

CIGS thin-film solar cells – Breakthroughs for further efficiency improvements

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ABSTRACT

During the past two years remarkable performance improvements have been reported for polycrystalline $\text{Cu}(\text{In,Ga})\text{Se}_2$ (CIGS), CdTe and perovskite thin-film solar cells. In this paper the key breakthroughs in CIGS thin-film technology are reviewed and the scope for further performance improvements by analysing the still-remaining electrical and optical losses in record-efficiency CIGS solar cells is discussed. On the basis of this analysis it is believed that conversion efficiencies up to 25% are achievable with CIGS solar cells in the mid term. Furthermore, the potential for the concept of polycrystalline multi-junction solar cells to push efficiencies even further, towards 30%, is discussed. Finally, a short review of the CIGS market and an outlook from an industrial perspective are presented.

Introduction

Recent progress in the field of thin-film PV demonstrates the potential for the production of low-cost, highly efficient and durable solar modules in the near future. In particular, an outstanding performance of close to 22% energy conversion efficiency has been achieved at a research level by technologies based on $\text{Cu}(\text{In,Ga})\text{Se}_2$ (CIGS), exceeding the highest reported efficiency of 20.4% of the current market-dominating polycrystalline silicon (px-Si) wafer-

based technologies.

The minimum layer thickness of the materials required to efficiently absorb sunlight is 2–3 μm , which is about one hundredth of what is necessary for px-Si wafers. This carries obvious advantages in terms of the amount of material needed, and the large-area manufacturing processes enable the construction of thin-film solar modules with shorter energy payback times. Furthermore, thin-film technology allows the creation of flexible solar cells

through deposition on polymer films or metal foils. Flexibility combined with light weight and high efficiency is a key advantage in applications such as building-integrated PV (BIPV) and rooftop installations; moreover, these characteristics have a significant impact on reducing overall system cost.

“The optimal band gap of an absorber layer for the most efficient conversion of sunlight into electricity is between 1.1 and 1.5 eV, with a corresponding maximum theoretical efficiency close to 33%.”

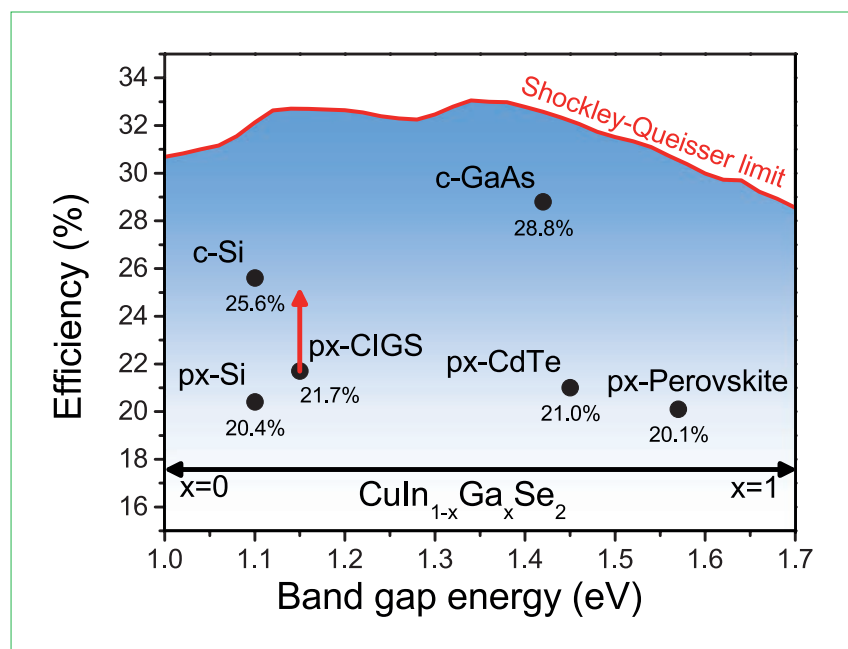


Figure 1. Shockley-Queisser limit (accounting for radiative recombination) for single-junction solar cells under an AM1.5G spectrum. Also shown are selected record efficiencies for different monocrystalline (c-) and polycrystalline (px-) PV technologies.

