, we will discuss how perovskite and silicon are well matched for tandem photovoltaics from the crucial perspectives of materials science, manufacturing, sustainability, and business, and how their union represents a technological disruption that will help accelerate global PV capacity growth and mitigate climate change.

The multi-junction photovoltaic device is a well-known and proven technological concept to increase the efficiency beyond the theoretical Shockley–Queisser limit for single-junction solar cells. An archetypal example that has been widely utilized for space applications, where the need for high-efficiency outweighs cost, is triple-junction solar cells made from III to V semiconductors. While III–V multi-junction devices are sold by several manufacturers, such as Spectrolab and Azur Space, for terrestrial use and can achieve efficiencies around 40%, ^{18–20} these most often employ concentrating optics to minimize wafer area and offset some of the high fabrication costs

arising from both sourcing these exotic materials and their required epitaxial growth.²¹ However, competing for market share in the terrestrial photovoltaics market requires minimizing the cost per watt value of an installation. Therefore, the simplest multi-junction configuration that provides efficiency gains without adding excessive cost to the balance of systems is a non-concentrated tandem design, where two sub-cells of inexpensive solar absorbers with different bandgaps are combined. These sub-cells can either be uncoupled semi-transparent devices that are mechanically stacked to form a four-terminal (4T) solar cell or serially connected in a monolithically integrated twoterminal (2T) configuration. Unlike 2T tandems, 4T tandems do not require current matching between the two sub-cells, which widens the material combinations of the sub-cells that can achieve high efficiencies.²² Although there are wiring designs that allow mechanically stacked tandems to voltage-match with only two terminals,²³ traditional 4T designs require twice the number of inverters of singlejunction and 2T tandem modules, and in a current market where the balance of system costs outweigh that of the module, increasing the cost of the power electronics is economically prohibitive. ^{22,24,25} In contrast, 2T designs are fully compatible with the standard power electronics of mainstream single-junction silicon modules and, therefore, represent the most cost-effective tandem configuration.

In 2T tandems, the maximum achievable efficiency is only a function of the incident solar spectrum and the bandgaps of the two sub-cell absorber materials.²⁶ It can be determined by performing the detailed balance calculation for a given bandgap combination. Figure 1(a) depicts the results under an AM1.5G solar spectrum when a perfect reflector is placed at the back of the bottom cell, and photon recycling from the top to the bottom cell material is considered. From Fig. 1(a), it is clear that the bandgap pairing determines the efficiency ceiling of the different tandem options, where the judicious combination of a bottom cell with a bandgap between 0.8 and 1.2 eV and a top cell with a bandgap between 1.5 and 1.8 eV could achieve efficiencies exceeding 40%. Within established PV technologies, copper indium gallium selenide (CIGS) and silicon display bandgaps best suited for the high-efficiency range of the bottom sub-cell. If we consider that Si represents the backbone of the current PV market with over 60 years of development, it is only logical to select it as the preferred bottom sub-cell candidate material. Regarding top cell options, there is a wide variety of materials with the preferred bandgap range of 1.5-1.8 eV, including well-known III-V semiconductors as well as emerging absorbers such as organic semiconductors and perovskites.

While the detailed balance approach provides a useful guidance for selecting the bandgaps of the sub-cell absorbers, predicting if a material combination is commercially competitive as a tandem cell requires considering practical factors such as module cost and achievable power conversion efficiencies. In general, the efficiency gain of a tandem solar cell must be sufficient enough to outweigh the additional associated manufacturing cost. Otherwise, a single-junction module of one of its constituent sub-cells would be a more economical option. A useful metric, previously described by Peters *et al.*²⁴ and adopted recently by Yu *et al.*²⁸ specifically for Si-based tandems, is the relative tandem cost benefit, ζ , which is a function of the cost and power output of the tandem, top, and bottom cells. As is illustrated in Fig. 1(b), when ζ is positive, the cost (relative to the area-related balance of systems, BOS_A) of a single-junction module based on the top and bottom sub-cells is sufficient to make a tandem configuration economically

