

Modeling low cost hybrid tandem photovoltaics with the potential for efficiencies exceeding 20%[†]

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It is estimated that for photovoltaics to reach grid parity around the planet, they must be made with costs under \$0.50 per W_p and must also achieve power conversion efficiencies above 20% in order to keep installation costs down. In this work we explore a novel solar cell architecture, a hybrid tandem photovoltaic (HTPV), and show that it is capable of meeting these targets. HTPV is composed of an inexpensive and low temperature processed solar cell, such as an organic or dye-sensitized solar cell, that can be printed on top of one of a variety of more traditional inorganic solar cells. Our modeling shows that an organic solar cell may be added on top of a commercial CIGS cell to improve its efficiency from 15.1% to 21.4%, thereby reducing the cost of the modules by $\sim 15\%$ to 20% and the cost of installation by up to 30%. This suggests that HTPV is a promising option for producing solar power that matches the cost of existing grid energy.

Introduction

Photovoltaics are already cost competitive with grid energy in places around planet where there is a lot of sunlight or where the price of electricity is relatively high, such as California and Germany.¹ In order for photovoltaics to compete widespread with power from fossil fuels, it is estimated that their cost must drop from $\sim \$1$ – $\$1.30$ per W_p today to under \$0.50 per W_p .² However, this reduction in module price is not itself sufficient for photovoltaics to match the cost of energy from fossil fuels; approximately 50% of the cost of harvesting solar energy comes from installing and maintaining the solar panels,³ and these costs must come down as well. While many researchers are pursuing technologies that have much lower installation costs, such as solar tarps⁴ and building integrated photovoltaics (BIPV),⁵ the most significant reductions in the installation costs of utility scale photovoltaics will come from increasing the power conversion efficiencies (PCEs) of the solar cells. Photovoltaic technologies that are more efficient have lower installation costs because they require fewer panels to produce a given amount of power, and much of the installation cost scales with the number (or area) of installed panels. Thus it has been estimated⁶ that photovoltaics will need to achieve PCEs above 20% if they are to have installation costs low enough to compete on the utility

scale with fossil fuels.[‡] Although solar cells exist that have passed this efficiency threshold⁸ none is definitely capable of meeting this high standard at such a low cost. Among the dominant photovoltaic technologies, only silicon and CIGS have produced laboratory-scale record cells with efficiencies exceeding 20% (Table 1), and the efficiencies of typical production modules are well below these records, in the range of 11–16%. It is possible to purchase high efficiency modules with lower installation costs, such as those made by SunPower,⁹ which have average efficiencies above 20%, but these modules are more expensive. Although module costs (Table 1) are dropping rapidly, the cost of even the cheapest modules is still far from the \$0.50 per W_p target, and it is estimated that for silicon cells (which currently account for over 85% of photovoltaics production) to reach it while maintaining high efficiency will require advanced designs not currently incorporated in industry roadmaps.²

One way to achieve very high efficiencies is to make a multi-junction (tandem) solar cell, in which multiple absorbing layers are used to divide the solar spectrum into parts. This allows

[‡] The measure of cost that most accurately captures both module and installation costs is the levelized cost of energy (LCOE), which is the sum of all the costs associated with a photovoltaic system divided by its total energy production over the system lifetime (*e.g.* \$ per kW·h). Many of the factors that make up LCOE are location dependent and difficult to estimate, and because of this a photovoltaic technology does not have a specific LCOE, but rather a range of possible LCOEs.⁷ A general estimate is that photovoltaics must have a total cost below \$1.00 per W_p (\$0.50 per W_p module cost and \$0.50 per W_p installation costs) and a PCE above 20% for their LCOE to match that of grid energy ($\sim \$0.05$ kW·h).⁶ For future technologies with much lower installation costs (*e.g.* BIPV), the efficiency target will be lower.

