ACS Energy Letters

http://pubs.acs.org/journal/aelccp

VIEWPOINT

Perovskite Photovoltaics: The Path to a Printable Terawatt-Scale Technology

Abundant, low-cost, reliable, and clean energy is critical not just to maintain but also to improve the living conditions across the globe. Because of the world's unrelenting population and GDP growth that is only slightly offset by gains in energy intensity per $ of GDP, it has been estimated that by 2050 the world will consume an average of 30 Terawatts (TW) of *total* (i.e., not just electricity) energy, of which, assuming no current generation capacity retires, at least between 10−15 TW will be completely new capacity.[1] Addressing this "Terawatt challenge", a term originally coined by Richard Smalley[2] in 2004, in a nonpolluting and sustainable way is integral to addressing socioeconomic needs in both industrial and rapidly developing countries worldwide. In conjunction with ongoing electrification of the energy system, the vastness of the available solar resource will provide a solution to the world's energy needs if we can develop sufficiently low-cost, high-performance, and massively scalable photovoltaic (PV) technology.

PVs based on halide perovskite materials appear to have attributes to assist in the process of large-scale electrification, thus offering a pathway to develop a truly transformational technology at the TW scale. Uniquely, halide perovskite solar cells (HPSCs) have the highest efficiencies when they are solution-processed; therefore, one can envision solar cells printed in a similar manner and scale as newspapers. Also, the optoelectronic tunability of perovskites can enable multi-junction devices—perovskite/perovskite as well as perovskite/silicon or perovskite/thin-film absorber (e.g., CIGS)—that yield very high efficiencies. HPSCs are therefore suited to helping address the challenge of large-scale, PV-based electricity production that can power the future world.

In this Viewpoint, we discuss how and why HPSCs can provide a path to TW-scale PVs because of potentially extremely low cost cell technology coupled with an inherently scalable production technology. Realizing low-cost, high-performance HPSC technology at the TW scale will shift research needs from the device level to other critical elements required to implement solar at these scales—elements such as distribution along with associated power systems, electrochemical storage, and electrification of industry. The opportunity for HPSCs to positively change the dynamics of the solar market justifies investing the resources for their development.

Printable, Terawatt PV Technology. The PV market is currently dominated by silicon (Si)-based devices. In part, this is a result of the massive investments in Si-based research and development that created a foundational understanding of this semiconducting material that is unrivaled. Despite the basic materials physics limitations of Si (e.g., indirect bandgap and modest absorption cross sections), this scientific platform has allowed PV devices based on this Earth-abundant material to approach their practical performance limit.[3,4] The solar industry, dominated by Si technology, has exhibited massive success with average growth of ∼40% each year for the last 10 years. However, as much of the world looks to add even more production capacity, sustaining even a 20% level of growth will require innovative low-cost technologies to allow PVs to be coupled with a flexible grid at a combined cost that is lower than conventional power. As the industry has demonstrated, continued innovation is happening within the silicon industry that will continue to reduce both product and CapEx costs. However, the strong need for low-CapEx, highly scalable, and low-cost overall technologies makes perovskite-based technologies a prime candidate to enhance Si and existing thin-film technologies toward TW-scale production and to provide an additional scalable PV technology in its own right to help enable the TW PV revolution.

PV technologies can offer a streamlined path toward TW-scale production and deployment if several basic constraints are met. First, the materials in the device stack must be sufficiently abundant in the Earth's crust to not fundamentally limit the amount of PVs that can be produced. Second, the technology must be stable, scalable, and have a sufficiently high efficiency. In contrast to existing thin-film PV technologies, HPSCs, with their remarkable performance, have propelled perovskite-based PVs to efficiency parity on a time scale an order of magnitude faster, making them an exciting new candidate to develop for TW-scale deployment.

Two main methods for producing HPSCs are vapor-phase deposition and solution processing, which are both inherently scalable technologies. Solution processing, in particular, enables unique opportunities for large-scale production at relatively low CapEx. Considering solution processing and sheet-to-sheet (S2S) production approaches, technoeconomic analyses indicate that HPSCs can compete with other PV technologies.[5] Furthermore, roll-to-roll (R2R) approaches to fabricate the entire HPSC module could further accelerate the rate of production. The opportunity to rapidly scale PVs manufactured using a R2R printing process is readily illustrated by the following calculation: Imagine we are able to develop a R2R-printed PV technology with speed comparable to news print based on the daily circulation of news print in China, India, Japan, United States, and Germany—about 300 million copies per day and ∼30 m^2 per copy. We would then have a total production capacity of $\sim 3 \times 10^{12}$ m^2 per year. If this equipment were used to print modules with efficiencies of 15% or 150 W/m^2, this would enable printing of modules at a rate close to 500 TW per year! This is a factor of 5000 greater than the current manufacturing capacity for PV modules!