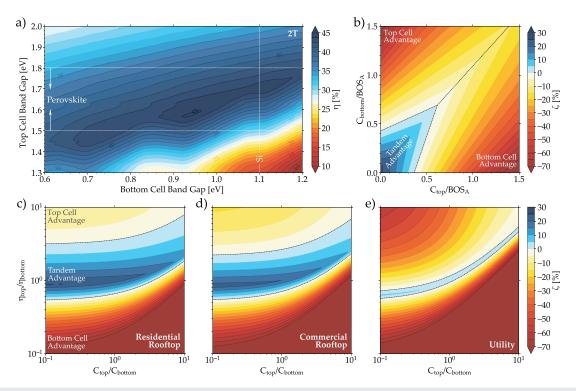


FIG. 1. (a) Theoretical power conversion efficiency, η , for any combination of bottom and top cell bandgaps in a two-terminal configuration as determined by detailed balance calculations with photon recycling from the top to bottom cells and a perfect reflector at the backside of the rear cell. (b)–(e) The cost-competitiveness of a tandem system compared against single-junction modules made with either the bottom or top cell material alone as quantified by the relative tandem system cost benefit, ζ . Panel (b) presents the cost-competitiveness as a function of the cost of the top, C_{top} , and bottom, C_{bottom} , modules relative to the area-related balance of systems, BOS_A , for a 22% efficient silicon device, a 21% efficient 1.7 eV bandgap perovskite device, and a 30% efficient perovskite/Si tandem. Panels (c)–(e) present the cost-competitiveness as a function of the relative cost and efficiencies of the top (a 1.7 eV absorber) and bottom (silicon) modules assuming BOS_A values appropriate for (c) residential rooftop, (d) commercial rooftop, or (e) utility applications.

advantageous. Following the methodology explained in the supplementary information, ζ was rewritten and plotted as a function of the efficiency and cost ratios for top and bottom cell, $\eta_{\rm top}/\eta_{\rm bottom}$ and $C_{\rm top}/C_{\rm bottom}$, respectively. Figures 1(c)–1(e) shows the relative cost benefit ζ , for a 2T tandem solar cell with a 1.7 eV top cell and a 1.1 eV Si bottom cell as a function of the efficiency and module cost ratios for residential rooftop, commercial rooftop, and utility applications. A bandgap of 1.7 eV was selected based on the results shown in Fig. 1(a) to maximize the theoretical efficiency achievable by a 2T tandem employing Si for the bottom cell.

The trends observed in Figs. 1(c)-1(e) are common regardless of the specific photovoltaic application: the deposition of an inexpensive or an efficient top cell on a Si bottom cell will not necessarily make a tandem economical. To achieve a maximum cost benefit from a tandem solar cell, the constituent sub-cells should have similar efficiencies with the top cell also being as inexpensive as possible. However, there is always a pathway for a more expensive top cell to still be advantageous if its efficiency sufficiently surpasses the bottom cell. This illustration offers an explanation of why Si-based tandems have not yet seen uptake into the PV market. Although III-V/Si solar cells are efficient, the high fabrication cost of a III-V semiconductor top cell places it on the far right of Figs. 1(c)-1(e). Likewise, despite their low module manufacturing cost, the historically low efficiencies of

organic-based photovoltaics would tend an organic/Si tandem toward the lower left corner. However, recent developments of high-efficiency pairings of new polymer donors and non-fullerene acceptors could favorably shift its positioning in the future. The parameter space available for tandem advantage is similar for both rooftop applications, but narrows considerably at the utility level where the BOS_A cost reduces substantially. Even so, a benefit exists for all applications when a 1.7 eV top cell solar material matches both the cost and efficiency of silicon, with stronger advantage occurring when the top cell cost is further minimized with an efficiency slightly below that of silicon [Fig. 1(e)].

Following on the previously outlined criteria, perovskites offer an ideal choice for the top cell in a Si-based tandem. Their bandgap can be easily adjusted by varying material composition, with achievable values ranging from 1.22 eV, through partial Sn²⁺ substitution at the B-site^{31–34} of the ABX₃ perovskite lattice, up to 2.4 eV by altering the halide content at the X-site.³⁵ The 1.5–1.8 eV range previously identified for the top sub-cell to maximize potential efficiency is well-suited for perovskites for several practical reasons. First, the lower end point of this range can be achieved by formamidinium lead triiodide,³⁶ and all other wider bandgaps are accessible through several different combinations of A-site and X-site ions, which provides flexibility in materials selection.^{37–39} Second, a phenomenological voltage pinning

effect has been observed for bandgaps between 1.8 and about 2.2 eV, which limits the usefulness of current materials in this range, whereas bandgaps below 1.8 eV are unhindered and display open-circuit voltages that scale linearly with bandgap. Finally, halide segregation under illumination is a well-known phenomenon in perovskites with wide bandgap compositions; however, several compositions within this range consist of either neat iodide or have been identified to be stable under photoexcitation. According to the 2T-detailed balance calculation results presented in Fig. 1(a), a perovskite top cell with a bandgap around 1.7 eV is perfectly matched with a 1.1 eV silicon bottom cell to achieve theoretical efficiencies above 43%. However, there is a healthy margin of variation for the top cell bandgap that still achieves high efficiencies, which provides some flexibility in selecting the precise perovskite composition.