Polycrystalline CIGS is a stable p-type semiconductor material that can be used as the absorber layer in a thin-film solar cell. Its band gap can be continuously tuned from 1.0 to 1.7 eV by changing the ratio of In to Ga. This covers the ideal band-gap range for converting solar energy to electricity if compared with the Shockley-Queisser limit, which defines the theoretical maximum achievable conversion efficiency (Fig. 1). On the basis of those calculations, the optimal band gap of an absorber layer for the most efficient conversion of sunlight into electricity is in fact between 1.1 and 1.5 eV, with a corresponding maximum theoretical efficiency close to 33%. For CIGS solar

cells, the highest efficiencies have so far been achieved with an average energy band gap of 1.15–1.2eV.

There is still a large gap, however, between the Shockley-Queisser limit and the actual achieved values for current record 21.7%-efficiency CIGS solar cells. An argument that further progress can still be made to increase efficiencies by reducing the optical and electronic losses is discussed in this paper. It is now believed that efficiencies up to 25% are within reach in the mid term. Some aspects of CIGS development that have led to the current solar cell configuration and efficiency are presented here, and the origin of the remaining losses in record-efficiency devices is discussed.

Finally, the path towards multiple-junction polycrystalline thin-film solar cells in order to achieve even higher efficiencies approaching 30% is presented. Some aspects of production costs and industrial players are discussed, highlighting the unique advantages of this technology that should allow the cost of PV modules to be brought down even further, while opening the door to innovative applications and new markets.

A brief history of CIGS solar cell processing

From the appearance of the first solar cells with copper indium selenide (CIS) single crystals combined with evaporated CdS in 1974 [1] to the achievement of the recent record efficiency of 21.7% [2], many material combinations and growth methods have been tested, leading up to the current CIGS stack configuration (shown as an inset in Fig. 3). The typical substrate of the record-efficiency solar cells is soda-lime glass, but recently similar efficiencies have been reported on flexible polyimide substrate, in spite of the limitation of the low deposition temperature (<500°C) imposed by the plastic substrate. A sputtered Mo layer is used as a back contact, followed by a CIGS absorber grown by evaporation of individual elements or by sputtering followed by selenization. The pn junction is formed by the deposition of a so-called 'buffer layer', which is usually of n-type CdS deposited in a chemical bath, but alternative buffer layers, such as Zn(O,S), can also be used. The device is finished by a transparent conductive oxide (TCO), such as ZnO doped with Al or ITO, as well as a metal grid for better current collection.

Some of the advances have had a significant impact on the progress of technology. The first innovation was

the introduction of a thin (< 100nm) n-doped CdS buffer layer [3], which allowed the formation of a good-quality p-n junction with limited interface recombination between the CIGS absorber and the next layer, along with reduced optical losses due to absorption in the CdS layer.

A second major leap forward was realized when the role that Na plays in the CIGS electronic properties was reported [4]. Na is naturally present in the soda-lime glass substrate material and diffuses into the CIGS layer during growth at an elevated temperature, improving the electronic quality of the CIGS, which significantly enhances the solar cell efficiency. Furthermore, the partial replacement of In by Ga to increase the band gap of the absorber has led to a further efficiency improvement [5].

From a CIGS growth perspective, the shift from the co-evaporation process introduced by Boeing (evaporation from separate elemental sources) [6] to the so-called 'three-stage method' by NREL [7] paved the way for all further efforts towards achieving high efficiencies, allowing the growth of CIGS layers with enhanced crystalline quality, grain size and electronic properties; those concepts led to a 19.9% cell efficiency in 2007 [8]. Fig. 2 shows the subsequent improvements in cell efficiency, comparing high- and low-temperature deposition processes, along with the

progress of the highest-reported mini-module efficiency on plastic substrate. After 2007, the efficiency gap with px-Si solar cells became even less: incremental improvements in cell efficiency reached 20.3% [11] in 2011, mainly driven by optimization of the deposition methods, without any actual breakthrough in terms of material composition and deposition techniques.

A new boost in efficiency improvement, however, was recently triggered by Empa with the introduction of a novel processing step based on a post-deposition treatment (PDT) of the CIGS absorber layer with potassium fluoride (KF) [12,13].