For vapor deposition, the second main method of producing HPSCs, we can make a similar comparison—with similar

Received: October 3, 2017
Accepted: October 9, 2017

ACS Publications © XXXX American Chemical Society

DOI: 10.1021/acsenergylett.7b00964
ACS Energy Lett. XXXX, XXX, XXX−XXX

Pursuant to the DOE Public Access Plan, this document represents the authors' peer-reviewed, accepted manuscript. The published version of the article is available from the relevant publisher.

conclusions—to producing potato chip bags, where vapor deposition is used to produce the metal layers that keep potato chips fresh. These analyses, of course, presuppose a component supply chain and module processing that can sustain this production level, but it impressively conveys the opportunity for disruptive R2R processing technologies.

HPSCs clearly represent a major opportunity for a new technology that can drive increased competition in the global PV market and because of this increased diversity of technologies and radically different production methodology might drive a new maturation rate for the entire industry. Increased technology diversity is important for the success of solar in the market, and increased consumer choice is highly desirable. It also appears to be a technology that could open new markets because of the ease with which it could be integrated into thin flexible packages and could have a significant domestic manufacturing base because of the presence of a complete supply chain and the coupling to application into products that are manufactured locally.

Solid demonstrations of high-efficiency, long-term operational stability as well as scalability at the cell and module levels are critically required to move perovskite PVs from laboratory-scale devices to industry-relevant PV technology if we are to even begin considering the extreme scaling envisioned above. During the past several years, significant improvements have been demonstrated on achieving >20%-efficient devices with much improved stability of the perovskite composition and device stack.[10−13] During the last year, we have also seen rapid progress on module development faster than any technology in history.[9,14−16] Figure 1 compares perovskite PVs to single-crystalline Si and polycrystalline CdTe PV technologies based on device and module (or mini-module) levels from the past 20 years.[6−9] It is worth noting that PV efficiency normally decreases with increasing device area for all PV technologies. However, this would not change the development trend revealed from Figure 1. In general, the module development follows behind developing laboratory-scale devices with a performance gap gradually decreasing to around 3−4%.

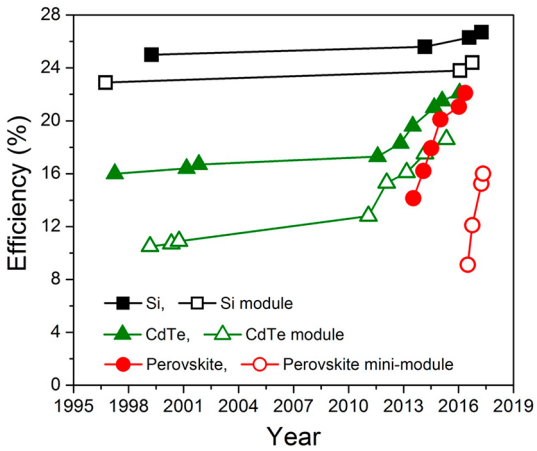

Figure 1. Comparison of perovskite PVs to single-crystalline Si and polycrystalline CdTe PV technologies based on device and module (or mini-module) results from the past 20 years.[6−9]

The progress reported for perovskite PVs is advancing more rapidly than all other PV technologies. Laboratory-scale perovskite devices should soon achieve efficiencies of 24−25% via continuous advances on perovskite composition, device architecture, optical management, and other aspects already applied to other thin-film technologies but which have only been explored cursorily for HPSCs.[17−19] Thus, it is reasonable to expect the demonstration of >20%-efficient perovskite PV modules in a few years. Critical to the ability to realize HPSCs at the TW scale is the ability to sustain the rate of production across various manufacturing inputs (e.g., glass, refined raw materials). Advances in stability, scalability, and efficiency of devices are the foundation of applied PV research. However, clear needs exist to develop advanced manufacturing approaches that can enable realizing these levels of production capacity for full PV modules and the associated materials and component inputs.