In terms of efficiency, metal halide perovskites have developed at the fastest rate of any solar cell technology, from an initial reported value of 3.9% (Ref. 46) to above 25% (Refs. 15 and 47) in just over a decade. This rapid evolution can be largely attributed to their excellent optoelectronic properties and relative ease of processing. In many respects, perovskite is a foil to c-Si. Whereas silicon is a weakly absorbing material that requires hundreds of micrometers of thickness to fully absorb sunlight due to its indirect bandgap, perovskites are strongly absorbing, direct bandgap semiconductors that require less than a micrometer of material thickness. Silicon is a covalently bonded material that requires energy-intensive fabrication processes to synthesize high-purity single-crystals necessary for low defect densities, whereas perovskites are ionic semiconductors that are inherently defect tolerant and can be processed at low-temperatures into poly-crystalline films. Whereas the charge transport properties of c-Si arise from extrinsically doping different regions of the absorber thickness, perovskites are effectively intrinsic semiconductors with inherently high ambipolar charge carrier mobilities under illumination. While the differences in material properties are striking, the manifested end result is similarhigh-quality optoelectronic materials that can deliver power conversion efficiencies around 26% using low-cost fabrication routes. Therefore, the combination of perovskite and silicon comprises an ideal match from a materials perspective to deliver a cost-effective tandem solar cell capable of competing in a market dominated by silicon.

Before we move to the next paragraph, it is worth highlighting that although the most efficient combinations with perovskite are achieved by selecting either CIGS or c-Si as the bottom cells, other potential alternatives⁴⁸ are possible with wider bandgap bottom cell materials, such as CdTe⁴⁹ or GaAs,⁵⁰ due to the tunability of the perovskite bandgap to even higher values. Notably, such wider bandgap perovskite top cells can also be paired with narrow bandgap perovskite bottom cells to form all-perovskite tandem devices which have reached research efficiencies near 25% (Ref. 51) and are also a subject of intense research and development by the perovskite community.

We have demonstrated that the material properties of perovskite and silicon offer a highly promising combination for a theoretically cost-competitive 2T tandem module. However, commercialization of an actual product requires that the manufacturing processes for both sub-cells are technologically compatible, scalable, and economical. Toward explaining why silicon provides an excellent foundation for building a first-generation terrestrial tandem, it is helpful to briefly consider how wafer-based silicon has become the dominant solar

technology, accounting for 95% of global PV production in 2019. 12 At first glance, its weakly absorbing character and its requirements for high-purity and extrinsic doping make silicon an unnatural solar absorber choice solely on the basis of inherent materials properties. What is needed to explain its emergence is some historical context. The theoretical and technological foundations for silicon-based PV were established early in its development as a result of the rapid advancements achieved by the semiconductor community for solidstate electronics and computing applications. The quickly advancing technological readiness of silicon solar cells, coupled with the need for lightweight and reliable power for burgeoning applications in space, provided both the means and the market to initialize solar energy applications in the 1960s. Since the 1970s, silicon PV has followed an extraordinary trajectory of technological development that is often expressed as an experience curve where the inflation-adjusted module price per watt is plotted against the cumulative PV production. Figure 2(a) reproduces the experience curve with logarithmic scales for both axes using data published in the 2019 ITRPV report, 10 which clearly demonstrates that exponential price reductions have accompanied the exponential growth of manufacturing capacity. While this curve undoubtedly results from the complex confluence of many factors, a paper by Kavlak et al. has developed economic models to identify the key low-level and high-level factors that have driven the learning rate.⁵² In their analysis, the authors identified that the primary highlevel motivator for the development from 1980 to 2001 was public and private investments in research and development, which contributed to about 60% of module cost reductions due to efficiency improvements, reductions in silicon and non-silicon material costs, and increases in wafer size from growing large crystal ingots.⁵² Whereas cost reductions due to economies of scale contributed to roughly 20% of the module cost change in this period, it became the primary highlevel driving factor from 2001 to 2012 and contributed to nearly 50% of the cost reductions across the industry.⁵² As is shown in Fig. 2(a), the module cost per watt decreased steadily by about 24% for every doubling of cumulative production capacity prior to 2008. However, the learning rate has quickened to about 40% in the past decade. This has been attributed to steady module efficiency improvements, further increases in wafer size and materials utilization, and coordination between manufacturers, which has led to increased standardization and the establishment of common supply chains.¹² The outcome is a streamlined industry that has reduced the module price in 2019 to only \$0.23/W_p¹⁰ and gives no indications that price reductions will not continue in the near future. Therefore, selecting silicon as the bottom cell in a tandem product provides the opportunity to leverage the industrial inertia of the silicon PV industry that has been building up over decades of development by riding the experience curve to bottom cell cost reductions and efficiency gains. Similarly, a wafer-based tandem capitalizes upon the availability and affordability of efficient and high-throughput production and automation tools that support the silicon PV industry, 12 while also maintaining the cell format that is already familiar to module manufacturers and installers. Therefore, a silicon-based tandem acts as a drop-in replacement to the existing production infrastructure, which, we believe, represents the quickest delivery mechanism for its adoption and integration into the global energy economy.