In the process of depositing CIGS layers onto alkali-free substrates, alternative methods had to be developed in order to add the alkali, mainly Na, to take advantage of its beneficial effect on performance. In 2004 the concept of a PDT for doping CIGS layers with alkaline was developed at ETH Zürich (the research group relocated to Empa in 2009) [14]: NaF is evaporated onto the CIGS absorber at a temperature of around 350°C after growth has finished, allowing diffusion of Na into the layer and yielding similar effects on the absorber electronic properties to those when diffusing from the glass substrate. Most notably, this immediately allowed an improvement in record efficiencies of cells grown on flexible plastic films [15].

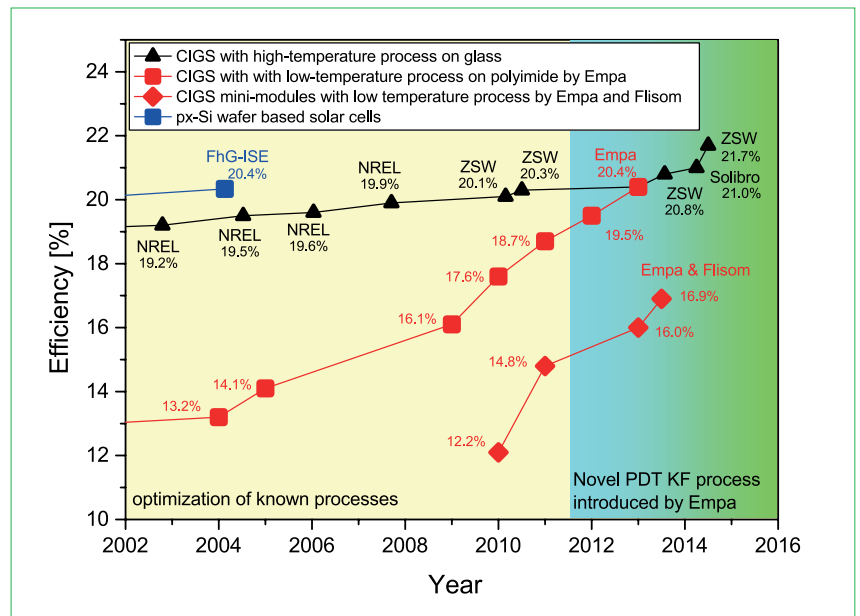


Figure 2. Efficiencies of CIGS solar cells processed by co-evaporation methods on glass substrates at high temperatures and on flexible polyimide films at low temperatures. Efficiencies are also shown for mini-modules processed by low-temperature co-evaporation (but not for those processed at high temperatures). With such high-temperature processes, Solibro has achieved a designated area efficiency of 18.7% for a mini-module [9]. The values for CIGS solar cells using a high-temperature process, as well as the values for px-Si, are taken from NREL [10].

“The introduction of potassium after the absorber growth has led to an efficiency of 20.4% on a flexible polyimide substrate.”

For a long time, Na was believed to be the most effective alkali element, which is a valid argument when it is present before or during CIGS growth. However, the introduction of potassium after the absorber growth has led to an efficiency of 20.4% on a flexible polyimide substrate and has proved advantageous in improving the quality of the interface between the CIGS absorber and the CdS buffer layer. Not long after that was announced, a new record of 20.8% on glass using a process based on KF PDT was reported [16]; more recently, further outstanding improvements in efficiency of up to 21.0% at Solibro [17] and 21.7% at ZSW [2] have been achieved using KF PDT.

Conventionally, these CIGS absorbers are grown by the co-evaporation of Cu, In, Ga and Se following a three-stage or multi-stage process. However, sputtering of Cu, In and Ga metallic precursors, followed by selenization and sulphurization processes, has also yielded a cell efficiency above 20% [18]

using a Cd-free buffer layer.

Overall, it is of fundamental importance not only to control the actual compositions of the CIGS and the other layers in a solar device, but also to adapt suitable techniques to grow and combine the different layers without any adverse effects on the properties of the previous and subsequent layers. Aspects such as compositional grading throughout the absorber layer, the amount and nature of crystal defects, or the elemental inter-diffusion at interfaces all have consequences for the final photovoltaic properties of the solar cells; understanding their interdependencies is essential for the processing of high-efficiency solar cells.