Finally, we note that permitting, financing, and other soft costs are often rightly identified as critical cost bottlenecks for PV deployment. The existence of these and other bottlenecks to PVs not discussed here is also critical, but no high penetration PV scenario is possible without lowered cost.

Low-Cost, Ultrahigh-Efficiency Tandem Devices. As the record efficiencies of single-junction PV technologies steadily approach their theoretical efficiencies limited by thermodynamic principles, i.e., the Shockley−Queisser (SQ) radiative efficiency limit,[20] new approaches may significantly improve their efficiency levels. One proven approach to break the single-junction SQ efficiency limits is to form tandem devices by stacking two or more solar cells with different bandgaps.[21] Each subcell is optimized to absorb a different portion of the solar spectrum, reducing voltage loss caused by thermalization of photogenerated carriers. In general, the subcells can be stacked in two different ways: (1) 2*n*-terminal (2*n*-T), where *n* is the number of subcells and each subcell works independently, or (2) 2-terminal (2-T) tandem cells, where subcells are monolithically integrated and current-match among all subcells is required. In principle, the 2*n*-T tandem cell configuration has a slightly higher theoretical efficiency than the 2-T configuration because it does not require current-matching between the two subcells as in the 2-T configuration. This is particularly advantageous when the solar spectrum varies, to which the 2-T configuration is more sensitive.[22,23] Compared to the 2-T structure, the 2*n*-T configuration also allows selection from a wider range of absorbers with different bandgaps, as well as offers fabrication advantages because the subcells can be developed separately without worrying about the processing compatibility. However, the 2*n*-T tandem cells may require more materials such as substrates and contacts, which potentially can lead to higher module costs and more parasitic optical/electric losses.

Ultrahigh-efficiency tandem solar cells have been fabricated with III−V semiconductors. The 2-T five-junction solar cells based on III−V materials have demonstrated efficiencies up to 38.8%.[24] Unfortunately, these cells have rather limited use for terrestrial applications due to the high costs associated with the complex epitaxial growth and expensive substrates. Recently, tandem cells combining III−V top cells and Si bottom cells have reached ultrahigh efficiencies—32.8% for a 4-T dual-junction III−V/Si tandem cell and 35.9% for a 4-T tandem cell combining a GaInP/GaAs dual-junction cell with a Si single-junction cell.[25] Although the Si single-junction bottom solar cells are much less expensive than the III−V cells, technoeconomic analysis[25] reveals that the costs of III−V/Si tandem solar cells are an order-of-magnitude more expensive than the Si single-junction cells. Thus, to reduce the cost,

DOI: 10.1021/acsenergylett.7b00964
ACS Energy Lett. XXXX, XXX, XXX−XXX
Pursuant to the DOE Public Access Plan, this document represents the authors' peer-reviewed, accepted manuscript. The published version of the article is available from the relevant publisher.

alternative deposition and lift-off technologies for III−V have to be developed that radically lower the cost of the expensive wide-bandgap epitaxially grown III−V absorbers. Alternatively, these materials could be replaced by lower-cost polycrystalline wide-bandgap thin-film absorbers. In addition, the low-bandgap III−V absorbers could be replaced by less expensive absorbers with similar bandgaps (e.g., Si, CIGS, or perovskite).

To date, polycrystalline thin films have not become mainstream contributors to tandem cells because of two main reasons: (1) The wide-bandgap thin-film top cells all have low efficiencies due to significant open-circuit voltage (V_{oc}) deficits; and (2) the cell fabrication processes require high temperatures and chemical treatments that degrade the performance of bottom cells.

Here again, the emergence of HPSCs can remove these obstacles. First, HPSCs exhibit V_{oc} deficits close to the epitaxial single-crystalline III−V thin-film solar cells, and the V_{oc} deficit does not significantly increase as the bandgap increases. Second, HPSCs can be fabricated by low-temperature solution processing. The low processing temperature and use of organic solvents that do not attack inorganic low-bandgap solar cells such as Si and CIGS can enable tandems with these technologies. Halide perovskites exhibit excellent tunability of bandgap—from about 1.2 to >2.3 eV. Therefore, halide perovskites also provide an unprecedented opportunity for fabricating low-cost and ultrahigh-efficiency perovskite-based tandems with established thin-film technologies based on lower-bandgap materials (e.g., Si, CIGS). The ability to make tandems across technologies naturally enables perovskite/perovskite tandems and the potential for multijunctions at the TW scale.