For a perovskite top cell to be successfully integrated with silicon into a tandem, deposition methods must also be available, which are

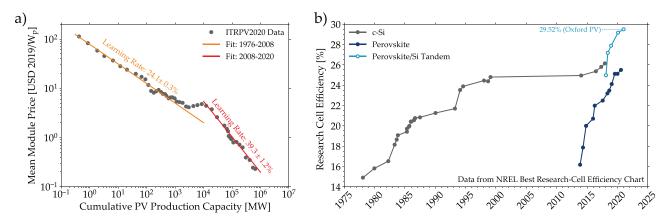


FIG. 2. (a) The historical experience curve of silicon photovoltaics shows how module price reductions accompany increasing cumulative production capacity. Data points are from the ITRPV 2019 Results. (b) The record-certified research efficiency solar cells for c-Si, perovskite, and perovskite/Si tandems using data courtesy of the National Renewable Energy Laboratory (NREL). (5)

scalable, compatible, and economical. Perovskite is a known soft material with high tolerance to defects. These unique properties make its material deposition compatible with almost all known scalable deposition techniques. Vapor-based processes include traditional vacuum thermal evaporation, 53-55 chemical vapor deposition, 56,57 and high-speed vapor transport deposition akin to organic vapor phase deposition.⁵⁸ While spin-coating remains a preferred technique for lab-scale research devices, scalable solution-based techniques have been successfully utilized with sheet-to-sheet methods like blade coating, 59,60 inkjet printing,⁶¹ and screen printing,⁶² or roll-to-roll compatible techniques such as slot-die coating^{63,64} and gravure printing.⁶⁵ Many of these processes are relatively simple, affordable, accessible, and capable of producing high-quality perovskite thin-films at low-temperatures, which have contributed substantially to the rapid development of high-efficiency perovskite solar cells by the international research community. Not all of these methods are compatible with wafer-based manufacturing on silicon substrates, but most of them are, which give plenty of choice for designing production lines for perovskite/Si tandems.

The combination of silicon and perovskite into an integrated tandem product capitalizes on both the experience and momentum of the silicon PV industry garnered over decades of development as well as the incredible growth potential of perovskite. This concept manifests itself elegantly in Fig. 2(b), which reproduces the historical advancement of record research cell efficiencies as chronicled by the United States National Renewable Energy Laboratory (NREL) for these technologies. Over decades of research and development, lab-scale c-Si devices have gained steady efficiency improvements, but progress has nearly plateaued in recent years. In sharp contrast, the trajectory of perovskite development has nearly matched silicon in less than a decade of development. When combined into a tandem, the resultant efficiency gain constitutes a technological jump above the practical limit that c-Si can foreseeably achieve, and the upward trajectory is indicative of the headspace available for further performance growth.