Potential for further performance improvement of up to 25% efficiency

As previously mentioned there exists a fundamental physical limit to the performance of single-junction solar cells (Shockley-Queisser limit) which assumes that, in equilibrium, the generation of charge carriers has to be balanced by an equal amount of recombination, and no other losses are present [19]. However, in reality other losses limit the performance of solar cells: these can be split into *optical* and *electronic* losses.

In a CIGS device, optical losses arise because of reflection at the surface, parasitic absorption of photons in the TCO and the buffer layer, and incomplete light absorption in the CIGS layer. The individual losses for a device produced at Empa with an efficiency of 20.4%, and using a CdS buffer layer, are quantified in Fig. 3. With an absorber band gap of 1.15eV, a maximum short-circuit current density (J_{sc}) of 42.3mA/cm² can be achieved in an ideal situation. As shown in Fig. 3, the most severe optical losses in a CIGS device occur as a result of parasitic absorption in the TCO and buffer layers. To eliminate, or at least reduce, the influence of the TCO on the optical losses, suitable materials with higher optical band gaps and higher mobilities are required. This is a big challenge for future research because several factors need to be considered: the optoelectronic properties, the limited deposition temperature the device can withstand (<200°C), and the price of materials.

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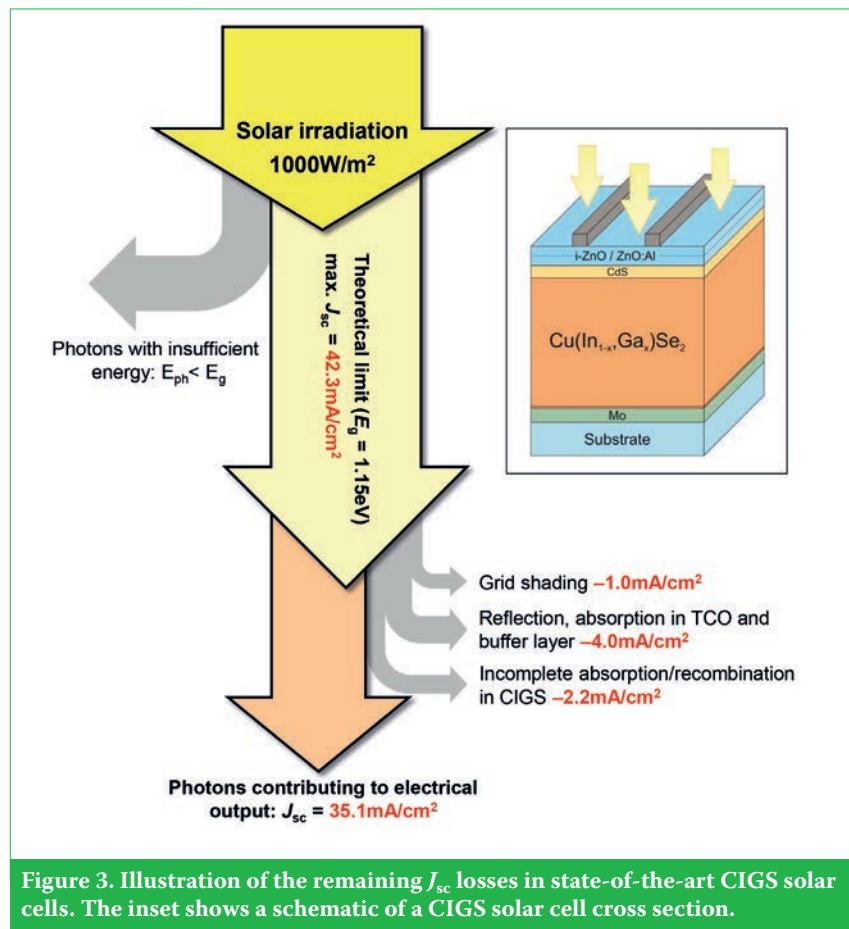


Figure 3. Illustration of the remaining J_{sc} losses in state-of-the-art CIGS solar cells. The inset shows a schematic of a CIGS solar cell cross section.