Figure 2 summarizes the current status of state-of-the-art two-junction tandem solar cells. At present, the best 2-T tandem is based on GaInP/GaAs with an efficiency of 31.6%, and the best 4-T tandem is achieved using GaInP/Si with an efficiency of 32.8%.[25,26] These results were achieved after decades of research. With the rapid progress of HPSCs in the past several years, however, we have already seen impressive advancement of perovskite-based tandem devices. 4-T and 2-T tandem cells combining wide-bandgap HPSCs with low-bandgap Si or CIGS solar cells have been demonstrated, showing the best efficiencies of 26.4% for 4-T perovskite/Si,[27] 23.6% for 2-T perovskite/Si,[28] 22.1% for 4-T perovskite/CIGS,[29] 21% for 4-T perovskite/perovskite,[30] and 18.5% for 2-T perovskite/perovskite tandem configurations.[31]

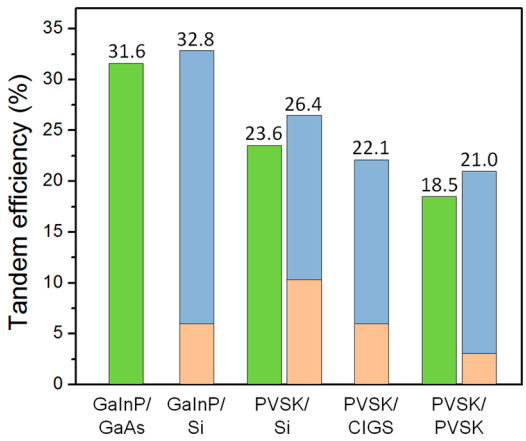

Figure 2. Comparison of the current status of state-of-the-art two-junction tandem solar cells including 2-T (green) and 4-T (blue: top cell; brown: bottom cell) configurations.[25−31]

The development of HPSCs is much less mature than III−V cells; therefore, we expect significant advances in the coming years for perovskite/X (where X = [Si, CIGS, or perovskites]) tandem solar cells, with the potential of >30% efficiency. To reach this target, a key effort is to further improve the performance of wide-bandgap (∼1.7−1.9 eV) HPSCs to an efficiency level of about 20% or higher. This advance would lead to about 30 and 33% tandem efficiency when paired with 20 and 25% Si bottom cells, respectively, based on a recent spectral efficiency analysis.[29] Reaching this target requires a continuous increase of V_{oc} via reduction of recombination in the bulk and at the interfaces, as well as the advances in contact/transport materials with optimized energetic levels and transport properties.

All-perovskite-based tandems require low-bandgap (∼1.2−1.3 eV) HPSCs with high response in the near-infrared wavelength range (∼700−1000 nm) and low processing temperature. For monolithic integration, the interconnection layer/structure needs to have good optical and electronic coupling, but it should also be effective at protecting the lower subcells during processing. The deployment of perovskite/X multijunctions at the TW scale will depend on some of the constraints for the bottom-cell technologies. Here again, perovskite-based multijunctions appear to have advantages that could make them transformational, even in the context of existing technologies.

Life Cycle and Environmental Considerations. At TW-scale production, we must consider the life cycle for modules in a more integral fashion. The cost of recycling, refurbishing, or disposing will grow with increasing market size, and basic research is needed on how to address these costs—both by simply prolonging module lifetime and by fundamentally addressing recyclability. At the TW scale, any technology faces challenges related to module disposal, costs associated with material reclamation and reuse in production, as well as environmental impacts (e.g., CO_2 production).[32] Existing cost models for HPSCs as well as other PV technologies typically do not factor in the impacts of the product life cycle and associated economic cost nor do they provide a comparison across technologies at the TW scale. This is a major shortcoming of such analyses because the actual cost of electricity is directly controlled by the life cycle of the modules, among other factors.