We have argued that perovskite/Si technology offers the fastest route to deploy cost-competitive tandems capable of increased power generation per installation. However, as with any new technology, its readiness is limited by the maturity of manufacturing processes and a sustainable chain of supply and operations over its full life cycle. A valuable tool to quantify the environmental impact of any technology is the life cycle assessment (LCA) methodology, which aims to consider every phase of a product's lifespan across a standard set of key impact categories. One of these is climate change, which quantifies the global warming potential embedded in the product over the course of its lifetime by many factors, including its associated raw materials, production processes, transportation, installation, and operations. While, undoubtedly, any given renewable energy technology will have a lower carbon footprint than its fossil fuel counterpart, maximizing its opportunity to decarbonize the energy sector requires understanding and minimizing its own contributions to global warming. Since photovoltaics do not generate any greenhouse gases during operation, the largest contributor to their carbon footprints is usually the primary energy consumption embodied in their production; the level of which can strongly vary between technologies.^{66,67} In the case of c-Si, LCA studies reveal that the carbon footprint is relatively high compared to other PV technologies due to the energy intensive Czochralski process that is used to grow high-purity solar grade single-crystal ingots, which accounts for up to 90% of the total energy used in silicon cell production. 66,68 In contrast, applying the same assessment methodology to perovskite technologies has led to very different results, where the carbon footprint and overall environmental impact are decreased substantially by the use of low-temperature fabrication processes. 60 There is general consensus amongst LCA studies for single-junction perovskite solar cells that even though the required power consumption is lower than c-Si, it still remains the primary hot spot for environmental impact in part due to the high level of energy embedded in sourcing and depositing transparent conductive oxides, ^{66,68} whereas the impact from metals contained in the absorber layer is relatively small.^{69,70} Now, if we take into consideration both parts, it will become obvious that the addition of a top cell to the Si will only increase the energy consumption, and thereby the carbon footprint. However, photovoltaic performance and lifetime play a key role in the assessment, and since the additional impact of the top cell is relatively low, the surplus clean power produced by a tandem module relative to c-Si alone can offset the overall carbon footprint. A recent report shows that the greenhouse gas emission factor of a 25% efficient perovskite/c-Si tandem would break even with c-Si alone if its lifetime is about 28 years.

Tandems with higher efficiencies or longer lifetimes would produce enough clean power to partially compensate for the high embodied energy in the c-Si bottom cell and offer a greenhouse gas emission factor below that of the dominant PV technology.

It is important to note that the carbon footprint of a solar technology is strongly linked with its embedded energy largely because the current energy mix for electricity still relies heavily on fossil fuel sources in most places. As nations decarbonize their energy infrastructure and the electricity used by PV manufacturers becomes cleaner, the carbon footprints of all PV will correspondingly reduce in the manner of a virtuous cycle as we approach a future where 90% of global electricity is supplied by clean technologies. However, even now, there are silicon manufacturers, and certainly more will follow, that either fully source their electricity from renewable sources or employ less energyintensive fabrication processes, which both provide the immediate opportunity for the carbon embodied in silicon-based PV to be dramatically reduced. For instance, NorSun is a monocrystalline silicon ingot and wafer producer, which uses hydroelectric power and natural cooling water in its production line,⁷¹ whereas NexWafe and 1366 Technologies directly fabricate kerfless silicon wafers using processes that have an inherently lower embodied energy than traditional routes.^{72,73} Through a combined strategy of increasing both module efficiency and the fraction of renewably sourced electricity used in production, there is a pathway for perovskite/Si tandem technology to accelerate decarbonization of the energy sector.

Beyond the impact associated with solar cell manufacture, a cradle-to-grave assessment requires considering the sourcing of raw materials at the beginning of the supply chain and product disposal at its end-of-life. With regard to the latter, there are different approaches in PV waste management worldwide, depending on each country's regulatory framework for general waste and PV waste treatments. The European Union (EU) and United Kingdom have enacted Waste Electrical and Electronic Equipment (WEEE) directives that include specific regulations and policies for managing the end-of-life of PV modules.⁷⁴ In parallel, voluntary schemes have been established and fully financed by leading PV manufacturers, such as PV CYCLE, which undertakes the collection and recycling of defective and end-of-life PV modules, in compliance with EU legislation.⁷⁵ Notably, the EU is the only geographic region where PV-specific obligations exist. No federal regulations exist in the United States, although the state of California's Department of Toxic Substances Control (DTSC) has taken over the responsibility of solar waste treatment and a takeback program is under consideration in Washington State. 76,77 Similar to PV CYCLE and in close collaboration with it, Recycle PV Solar was founded in 2018 with the main goal to recycle up to 95% of a discarded module's material.⁷⁸ In Japan, the government is responsible for overseeing solar panel recycling, and in parallel, industrial research is conducted to develop recycling technology following the respective European standards. 79 In countries like China and South Korea, although the need of special legislation has been realized, there are still no strong policies regarding PV waste management. However, many companies worldwide have their own internal environmental management system, for example, by participating in the ISO 14040 framework related to LCA. 80 Voluntary takeback or product stewardship programs offering the collection of solar panels for recycling either by the manufacturing company or by external collaborating organizations like PV CYCLE⁸¹ are not unknown. Regardless of where perovskite/Si tandems are

marketed, at a minimum, producers and sellers will need to fulfill all local regulations and ensure that the modules are disposed of safely.