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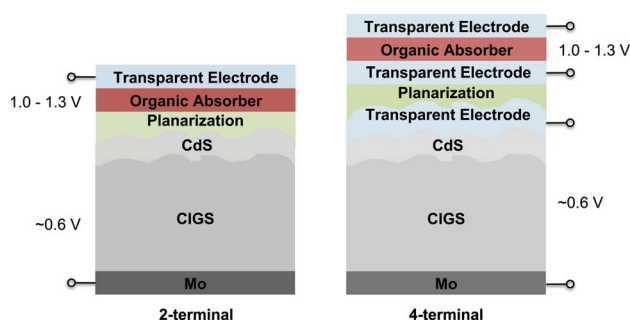
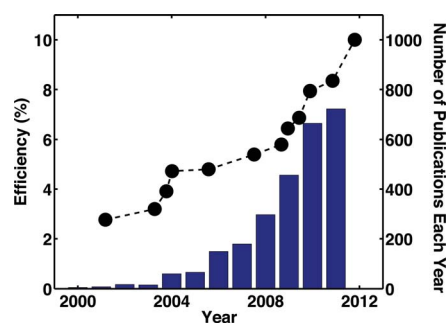
[†] Electronic supplementary information (ESI) available: Experimental methods, estimate of HTPV costs, and supplementary figures. See DOI: 10.1039/c2ee23073a

Table 1 A survey of photovoltaic technologies and costs

Technology	Laboratory record efficiency ⁴⁰	Production module efficiency	Module manufacturing cost (per W_p)
Si	25.0%	14.5% (ref. 2)	\$1.29 (ref. 2)
CIGS	20.3%	11.5% (ref. 55)	\$1.13 (ref. 3)
CdTe	17.3%	11.7% (ref. 56)	\$0.74 (ref. 56)
Dye cell	11.4%		
Organics	10.0%		
Multijunction (III–V)	43.5%		

tandem cells to reach efficiencies far beyond those achievable by single junction technologies, with the current record efficiency at 43.5%.⁸ Traditionally, tandem cells are made from very high quality III–V semiconductors, which are deposited using slow epitaxial growth and require sunlight concentrators in order to offset their enormous costs ($> \$40\,000\text{ m}^{-2}$).¹⁰ We believe there is a great opportunity for a low cost tandem device: one that combines the high efficiencies of multijunction cells with the lower cost, higher throughput and relative defect tolerance of thin film technologies. Here we argue for the potential of a hybrid tandem photovoltaic (HTPV), in which the top cell is an emerging photovoltaic technology that can be deposited at low temperatures and with rapid throughput, such as an organic or dye-sensitized cell, and the bottom cell is one of a variety of traditional inorganic photovoltaics (Fig. 1). The advantage of HTPV over traditional tandem cells is that the top cell may be printed on existing inorganic cells at or near room temperature with little additional cost; the cost of the organic layers in a bulk heterojunction solar cell, for example, is estimated^{11,12} to be as low as $\$10\text{ m}^{-2}$. Low temperature processability is critical because most inorganic photovoltaic technologies have a highly optimized thermal processing flow, and adding a top cell at high temperatures can damage the layers already there. Depositing the top cell layers near room temperature makes HTPV highly versatile in that it can be applied to nearly any inorganic cell to convert it to a tandem device. In this work, we model the efficiencies of HTPV devices where the top cell is an organic bulk heterojunction solar cell,^{13–15} though dye-sensitized,^{16–18} inorganic nanocrystal^{19,20} and other low temperature processed photovoltaic technologies are also well suited for this purpose.

The field of organic photovoltaics has advanced rapidly (Fig. 2), and it is only recently that organic solar cells have

**Fig. 1** Schematics of current matched (2-terminal) and independently operated (4-terminal) hybrid tandem photovoltaic devices with CIGS bottom cells.**Fig. 2** Record power conversion efficiencies (points) and number of publications in the literature (bars) for organic bulk heterojunction solar cells. Record efficiencies were taken from the National Renewable Energy Laboratory chart of Best Research-Cell Efficiencies.⁴⁰ The number of publications in the literature were taken from a Web of Knowledge search by topic for “bulk heterojunction” and “solar”.

improved to the point that they may be used to make effective HTPV devices. The record efficiency for an organic solar cell is now over 10%,⁸ and wide band gap organics that will be used in HTPV can achieve voltages close to 1 V with charge collection efficiencies exceeding 90%.¹⁵ Our modeling shows that today’s organic photovoltaic technology is capable of significantly improving the efficiencies, and thus lowering the costs, of some of the lowest cost commercial photovoltaic technologies (silicon and CIGS). This is in part due to the benefits of a 4-terminal device architecture, which allows an organic top cell to add power to an inorganic bottom cell, even if it is not an equal partner in absorbing the solar spectrum. Furthermore we show that as the continued development of organic technology brings cell voltages toward 1.2 V, organic absorbers toward opaque thicknesses, and module lifetimes toward 25 years, HTPV has the potential to boost the performance of commercial inorganic photovoltaics beyond the 20% threshold while reducing costs toward the goal of $\$0.50$ per W_p .