Existing HPSC cost models typically indicate that the primary cost of HPSC modules, if a reasonable lifetime is assumed, is dominated by the back- and front-plane glass and electrodes. The low formation energy and solution processability of the active and transport materials could enable a much more efficient and cost-effective approach to refurbishing or recycling HPSC modules. Indeed, the reuse of substrates, along with reclamation of electrode and active layer materials, has been demonstrated for laboratory-scale HPSCs.[33−35]

Many have viewed the reliance of HPSCs on lead (Pb) in the active layer to be a significant concern for deploying at the TW scale because of possible environmental contamination and associated human and animal health impacts. Objectively, this concern does not appear to be significant because the amount of Pb in the active layer is minimal due to the strong absorption coefficient of the perovskite; this results in panels that would contain less Pb per unit area than presently found in an equivalent-area Si-based module.[36] Analysis performed thus far indicates that HPSCs deployed at the TW scale will not present

DOI: 10.1021/acsenergylett.7b00964
ACS Energy Lett. XXXX, XXX, XXX−XXX
Pursuant to the DOE Public Access Plan, this document represents the authors' peer-reviewed, accepted manuscript. The published version of the article is available from the relevant publisher.

a serious hazard of Pb contamination into the environment. With the thin-film absorber being ~200−500 nm thick and the relatively low density of typical HPSC absorber layers, the amount of Pb in 1 t of coal represents enough Pb to produce more than 400 m^2 of HPSC absorber. The 2015 U.S. domestic coal consumption of 900 million short tons[37] can be estimated to have released enough finely dispersed Pb into the environment to produce on the order of 50 TW of nameplate capacity HPSC. Nevertheless, one cannot discount the impact of public concern about Pb, and significant public education about these issues must occur if HPSCs based on Pb compounds are to be widely deployed.

Of considerably larger and more realistic concern are the solvents currently used in producing HPSCs that are often highly toxic or have major environmental impacts.[38,39] Development of greener solution chemistry will likely be required to enable production of HPSCs at the TW scale or, at a minimum, development of highly efficient solvent-recapture processes for the current generation of solvent chemistries. Indeed, failure to make advances on this front would undercut the solution-processing advantage of HPSC technologies. In this case, vapor-phase-based methodologies may still be able to have impact at the TW scale. However, a path to the massive TW-scale production could become more difficult as a result of the increased CapEx associated with vacuum processing. The future path will depend on what other gains can be made in efficiency and device lifetime by changing these processing details.

To conclude, while explosive growth in research on halide perovskite PVs has been made possible by the low barrier to entry for researchers to enter the field commensurate with its low-CapEx potential and the rapid efficiency enhancements that have been made, the promise of the technology is predicated on the exciting possibility of a TW-scale technology that could address the future societal needs for energy. The potential that has driven the research community and the associated financial investment is a testament to the potential of these materials to be transformational in meeting the world's future energy needs. On the basis of their physical properties, meteoric rise in efficiencies, ease of processing, and abundances, HPSCs offer a path to addressing TW-scale energy production. In addition, these materials can impact other parts of the energy sector such as solid-state lighting via improved efficiencies. For energy generation via ubiquitous installed HPSCs, we must tackle critical research needs associated with stability as well as make continued advances in efficiencies and in scalability of both cells and modules. The rapid advances in scientific understanding, coupled with ongoing technology development efforts across the globe, make HPSC and tandem cells based on hybrid perovskites a very real pathway to energy at the TW scale.

Joseph J. Berry[*,†]
Jao van de Lagemaat[†]
Mowafak M. Al-Jassim[†]
Sarah Kurtz[†]
Yanfa Yan[‡]
Kai Zhu[*,†]

[†]National Renewable Energy Laboratory, Golden, Colorado 80401, United States
[‡]University of Toledo, Toledo, Ohio 43606, United States

AUTHOR INFORMATION

Corresponding Authors
*E-mail: Joe.Berry@nrel.gov (J.J.B).
*E-mail: Kai.Zhu@nrel.gov (K.Z.).

ORCID
Yanfa Yan: 0000-0003-3977-5789
Kai Zhu: 0000-0003-0908-3909

Notes

Views expressed in this Viewpoint are those of the authors and not necessarily the views of the ACS.
The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy under Contract No. DE-AC36-08GO28308 to the National Renewable Energy Laboratory. We would like to thank Lenny Tinker and Dana Olson of the DOE/EERE/SETO team for helpful discussions. We also acknowledge Zhen Li for contributions in preparing figures and references.

REFERENCES

(1) Hoffert, M. I.; Caldeira, K.; Jain, A. K.; Haites, E. F.; Harvey, L. D. D.; Potter, S. D.; Schlesinger, M. E.; Schneider, S. H.; Watts, R. G.; Wigley, T. M. L.; et al. Energy Implications of Future Stabilization of Atmospheric CO_2 Content. *Nature* **1998**, *395*, 881−884.