At the opposite end of product life, there are crucial aspects to consider about sourcing materials and establishing a supply chain for a new technology. The abundance and commercial availabilities of the technology's constituent elements strongly influence resource sustainability, the rate of commercialization, and environmental impact. Reaching IRENA's 1.5 °C Scenario target of 14 TW PV by 2050 requires an average of about 0.46 TW to be deployed each year, roughly three times higher than the current rate.⁸ Practically manufacturing at such a massive scale in a short timespan requires not only employing elements with sufficient reserves but also the existence of primary supply chains capable of increasing production to support PV capacity goals. To place these considerations into context, we apply the approach taken by Jean et al. 82,83 to estimate the amount of primary material required to produce the absorber layer in representative device embodiments for several leading photovoltaic technologies. This is accomplished by first calculating the areal mass density (i.e., the mass per solar cell area) for each key element in the absorber layer of each solar technology considered using the material composition and thicknesses reported in literature for state-of-the-art c-Si, 84 perovskite, 85 perovskite/Si tandem, 86 CdTe, 87 CIGS, 88 GaAs/Si tandem, 21 and GaInP/Si tandem²¹ builds. Plotting the areal mass density for each key element against its average concentration in the Earth's crust⁸⁹ in Fig. 3(a) provides a means to compare the quantity required for a solar installation against a rough measure of terrestrial abundance. The elements that tend to the lower right corner are in the prime position of being both abundant and requiring a low mass loading in a solar cell technology, whereas the elements that tend to the upper left are those that are scarce, highly loaded, and will likely constrain the production capacity of its parent technology. Since its indirect bandgap makes silicon a weak absorber, the absorber layer thickness is at least 150 μm thick, which requires the highest areal mass density of the technologies considered here. However, it has the key advantage of having the second highest crustal abundance of all elements, surpassed only by oxygen, and therefore high areal mass density is inconsequential. In the case of perovskite, the limiting element is iodine. However, it is noteworthy that the perovskite/Si tandem build86 requires about half as much iodine loading as the single-junction analog⁸⁵ because some of the halide content is replaced with bromide to widen the bandgap, and the required perovskite layer is thinner, which maximizes the overall device current by allowing additional photons through to the silicon bottom cell to achieve current matching between the two subcells. Second, iodine is more abundant and requires less loading than the limiting element(s) for most of the other technologies considered: Te and Cd for CdTe, Se and In for CIGS, and In for GaInP/Si.

While Fig. 3(a) provides some context for comparing the material demand and abundance of solar technologies, the metric of crustal abundance does not capture the practical availability of raw material supply chains for PV manufacturing. To address this, Fig. 3(b) presents an estimate of the time necessary to produce the required material for solar deployments of 1, 14, and 30 TW of each technology based on the reported efficiencies for those builds^{21,84–88} and the current production rates of primary materials.⁹⁰ It is important to highlight that these calculations have been made assuming a full deployment of each technology and not partitions in a shared market. The analysis is based on the approach that each technology is a

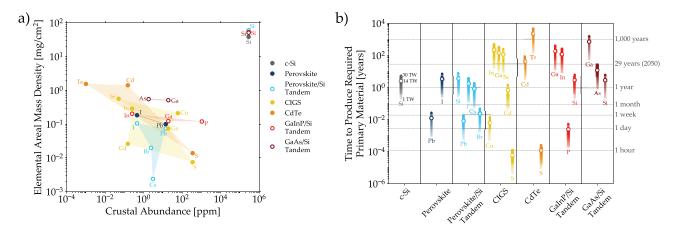


FIG. 3. (a) Comparisons of the elemental mass per square meter of solar cell required for the key elements comprising the active layers of current photovoltaic technologies against each element's abundance in the Earth's crust. (b) The time it would take to produce the required material to create between 1 and 30 TW of PV from current photovoltaic technologies at the current rate of global primary production. These calculations use material compositions and thicknesses reported from literature for state-of-the-art device builds for c-Si, ⁸⁴ perovskite, ⁸⁵ perovskite, ⁸⁵ clGS, ⁸⁸ CdTe, ⁸⁷ GalnP/Si, ²¹ and GaAs/Si tandems. ²¹