Methods

In this work we have explored the efficiency potential for HTPV devices by modeling organic top cells in hybrid tandems with silicon or CIGS bottom cells. We have chosen these inorganic cells, because they are mature technologies with band gaps that are smaller than is ideal for a single junction solar cell (1.4 eV), but close to the 1.1 eV band gap that is ideal for a two-junction solar cell.²¹ Because of their low band gaps, both of these technologies also have relatively low open circuit voltages (~ 600 mV to 700 mV);²² they will benefit more from the additional energy provided by a high voltage organic top cell than would a higher band gap cell like CdTe, GaAs or amorphous silicon. We have assumed that the band gap of the organic top cell can be finely tuned, since the flexibility of organic chemistry has shown that organic absorbers can be made with band gaps spanning the entirety of the visible part of the solar spectrum. We modeled HTPV devices using a simple photon accounting method; we assumed light is first incident on the organic top cell, which absorbs only light above its band gap, and allows lower energy photons to pass through to be absorbed by the inorganic bottom cell. The organic top cells were modeled assuming a 90% internal

quantum efficiency^{23,24} and a 0.70 fill factor,^{14,23,25} which are typical of high performing polymer bulk heterojunction solar cells. We have assumed that the V_{oc} is 0.6 V less than the band gap of the absorber divided by the electron charge, because it is thought that it will be challenging for polymer solar cells to achieve a V_{oc} that exceeds this scaling.^{26–28} The characteristics of the inorganic bottom cells (ESI Table 1†) were obtained from the device simulator PC1D in the case of silicon bottom cells, and from the literature in the case of CIGS bottom cells (see ESI† for details). We have chosen an array of inorganic bottom cells to demonstrate that the HTPV concept is compatible with high efficiency technologies, as well as those with more moderate performance. In order to make our modeling more realistic, we have assumed that 10% of the incident light across the entire solar spectrum is lost to parasitic absorption and reflection in each transparent electrode²⁹ required for the top cell.

2-Terminal tandems

Fig. 3 shows the modeled PCE *versus* the top cell band gap for organic cells that absorb 50%, 75% and 100% of the above band gap light (after subtracting off parasitic absorption). We have modeled different top cell absorbances because many of the best performing organic solar cells today must use very thin absorber layers

(100–200 nm) in order to perform well. If the active layer is made thick enough to absorb more of the light, it often comes at the expense of the charge collection efficiency.³⁰ However, as organic photovoltaic technology advances, we expect high performing cells to approach unity absorbance.³¹ In this case Table 2 shows that in the traditional tandem configuration, which is a 2-terminal, current matched design, the top cell can improve a commercial HeliVolt CIGS cell from 15.1% to 21.4% (details in ESI†). The significance of these gains becomes apparent when considering the range of efficiencies achieved by different photovoltaic technologies (Table 1). The addition of an organic top cell can bring a commercially produced CIGS cell to efficiencies that are better than the best laboratory-scale CIGS cell ever made (PCE = 20.3%). For CIGS cells in particular, there is often a large difference in efficiency between record laboratory and commercially produced devices because of the challenges of large-area uniformity; it is very expensive to extend the perfection of a very small laboratory cell to an entire module more than 10 000 times its size. By contrast, adding an organic top cell can be very inexpensive due to the low materials costs and compatibility with high throughput processing.¹² We estimate (see ESI†) that adding an organic top cell at scale can bring the cost of commercial CIGS modules down by 15–20% to as low as \$0.47 per W_p . Furthermore, the boost in cell efficiency means that fewer panels must be installed to produce equivalent