(2) Smalley, R. E. Future Global Energy Prosperity: The Terawatt Challenge. *MRS Bull.* **2005**, *30*, 412−417.

(3) Taguchi, M.; Yano, A.; Tohoda, S.; Matsuyama, K.; Nakamura, Y.; Nishiwaki, T.; Fujita, K.; Maruyama, E. 24.7% Record Efficiency HIT Solar Cell on Thin Silicon Wafer. *IEEE J. Photovoltaics* **2014**, *4*, 96−99.

(4) Yoshikawa, K.; Kawasaki, H.; Yoshida, W.; Irie, T.; Konishi, K.; Nakano, K.; Uto, T.; Adachi, D.; Kanematsu, M.; Uzu, H.; et al. Silicon Heterojunction Solar Cell with Interdigitated Back Contacts for a Photoconversion Efficiency over 26%. *Nature Energy* **2017**, *2*, 17032.

(5) Song, Z.; McElvany, C. L.; Phillips, A. B.; Celik, I.; Krantz, P. W.; Watthage, S. C.; Liyanage, G. K.; Apul, D.; Heben, M. J. A Technoeconomic Analysis of Perovskite Solar Module Manufacturing with Low-Cost Materials and Techniques. *Energy Environ. Sci.* **2017**, *10*, 1297−1305.

(6) Green, M. A.; Hishikawa, Y.; Warta, W.; Dunlop, E. D.; Levi, D. H.; Hohl-Ebinger, J.; Ho-Baillie, A. W. H. Solar Cell Efficiency Tables (Version 50). *Prog. Photovoltaics* **2017**, *25*, 668−676.

(7) NREL Solar Cell Efficiency Chart. http://www.nrel.gov/pv/assets/images/efficiency_chart.jpg (accessed October 3, 2017).

(8) Priyadarshi, A.; Haur, L. J.; Murray, P.; Fu, D.; Kulkarni, S.; Xing, G.; Sum, T. C.; Mathews, N.; Mhaisalkar, S. G. A Large Area (70 cm^2) Monolithic Perovskite Solar Module with a High Efficiency and Stability. *Energy Environ. Sci.* **2016**, *9*, 3687−3692.

(9) Chen, H.; Ye, F.; Tang, W.; He, J.; Yin, M.; Wang, Y.; Xie, F.; Bi, E.; Yang, X.; Grätzel, M.; et al. A Solvent- and Vacuum-Free Route to Large-Area Perovskite Films for Efficient Solar Modules. *Nature* **2017**, DOI: 10.1038/nature23877.

(10) Saliba, M.; Matsui, T.; Domanski, K.; Seo, J.-Y.; Ummadisingu, A.; Zakeeruddin, S. M.; Correa-Baena, J.-P.; Tress, W. R.; Abate, A.; Hagfeldt, A.; et al. Incorporation of Rubidium Cations into Perovskite Solar Cells Improves Photovoltaic Performance. *Science* **2016**, *354*, 206−209.

(11) Tsai, H.; Nie, W.; Blancon, J.-C.; Stoumpos, C. C.; Asadpour, R.; Harutyunyan, B.; Neukirch, A. J.; Verduzco, R.; Crochet, J. J.; Tretiak, S.; et al. High-Efficiency Two-Dimensional Ruddlesden−Popper Perovskite Solar Cells. *Nature* **2016**, *536*, 312−316.

(12) Tan, H.; Jain, A.; Voznyy, O.; Lan, X.; Garcia de Arquer, F. P.; Fan, J. Z.; Quintero-Bermudez, R.; Yuan, M.; Zhang, B.; Zhao, Y.; et al. Efficient and Stable Solution-Processed Planar Perovskite Solar Cells via Contact Passivation. *Science* **2017**, *355*, 722−726.

DOI: 10.1021/acsenergylett.7b00964
ACS Energy Lett. XXXX, XXX, XXX−XXX

Pursuant to the DOE Public Access Plan, this document represents the authors' peer-reviewed, accepted manuscript. The published version of the article is available from the relevant publisher.

(13) Shin, S. S.; Yeom, E. J.; Yang, W. S.; Hur, S.; Kim, M. G.; Im, J.; Seo, J.; Noh, J. H.; Seok, S. I. Colloidally Prepared La-doped $BaSnO_3$ Electrodes for Efficient, Photostable Perovskite Solar Cells. *Science* **2017**, *356*, 167−171.