separate entity that could independently supply the market with the required PV capacity, setting in this way a baseline metric of how easily established materials supply could accommodate increased demand. As an aside, we do not consider here the scarce but crucial element of silver, which is currently common to all solar technologies as a metalization material, instead only focusing on the elements specifically contained in absorber materials. While there is concern over whether silver could meet the future demand placed on it by renewable technologies, 12,82,91 this will need to be collectively addressed by the industry. Taking the 29 years timeline as the reference to meet 2050 targets, Fig. 3(b) demonstrates that producers of Si and perovskitebased technologies would be able to source all the required elements well in advance, whereas this would not be achievable at current production levels for any of the other technologies. The analysis presented in Fig. 3(b) provides a more realistic snapshot of the materials that would limit production for different PV technologies than Fig. 3(a), which is highlighted by the case of gallium. Although Ga is, in principle, abundant with a crustal concentration of 19 ppm, 89 it is practically scarce because it is only produced as a by-product of bauxite and zinc mining operations, and less than 10% of the Ga content is recoverable.90 In the case of all the other thin-film technologies considered, the limiting element (i.e., the one that requires the longest production time) is mined as a by-product, which can limit the practical opportunity to scale-up since it is dependent on operations for other principal ores. In contrast, the limiting element for perovskite/Si tandems is the silicon required for the bottom cell. Production of the perovskite top cell absorber is limited by iodine, which can be mined either as a byproduct of nitrate deposits found in salt flats, extracted from underground brines associated with gas fields, or harvested from seaweeds that accumulate it from the ocean. 92 Furthermore, perovskite's second-most limiting element, cesium, is mined as a primary material, but the current market is so small that active mining is limited even though the world's known reserves are vast in comparison. 90 In fact, the mining operation at the world's largest deposits of its principal ore, pollucite, in Canada ceased active production in 2015 because demand could be met by its stockpiles.⁹³ While the current production rates and size of known reserves are sufficient for perovskite/Si tandems to wholly supply the projected 30 TW (Ref. 8) of global power demand by 2050, it is entirely feasible that iodine and cesium mining operations could scale-up in direct response to increased demand from PV, if that were warranted, and the results could be economically favorable for the technology.

While we have shown that perovskite and silicon are well matched for two-terminal tandems from several different scientific and technological perspectives, in order for this product to contribute to the decarbonization of the global energy economy, it must also be profitable. Aside from all the practical engineering challenges of developing a new solar product, one of the biggest hurdles for startup companies is successfully entering the market. On the one hand, the astonishingly rapid reductions in the cost per watt of solar modules by the photovoltaics industry are a major achievement in addressing climate change. On the other hand, fierce competition has generated an intense pressure to reduce costs across the industry, which means that profit margins are razor thin. Whereas established manufacturers benefit from the inherent cost reductions that accompany high volume production, newcomers to the PV market are placed in a vulnerable position where they must first offer an initial cost-competitive product that is fabricated at low production volumes and then quickly scale-up manufacturing to match momentum with the industry, so that they are not out-paced before the window of opportunity closes. In this current environment, a business model based primarily on cost reductions is unlikely to be successful, which is why our approach at Oxford PV is one of product differentiation on the basis of efficiency. By combining the proven incumbent silicon technology of the PV industry with an efficient and cost-effective wide bandgap perovskite top cell, the resultant tandem yields a product with the familiar form factor of silicon, but with a technological jump in efficiency that is out of reach for silicon alone. Because perovskite/silicon tandems will be able to offer a 20%-30% boost in relative efficiency over current PV modules with only a marginal cost added primarily at the cell level, it becomes particularly attractive for space-constrained applications, such as rooftop installations, where sun-facing surface area is at a premium.

This is clearly illustrated by the cost-competitiveness model shown in Figs. 1(c)-1(e) by rooftop applications having a larger parameter space than the utility scenario that allows for more expensive top cells to still maintain tandem advantage. In this setting, the balance of system is by far the largest contributor to the overall cost of the installation, and this is leveraged by the additional power output that is provided by perovskite/Si tandem cells, thereby driving down the overall cost per watt and creating a substantial added value over the lifetime of the system. This added value, after considering the full economic costs and benefits by way of a discounted cash flow analysis, is such that the net present value of the system would match that of a conventional silicon module installation even if the tandem modules were sold at a higher price point. It, therefore, ultimately makes economic sense for both the producer of the cells, who would achieve enhanced margins, and the end consumer who would enjoy cheaper electricity in the long-term and a faster payback period. The fact that this tandem technology can still be an economical choice even with a premium on the price may also provide the opportunity to select silicon wafers for the bottom cell that have a low carbon embodiment, as was discussed previously, which would dramatically reduce the overall carbon footprint of the technology. Furthermore, rooftop installations are expected to be significant contributors to the future energy economy, with IRENA's roadmap anticipating that they will supply 40% of the global electricity contribution from PV.8 Likewise, the demand for efficient rooftop modules is expected to rise as more governments join those of Dubai, California, and Baden-Württemberg in enacting mandates for the installation of rooftop PV in new building projects. 94-97 Therefore, the efficiency gains tendered by perovskite/Si tandems provide a viable pathway to directly enter the rooftop market and immediately contribute to accelerating the decarbonization of electricity.