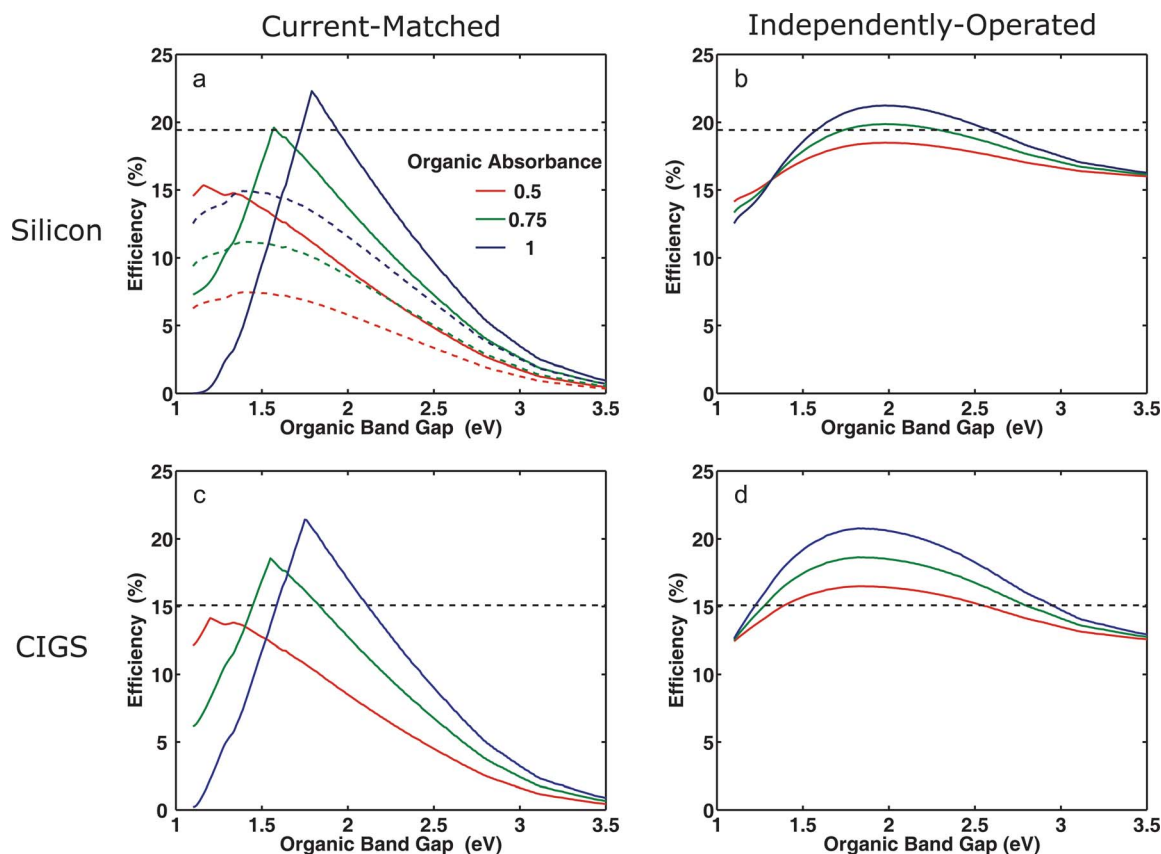


Fig. 3 Power conversion efficiencies of HTPV devices *versus* organic band gap and absorbance using (a and b) a high quality well-passivated silicon and (c and d) a HeliVolt CIGS bottom cell. Panels (a) and (c) show efficiencies assuming current matched (2-terminal) subcells, while panels (b) and (d) show efficiencies assuming independently operated (4-terminal) subcells. The horizontal black dashed lines show the efficiencies of the high quality silicon and HeliVolt CIGS bottom cells by themselves (*i.e.* with no organic top cell); the portions of the solid curves that extend above the dashed lines represent the improvements that are possible with the addition of an organic top cell. The colored dashed lines in panel (a) show the assumed efficiency of the organic top cell by itself.