As regards the buffer layer, a lot of effort has been put into replacing CdS by a material with a larger band gap, such as Zn(O,S); Solar Frontier recently demonstrated that a very high efficiency of 20.9% can be achieved [18] this way. Another, even more challenging, approach would be to completely redesign the CIGS device structure in the direction of a rear-contact solar cell with a passivated front surface, so that the TCO and buffer layers can be completely omitted and both contacts are located on the back side. Such an approach has already been successfully applied to Si-based solar cells. To apply this concept to a CIGS absorber layer, however, the quality of the material needs to be further improved, because a long diffusion length of the generated charge carriers is necessary, so that they can reach their respective electrical contacts.

Concerning the electronic losses, which mainly influence the open-circuit voltage (V_{oc}) and fill factor (FF) of the solar cells, there is still an ongoing discussion about their actual origin. In general, recombination occurring at defects in the CIGS bulk or at any of its interfaces, or at grain boundaries,

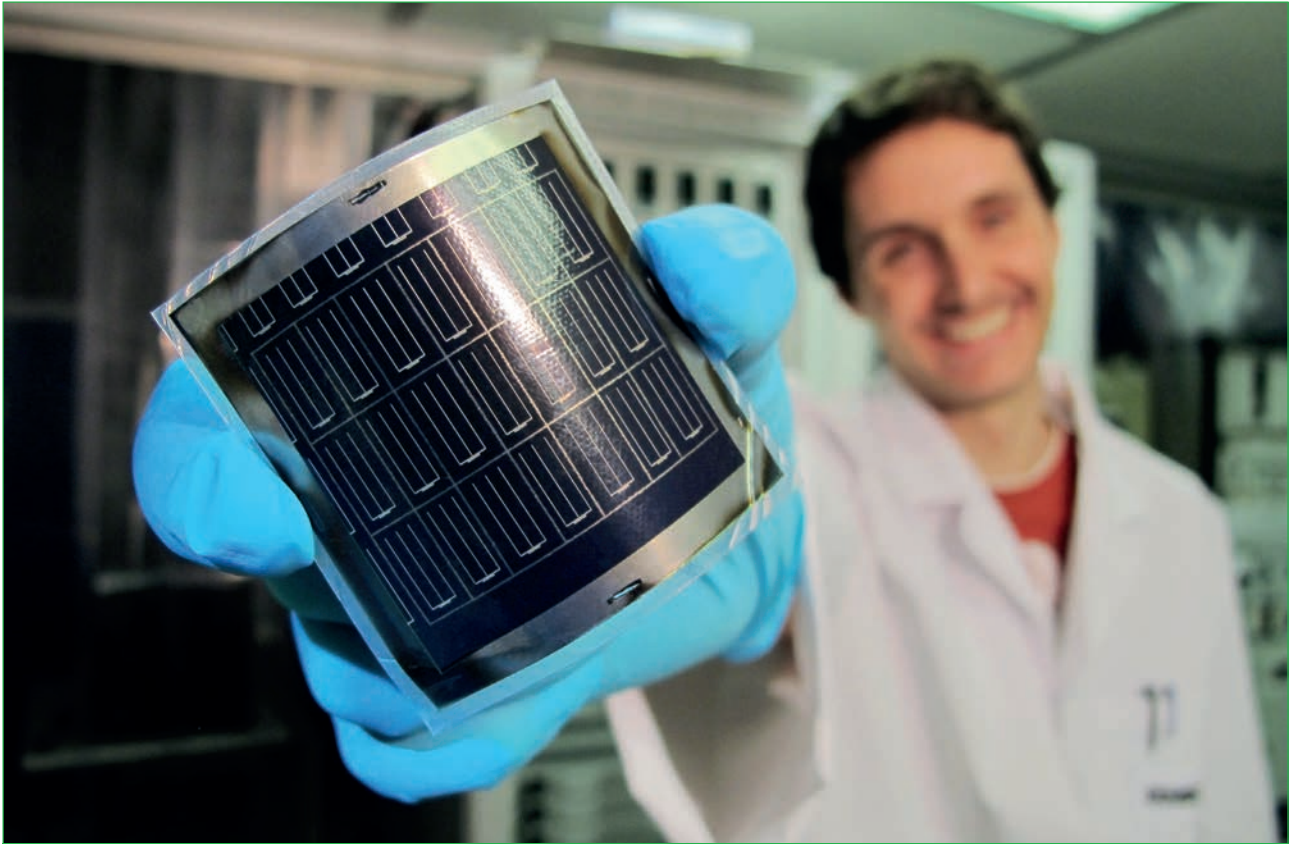


Figure 4. Empa flexible CIGS solar cell.

limits the device performance. Much effort is currently being devoted to better understanding each of these mechanisms and to developing new deposition methods and treatments for reducing their influence.

The PV parameters of a device with 20.4% efficiency produced at Empa (Fig. 4) are summarized in Table 1 and compared with the theoretically achievable values in the Shockley-Queisser limit. By overcoming some of the above-described remaining limitations, an improvement in J_{sc} by 10% to 38.5 mA/cm², in V_{oc} by 9% to 800 mV, and in FF by 3% to 81% seem feasible. In fact, such values have already individually been reported for this technology. Combining these best individual values in a single device would yield a CIGS solar cell efficiency of 25%. The first step in reaching this ambitious milestone should be to identify the basic properties of the layer and devices, and then design the processing accordingly. Indisputably, process optimization alone is not sufficient for achieving this goal – innovative approaches will also be required.

Advanced concepts for efficiencies towards 30% with tandem solar cells