(14) Yang, M.; Li, Z.; Reese, M. O.; Reid, O. G.; Kim, D. H.; Siol, S.; Klein, T. R.; Yan, Y.; Berry, J. J.; van Hest, M. F. A. M.; et al. Perovskite Ink with Wide Processing Window for Scalable High-Efficiency Solar Cells. *Nature Energy* **2017**, *2*, 17038.

(15) Ono, L. K.; Park, N.-G.; Zhu, K.; Huang, W.; Qi, Y. Perovskite Solar Cells - Towards Commercialization. *ACS Energy Lett.* **2017**, *2*, 1749−1751.

(16) Agresti, A.; Pescetelli, S.; Palma, A. L.; Del Rio Castillo, A. E.; Konios, D.; Kakavelakis, G.; Razza, S.; Cinà, L.; Kymakis, E.; Bonaccorso, F.; et al. Graphene Interface Engineering for Perovskite Solar Modules: 12.6% Power Conversion Efficiency over 50 cm^2 Active Area. *ACS Energy Lett.* **2017**, *2*, 279−287.

(17) Saliba, M.; Zhang, W.; Burlakov, V. M.; Stranks, S. D.; Sun, Y.; Ball, J. M.; Johnston, M. B.; Goriely, A.; Wiesner, U.; Snaith, H. J. Plasmonic-Induced Photon Recycling in Metal Halide Perovskite Solar Cells. *Adv. Funct. Mater.* **2015**, *25*, 5038−5046.

(18) Pazos-Outón, L. M.; Szumilo, M.; Lamboll, R.; Richter, J. M.; Crespo-Quesada, M.; Abdi-Jalebi, M.; Beeson, H. J.; Vrućinić, M.; Alsari, M.; Snaith, H. J.; et al. Photon Recycling in Lead Iodide Perovskite Solar Cells. *Science* **2016**, *351*, 1430−1433.

(19) Fang, Y.; Wei, H.; Dong, Q.; Huang, J. Quantification of Re-Absorption and Re-Emission Processes to Determine Photon Recycling Efficiency in Perovskite Single Crystals. *Nat. Commun.* **2017**, *8*, 14417.

(20) Shockley, W.; Queisser, H. J. Detailed Balance Limit of Efficiency of P-N Junction Solar Cells. *J. Appl. Phys.* **1961**, *32*, 510−519.

(21) Vos, A. D. Detailed Balance Limit of The Efficiency of Tandem Solar Cells. *J. Phys. D: Appl. Phys.* **1980**, *13*, 839.

(22) Almansouri, I.; Ho-Baillie, A.; Bremner, S. P.; Green, M. A. Supercharging Silicon Solar Cell Performance By Means of Multi-junction Concept. *IEEE J. Photovoltaics* **2015**, *5*, 968−976.

(23) Yu, Z.; Leilaeioun, M.; Holman, Z. Selecting Tandem Partners for Silicon Solar Cells. *Nature Energy* **2016**, *1*, 16137.

(24) Chiu, P. T.; Law, D. C.; Woo, R. L.; Singer, S. B.; Bhusari, D.; Hong, W. D.; Zakaria, A.; Boisvert, J.; Mesropian, S.; King, R. R.; et al. 35.8% Space and 38.8% Terrestrial 5J Direct Bonded Cells. In *2014 IEEE 40th Photovolt. Spec. Conf.*; IEEE: Denver, CO, 2014; pp 11−13.

(25) Essig, S.; Allebé, C.; Remo, T.; Geisz, J. F.; Steiner, M. A.; Horowitz, K.; Barraud, L.; Ward, J. S.; Schnabel, M.; Descoeudres, A.; et al. Raising the One-Sun Conversion Efficiency of III−V/Si Solar Cells to 32.8% for Two Junctions and 35.9% for Three Junctions. *Nature Energy* **2017**, *2*, 17144.

(26) Kayes, B. M.; Zhang, L.; Twist, R.; Ding, I.-K.; Higashi, G. S. Flexible Thin-Film Tandem Solar Cells with > 30% Efficiency. *IEEE J. Photovoltaics* **2014**, *4*, 729−733.

(27) Duong, T.; Wu, Y.; Shen, H.; Peng, J.; Fu, X.; Jacobs, D.; Wang, E.-C.; Kho, T. C.; Fong, K. C.; Stocks, M.; et al. Rubidium Multication Perovskite with Optimized Bandgap for Perovskite-Silicon Tandem with Over 26% Efficiency. *Adv. Energy Mater.* **2017**, *7*, 1700228.