Having passed through market entry, any new PV technology will establish a new experience curve that must match the industry's trajectory of cost per watt reductions to remain competitive. Perovskite/Si tandem technology also offers key advantages for this phase of business development. First, there is still much potential for overall efficiency gains by improving the perovskite top cell, with practical efficiencies of up to 33% currently envisaged. 98 Second, as production scales up to gigawatt production and beyond, cost savings associated with growing to new economies of scale will allow for additional reductions in cost per watt as this technology establishes its own learning curve alongside silicon and other thin film technologies. It is likely that the perovskite top cell will undergo a development pathway similar to silicon in its phases of growth along its experience curve initially benefitting by research and development driving efficiency improvements and engineering-driven cost reductions and later from savings associated with economies of scale cost reductions. Third, due to the drop-in compatibility of wafer-based tandems with the dominant wafer-based silicon PV technology, perovskite/Si tandems will also benefit from reductions in balance of system costs, as well as silicon efficiency gains and cost reductions achieved by the greater industry. The convergence of all of these factors should allow perovskite/Si tandems to maintain an advantage as it grows such that the levelised cost of electricity (LCOE) produced by the technology will decrease in time and eventually drop below that of single-junction silicon, which opens up the market opportunity for utility scale deployment. Calculations of perovskite/Si tandems suggest that the energy yield will maintain an advantage over silicon regardless of whether tracking is

used at utility level installations. ⁹⁸ Therefore, a strategic business pathway exists for this technology to contribute to advancing solar power generation in both distributive rooftop and localized utility scenarios without disrupting the firmly established industrial mechanisms currently tooled for wafer-based module production and installation. Finally, by virtue of the advanced level of technological readiness of our perovskite/Si tandem technology, our first products will be able to help accelerate the decarbonization of the energy sector as early as 2022 when we begin commercial production at our facility in Germany.

Limiting climate change is a race against time and necessitates an unprecedented increase in solar capacity to meet projected global energy demands. As silicon PV modules approach their practical efficiency limit, the acceleration of solar installation will inevitably slow since maintaining the current learning rate will become increasingly challenging through cost reductions alone. While perovskites comprise an incredibly promising emerging PV technology, penetrating the PV market and contributing toward climate change goals require competitive advantages at many technoeconomic levels over established commercial PV technologies. At the fundamental level, the optoelectronic properties of perovskite and silicon are well matched, which results in efficiencies and compatible manufacturing methods that allow these materials to practically integrate into a cost-competitive tandem product. Furthermore, the inertia generated over decades of development in the silicon PV industry could be leveraged by a perovskite/Si tandem that will confer cost benefits and allow it full access to the industrial machinery currently used to package and install modules for consumer use. Likewise, we have demonstrated that raw material supply chains exist that can accommodate a quick upscale in production, and pathways are available to reduce the carbon footprint below that of current silicon PV. These factors combined with a business model, which support market entry and growth on the basis of a technological jump in efficiency over incumbent technologies, provide the fastest avenue for terrestrial tandem technology deployment and do so by enhancing the infrastructure generated by silicon instead of competing with it. At Oxford PV, we have already demonstrated that high efficiencies are achievable with our certified 29.52% perovskite/silicon tandem in December 2020 and have been upscaling our fabrication capacity with plans to enter volume production in 2022. Therefore, perovskite/Si tandems not only offer several critical advantages but currently exist at a high level of technological readiness, which, we anticipate, will boost the power output of solar PV installations in the near future and help transform our fossil-fuel energy infrastructure into one that is clean, affordable, and sustainable.

AUTHORS' CONTRIBUTIONS

C.K. and M.T.K. contributed equally to this work.

See the supplementary material for details on the methods used for calculations of perovskite/Si tandem cost–competitiveness, learning rates for PV experience curves, areal mass density, and time to produce materials for selected solar absorbers.

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All of the authors are employees at Oxford Photovoltaics, Ltd.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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