A proven concept for enhancing efficiency beyond the values achievable with single-junction solar cells is a

	J_{sc} [mA/cm ²]	V_{oc} [mV]	FF [%]	η [%]
SQ limit (1.15eV)	42.3	887	87	32.7
Empa cell on polyimide	35.1	736	78.9	20.4
Difference relative to SQ limit	-17%	-17%	-9.3%	

Table 1. PV parameters of a CIGS solar cell processed on flexible polyimide substrate by Empa compared with the Shockley-Queisser (SQ) limit values for a single-junction solar cell with an energy band gap of 1.15 eV.

multi-junction device (e.g. tandem or triple-junction solar cell). Such a concept has been successfully applied to monocrystalline III-V semiconductors, yielding efficiencies well above 40% under concentrated light. Because of the high production costs, these devices are mainly used in very specialized applications, such as concentrated PV or space projects. In multi-junction solar cells, two or more cells with different energy band gaps are stacked on top of each other in such a way that high-energy photons are absorbed in the top cell and low-energy photons in the bottom one, leading to an optimal use of the solar spectrum. This raises the theoretical maximum efficiency from ~33% for single-junction solar cells to ~44% for tandem solar cells. However, this concept has not yet been successfully applied to polycrystalline thin-film solar cells, mainly because the technology for combining high efficiency and high transmittance for near-infrared light (NIR) is lacking.

This may change with the rapidly emerging perovskite solar cell, which has a tuneable band gap between 1.55 eV and 2.3 eV, making it an ideal candidate for the top cells in combination with CIGS solar cells at the bottom. Certified efficiencies of up to 20.1% have already been achieved for a single-junction perovskite solar cell [10]. For their implementation as top cells in tandem devices, perovskite solar cells with two transparent contacts are currently being developed in order to allow IR light to be transmitted to the CIGS bottom cell.

Different approaches can be used to combine CIGS and perovskite solar cells in tandem devices. The most straightforward is the separate production and subsequent stacking of the devices (Fig. 5); however, this can lead to losses of NIR light in three highly conductive TCO layers. Alternatively, perovskite solar cells can be monolithically grown on CIGS solar cells, which is facilitated by the low processing temperatures