(28) Bush, K. A.; Palmstrom, A. F.; Yu, Z. J.; Boccard, M.; Cheacharoen, R.; Mailoa, J. P.; McMeekin, D. P.; Hoye, R. L. Z.; Bailie, C. D.; Leijtens, T.; et al. 23.6%-Efficient Monolithic Perovskite/Silicon Tandem Solar Cells with Improved Stability. *Nature Energy* **2017**, *2*, 17009.

(29) Fu, F.; Feurer, T.; Weiss; Thomas, P.; Pisoni, S.; Avancini, E.; Andres, C.; Buecheler, S.; Tiwari; Ayodhya, N. High-Efficiency Inverted Semi-Transparent Planar Perovskite Solar Cells in Substrate Configuration. *Nature Energy* **2016**, *2*, 16190.

(30) Zhao, D.; Yu, Y.; Wang, C.; Liao, W.; Shrestha, N.; Grice, C. R.; Cimaroli, A. J.; Guan, L.; Ellingson, R. J.; Zhu, K.; et al. Low-Bandgap Mixed Tin−Lead Iodide Perovskite Absorbers With Long Carrier Lifetimes for All-Perovskite Tandem Solar Cells. *Nature Energy* **2017**, *2*, 17018.

(31) Rajagopal, A.; Yang, Z.; Jo, S. B.; Braly, I. L.; Liang, P.-W.; Hillhouse, H. W.; Jen, A. K. Y. Highly Efficient Perovskite−Perovskite Tandem Solar Cells Reaching 80% of The Theoretical Limit in Photovoltage. *Adv. Mater.* **2017**, *29*, 1702140.

(32) Varun; Bhat, I. K.; Prakash, R. LCA of Renewable Energy For Electricity Generation Systems - a Review. *Renewable Sustainable Energy Rev.* **2009**, *13*, 1067−1073.

(33) Kim, B. J.; Kim, D. H.; Kwon, S. L.; Park, S. Y.; Li, Z.; Zhu, K.; Jung, H. S. Selective Dissolution of Halide Perovskites as a Step Towards Recycling Solar Cells. *Nat. Commun.* **2016**, *7*, 11735.

(34) Binek, A.; Petrus, M. L.; Huber, N.; Bristow, H.; Hu, Y.; Bein, T.; Docampo, P. Recycling Perovskite Solar Cells to Avoid Lead Waste. *ACS Appl. Mater. Interfaces* **2016**, *8*, 12881−12886.

(35) Kadro, J. M.; Pellet, N.; Giordano, F.; Ulianov, A.; Muntener, O.; Maier, J.; Grätzel, M.; Hagfeldt, A. Proof-of-Concept for Facile Perovskite Solar Cell Recycling. *Energy Environ. Sci.* **2016**, *9*, 3172−3179.

(36) Park, N.-G.; Grätzel, M.; Miyasaka, T.; Zhu, K.; Emery, K. Towards stable and commercially available perovskite solar cells. *Nature Energy* **2016**, *1*, 16152.

(37) Arena, J. A.; Park, B. *Annual Coal Report 2015*. US Energy Information Administration. https://www.eia.gov/coal/annual/pdf/acr.pdf (2015).

(38) Gardner, K. L.; Tait, J. G.; Merckx, T.; Qiu, W.; Paetzold, U. W.; Kootstra, L.; Jaysankar, M.; Gehlhaar, R.; Cheyns, D.; Heremans, P.; et al. Nonhazardous Solvent Systems for Processing Perovskite Photovoltaics. *Adv. Energy Mater.* **2016**, *6*, 1600386.

(39) Noel, N. K.; Habisreutinger, S. N.; Wenger, B.; Klug, M. T.; Horantner, M. T.; Johnston, M. B.; Nicholas, R. J.; Moore, D. T.; Snaith, H. J. A Low Viscosity, Low Boiling Point, Clean Solvent System for The Rapid Crystallisation of Highly Specular Perovskite Films. *Energy Environ. Sci.* **2017**, *10*, 145−152.

DOI: 10.1021/acsenergylett.7b00964
ACS Energy Lett. XXXX, XXX, XXX−XXX

Pursuant to the DOE Public Access Plan, this document represents the authors' peer-reviewed, accepted manuscript. The published version of the article is available from the relevant publisher.