Table 2 Efficiencies of HTPV devices based on different inorganic bottom cells, for 2-terminal (current matched) and 4-terminal (independently operated) architectures. The organic top cell is assumed to have the optimal band gap for each device configuration and to absorb all of the above band gap light (after subtracting parasitic absorption)

Scenario	Inorganic only PCE	<i>Current matched</i>		<i>Independently operated</i>		
		Optimal organic E_g (eV)	Tandem PCE	Optimal organic E_g (eV)	Tandem PCE	
Silicon	High quality, good passivation	19.4%	1.79	22.3%	1.98	21.3%
	High quality, no passivation	17.5%	1.79	21.8%	1.93	20.8%
	Low quality, good passivation	15.4%	1.87	19.8%	1.90	18.7%
	Low quality, no passivation	14.2%	1.88	19.8%	1.91	18.6%
CIGS	HelioVolt cell	15.1%	1.76	21.4%	1.82	20.8%
	NREL cell	19.9%	1.76	23.5%	1.98	22.7%

power, leading to a reduction in the installation cost of the panels of up to 30%. This suggests that an HTPV device composed of an organic top cell and a commercial CIGS bottom cell may be among the most promising options for producing solar power at costs that match those of existing grid energy.

Higher performing inorganic cells, those with efficiencies that are already near 20%, can also be improved by the addition of an organic top cell. As shown in Table 2, the high quality silicon cell with good passivation can be improved from 19.4% to 22.3% when an organic top cell that absorbs all of the above band gap light is added. The high efficiency CIGS cell made by thermal evaporation at NREL can be improved from 19.9% to 23.5%. This demonstrates that HTPV is a versatile technology that is capable of benefiting high and moderate efficiency bottom cells alike. Furthermore, as inorganic technologies improve and their efficiencies rise, HTPV will not become obsolete; its efficiencies will rise as well.

The success of a 2-terminal HTPV tandem relies on the organic top cell absorbing enough light to match the current of the bottom cell. This is because in a 2-terminal device, the two subcells must be connected in series, so that the lowest current of the two subcells approximately determines the current of the tandem. If the organic cell absorbs all of the above band gap light, Table 2 shows that the optimal organic band gaps for 2-terminal devices are in the range of ~ 1.7 eV to 1.9 eV, which corresponds to the range of some of today's best performing polymer solar cells,³² and is similar to the ideal band gap for the top cell in two junction inorganic–inorganic tandem devices.²¹ When the organic top cell is too thin to absorb all of the above band gap light, Fig. 3 shows that the organic band gap must be made smaller so that the top cell generates enough current to match that of the bottom cell. The amount of light absorbed in an organic solar cell varies with the specific device architecture, since it depends on the effects of optical interference. Single-junction organic cells are made with a reflective back contact so that the optical path length is at least twice the film thickness, and often effectively even larger due to optical interference.^{33–35} However, it is reasonable to expect that with today's technology, an organic top cell can be made that absorbs between 50% and 75% of the above band gap light, which is approximately the amount of light absorbed in a single pass through organic active layers with thicknesses between 100 nm and 200 nm.³⁶ This puts the efficiencies achievable with today's organic absorbances somewhere between the red and green lines of Fig. 3. In order to improve the inorganic bottom cells in a current matched configuration, one must use relatively narrow band gap organics (< 1.6 eV), which today have

lower charge collection efficiencies and are not as high performing as those with wider band gaps.^{14,32} If the organic cell absorbs only 50% of the above band gap light, then it cannot improve either the silicon or the CIGS cell in a current matched configuration. Thus HTPV tandems in the traditional, current matched configuration have tremendous potential for low cost high efficiency photovoltaics, but in the near term will have trouble improving the dominant inorganic technologies because of limitations on the thickness of organic absorbers.

4-Terminal tandems

A 2-terminal configuration is not the only way that HTPV can be implemented, and inserting extra electrodes between the top and bottom cells to make a 4-terminal device (Fig. 1) provides additional degrees of freedom. While in a 2-terminal device the subcells must be current matched, in a 4-terminal device they can be wired at the module level in either current or voltage matched configurations (including multiple cell combinations, for example, two bottom cells matching the voltage of a one top cell), or can be independently operated so that the subcell powers add without restriction. Modules based on 4-terminal devices may be fabricated monolithically, like 2-terminal cells, or they can be assembled by making the subcells on separate substrates, then laminating them together to form a mechanically stacked device.³⁷ The disadvantage of a 4-terminal design, however, is that more light in the near infrared portion of the solar spectrum is lost due to (parasitic) absorption in the electrodes of the top cell. In modeling 4-terminal tandems, we have assumed that compared to the 2-terminal devices, an additional 10% of the light is lost in the intermediate electrode before it reaches the bottom cell. Fig. 3 shows that in a 4-terminal device where there is no current matching constraint, the optimal organic band gap does not depend on how much light it absorbs. In this configuration, even if the top cell has a band gap of ~ 1.8 eV and absorbs only 75% of the above band gap light it can still improve the CIGS cells by 3.5 percentage points, bringing its PCE from 15.1% to 18.6%. This means that today's organic absorbers have the potential to make up more than half the difference in efficiency between a commercially produced CIGS cell and the best CIGS cell ever made (PCE = 20.3%). These gains are due to the flexibility of a 4-terminal device, which allows the top cell to add power to the bottom cell even if it is thin and has a wide band gap so that it is not an equal partner in absorbing the solar spectrum.