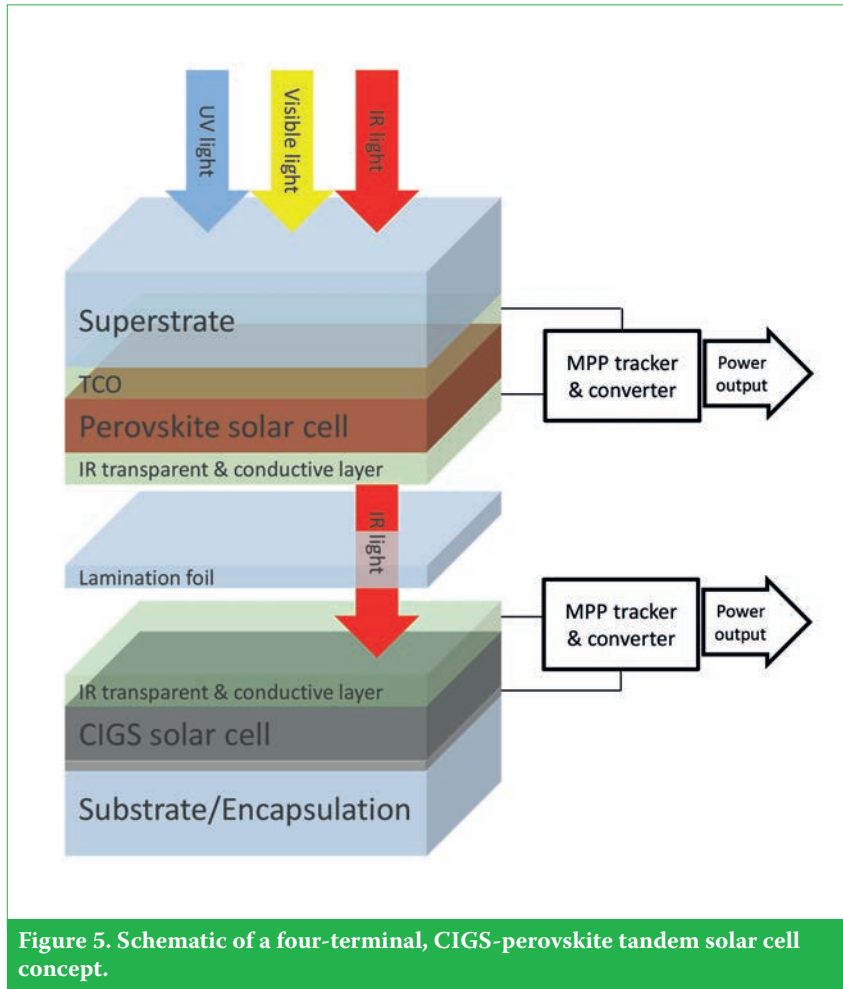


Figure 5. Schematic of a four-terminal, CIGS-perovskite tandem solar cell concept.

(<200°C) for perovskite solar cells. Several methods to prepare CIGS-perovskite tandem solar cells are currently being investigated and that research is expected to lead to these cells demonstrating efficiencies edging towards 30% in the near future [20]. Wafer-based tandem solar cells have shown their merits for better utilization of the solar spectrum, but the development of large-area manufacturable CIGS thin films as the bottom cell will break the bottleneck related to NIR-light loss, potentially leading to low-cost polycrystalline thin-film tandem solar cells.

CIGS from an industrial perspective – market for CIGS

Despite the emergence of several CIGS companies about 15 years ago, the full transfer of lab-scale results to industrial production is still ongoing. The crisis which hit the entire PV industry, mainly due to oversized production capacity, led to the disappearance of a significant number of companies in all types of PV technology in the last three years. The surviving CIGS companies are now scaling up production while still remaining active on the research front with the aim of further reducing

production costs and increasing module efficiencies. Solar Frontier is currently the largest CIGS player in the market, with a production capacity of more than 1GWp. Hanergy Group has acquired several CIGS manufacturers – such as Solibro, Global Solar and Miasolé – and is now planning to build up to a 3GWp production capacity within the next few years. A more detailed overview of some of the other active players is given in Reinhard et al. [21].

Key aspects for competitiveness of this technology are higher efficiencies and lower production costs of the modules. Solar Frontier, for example, is targeting to increase its average module efficiency to at least 16% by 2016 [22]. Solibro expects a module efficiency of 17% to be feasible by 2017 [23]. Manz announced a CIGS module production cost of around €0.41/W for a 147MW/annum production line. Bosch projects production costs below \$0.38/Wp in a 1GW plant with 16.3% average module efficiency.

Thin-film PV technologies currently make up 8% of the total market, whereas CIGS holds only 2%, but the production volume of CIGS is increasing. Besides ground-mounted CIGS modules, lightweight and flexible products in particular are advantageous

for BIPV and rooftop applications. The global market related to buildings is expected to grow from 23GW in 2013 to 33GW by 2017, while there remains a huge market if one considers metal roofs and facades [24]. It is predicted that the building-related CIGS market could achieve an additional 25–30GW by 2020. Flexible and lightweight solar modules can be directly laminated to such metallic building components, resulting in further cost benefits and additional functionality for electricity production.

“The recently achieved efficiency improvements at the solar cell level are a solid basis for constructing CIGS solar modules with efficiencies above 17%.”

Conclusions

In summary, the recently achieved efficiency improvements at the solar cell level are a solid basis for constructing CIGS solar modules with efficiencies above 17%. If the research community succeeds in driving solar cell efficiency towards 25%, full-area modules with efficiencies exceeding 20% will be possible. Conceivably, even higher performance is possible with the concept of tandem devices, though this would mean an additional processing line for the top cell. However, provided the production costs are mainly determined by the substrate and encapsulation, this is a viable possibility for creating highly efficient modules and generating low-cost solar electricity. The efficiency targets presented here might appear to be highly ambitious, but from a retrospective look at the historical trend of progress made in CIGS and compound semiconductor thin-film PV, the chances of their realization in the future are high.

Acknowledgement

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Dr. Stephan Buecheler studied physics at ETH Zurich and received his diploma in 2007. After receiving his Ph.D. from ETH Zurich in 2010, he took the position of group leader in the Laboratory for Thin Films and Photovoltaics at Empa, where his research group is currently working on highly efficient CIGS, CdTe and perovskite solar cells, including advanced tandem concepts.

Dr. Fabian Pianezzi studied physics at ETH Zurich and received his master’s in 2009. He then joined Empa as a Ph.D. student and focused on characterizing the electronic properties of CIGS solar cells. In 2014 he took up a postdoctoral position in the Laboratory for Thin Films and Photovoltaics at Empa, where his work focuses on solar cell device characterization using advanced diagnostic tools.

Patrick Reinhard studied materials science at ETH Zürich and received his master’s in 2011, with a specialization in material properties and analysis. He joined Empa as a Ph.D. student in

2012, and has since been involved in the development of high-efficiency, flexible CIGS solar cells. His work mainly focuses on the bulk and interface modifications of CIGS absorber layers induced by alkali treatments, especially potassium related.

Enrico Avancini received his bachelor’s in physics in 2012 from Trento University in Italy and later completed his studies at Freie Universität Berlin, obtaining an M.Sc. in physics in 2014. In November the same year he joined the Laboratory for Thin Films and Photovoltaics at Empa as a Ph.D. student.

Dr. Lukas Kranz studied nanoscale engineering at the University of Würzburg, Germany. He conducted his Ph.D. thesis at Empa, where he worked on the development of CdTe thin-film solar cells on glass and flexible metal-foil substrates. In 2014 he became a postdoctoral researcher at Empa, where his research focuses on the development of NIR-transparent perovskite solar cells for application with CIGS in tandem devices.

Fan Fu obtained his master’s in materials science from Wuhan University of Technology, China, in 2013, after which he started on his Ph.D. at Empa. His current research is focused on the design, synthesis and characterization of organic–inorganic hybrid perovskite solar cells and exploring the potential of their use in tandem structures in combination with CIGS.

Dr. Ayodhya N. Tiwari received his M.Sc. from the University of Roorkee, India, and his Ph.D. from the Indian Institute of Technology (IIT) in New Delhi. He heads the Laboratory for Thin Films and Photovoltaics at Empa and is a titular professor at the Swiss Federal Institute of Technology Zürich (ETH-Zürich). Prof. Tiwari is a co-founder and the chairman of Flisom AG, a Swiss company involved in the production of monolithically interconnected flexible and lightweight CIGS solar modules.

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