There are several reasons that the addition of an organic top cell can improve so significantly upon a variety of inorganic bottom cells. The primary benefit of adding a top cell is that high-energy photons are collected at a higher voltage than they would be if absorbed by the bottom cell. There is an additional benefit to HTPV, however, because many inorganic cells have low carrier collection efficiencies for blue photons, and absorbing high-energy photons in the top cell removes this loss. Silicon cells have low quantum efficiencies at wavelengths less than 500 nm, because high-energy photons are absorbed very close to the top surface where dangling bonds cause rapid surface recombination.²² This loss is mitigated to some extent by using quality top surface passivation, though even high performing silicon single junction cells have reduced quantum efficiencies at these wavelengths.⁹ In most CIGS cells, the quantum efficiency for high-energy photons is low because of parasitic absorption by the n-type junction partner, CdS. Most photons absorbed by CdS, which has a band gap of 2.43 eV, do not contribute to photocurrent.³⁸ These effects are illustrated in Fig. 4, which shows that the organic top cells absorb precisely where the inorganic cells have low carrier collection efficiencies, and organic solar cells typically have high carrier collection efficiencies in the ultraviolet portion of the solar spectrum.²⁴ The benefits of removing these losses from the bottom cells can be seen in Table 2. HTPV devices built upon silicon cells without top surface passivation are nearly as efficient as those built on passivated silicon cells, though without the top cell silicon cells are much worse without passivation. In this way, the organic top cell functions as a partial substitute for top surface passivation. Similarly for CIGS, the addition of an organic top cell narrows the difference between the high performing NREL cell and the Heliolt cell. In high efficiency CIGS cells the CdS layer is made as thin as possible to minimize its absorption. However, in an industrial setting the need for large area uniformity leads manufacturers to use thick CdS layers at the expense of efficiency. Placing a higher band gap organic top cell in front of these inorganic cells relaxes the need for a thin CdS layer. Thus not only does adding a top cell add power to the bottom, it also potentially relieves manufacturing constraints on the bottom cells, which may further reduce the costs of these technologies. These effects are similar to that of a luminescent down-conversion layer³⁹ added to the top surface of an inorganic cell, though the HTPV device has the additional benefit of capturing the high energy photons at a high voltage.

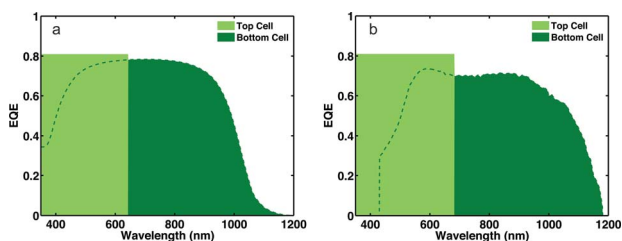


Fig. 4 External quantum efficiency (EQE) spectra for optimal HTPV devices with independently operated subcells for (a) a high quality unpassivated silicon bottom cell and (b) a Heliolt CIGS bottom cell. The plots assume the top cell has unity absorption above its band gap (after subtracting parasitic absorption). The dashed lines show the shape of the bottom cell EQE where it overlaps with the top cell absorption.

Hybrid tandems with today's organic photovoltaic technology

The above discussion represents an optimistic prediction for the PCE of HTPV devices, targets that may one day be achieved, but that require the advancement of organic photovoltaic technology. Specifically, the assumption that is not typical of today's technology is that the open circuit voltage is 0.6 V less than the organic band gap divided by the electron charge. Although this has been shown to be an empirical upper limit for V_{oc} 's in polymer/fullerene bulk heterojunction solar cells, and a 1.15 V V_{oc} has been demonstrated,^{27,40} the highest performing cells today with band gaps of ~ 1.6 to 2.0 eV can produce open circuit voltages of ~ 950 mV.^{15,41} In order to predict the efficiency of an HTPV device using current organic technology, we have modeled a device where the organic top cell has a band gap of 1.8 eV and a V_{oc} of 945 mV. We find that an organic cell absorbing 75% of the light above 1.8 eV that achieves a V_{oc} of 945 mV can improve a Heliolt CIGS cell by from 15.1% to 16.5% PCE (ESI Fig. 3†). If the top cell absorbs only 50% of the light, then the tandem cell more or less ties the efficiency of the Heliolt cell by itself. These figures suggest that even today an HTPV device with independently operated subcells is an attractive option for significantly increasing the efficiency of moderately performing inorganic photovoltaics.

The critical task in constructing high performing HTPV will be to develop wide band gap materials that achieve high V_{oc} 's while maintaining good performance with optically thick active layers. Recent work^{28,41} has shown that achieving V_{oc} 's higher than ~ 1.1 V may require moving away from the fullerene-derivative acceptors that are traditionally used to form the heterojunction with the absorbing organic. The organic top cells must also be made to absorb more light than they typically do. This can be accomplished either through light trapping schemes, or by simply making the absorber layer thicker. While many of the best performing organic photovoltaic materials optimize with active layers that are ~ 100 nm thick, some active layers can perform well with thickness up to 1 μm .³¹ The development of novel transparent electrodes, such as meshes of silver nanowires,^{42–44} will also aid the development of HTPV devices by reducing losses from parasitic absorption, particularly in 4-terminal devices. Lastly, the long-term reliability of organic photovoltaics is critical to the success of HTPV. While many inorganic technologies can last in excess of 25 years with little loss of efficiency, organic photovoltaics have historically been much shorter lived.⁴⁵ The study of the long-term degradation of organic photovoltaics is still in its infancy, though already high performing polymers with lifetimes in excess of 6 years have been demonstrated,⁴⁶ and Heliolt has reported organic small molecule based solar cells lasting the equivalent of 21 years in the sun.⁴⁷ Producing operational lifetimes at least this long in top performing organic cells will be necessary if they are to partner with inorganic cells to make commercial HTPV modules.

Conclusions

Our modeling has shown that organic solar cells have the potential to boost the efficiency of a variety of inorganic cells above 20%. In the nearer term, 4-terminal tandems allow

emerging technologies that can be processed at low temperatures to add power to more traditional inorganic cells, even if the top cells do not absorb enough of the solar spectrum to match the bottom cell's current. HTPV is a highly versatile concept that is compatible with high and moderate performing bottom cells alike; as inorganic photovoltaic technologies develop and efficiencies rise, the efficiencies of HTPV devices will rise with them. CIGS bottom cells can even be printed or sputtered onto flexible foils with great success,⁴⁸ opening up the possibility of flexible HTPV modules with PCEs over 20%. We have assumed the top cell was an organic bulk heterojunction solar cell, though the potential exists for other top cell types that are made from inexpensive materials and can be deposited near room temperature. The world record dye-sensitized solar cell produces a voltage of 0.965 V,¹⁶ and proof of concept tandem devices comprising a dye-sensitized cell on top of a CIGS cell have already been demonstrated.^{49,50}

All-organic tandems^{14,51,52} (where both the top and bottom cell are organic) offer another promising route toward low-cost photovoltaic energy, and have already reached efficiencies of 10.6%.⁴⁰ A 2008 study⁵³ estimated that they may reach efficiencies as high as 15%, and with the more current assumptions for fill factor and organic absorbance that are used in this work, this estimate rises to over 19%. However, achieving efficiencies this high with all-organic tandems requires high performing low band gap ($E_g < 1.4$ eV) organic cells, and to date little progress has been made toward devices that absorb in this range.⁵⁴

Photovoltaics are on the verge of being cost competitive with energy from fossil fuels in markets around the world. We have estimated that HTPV has the potential to decrease module costs by ~15% to 20% and installation costs by up to 30%, which would make solar power economically competitive in many more places. If the above goals for top cell voltage, absorbance and lifetime can be achieved, then HTPV may become the lowest cost photovoltaic technology that will bring us closer a planet with ubiquitous solar energy.

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