Thin Film Solar Cells: Research in an Industrial Perspective

Marika Edoff

Abstract Electricity generation by photovoltaic conversion of sunlight is a technology in strong growth. The thin film technology is taking market share from the dominant silicon wafer technology. In this article, the market for photovoltaics is reviewed, the concept of photovoltaic solar energy conversion is discussed and more details are given about the present technological limitations of thin film solar cell technology. Special emphasis is given for solar cells which employ Cu(In,Ga)Se_2 and $\text{Cu}_2\text{ZnSn(S,Se)}_4$ as the sunlight-absorbing layer.

Keywords Solar cells · Photovoltaics · Thin film · Electricity · CIGS · CZTS

INTRODUCTION

Motivated by the interest for climate change and in view of the limited resources for energy, electricity generation by photovoltaics (PV) is increasing in capacity worldwide. The merits of PV are obvious; it is silent, renewable and solar energy is the most abundant source of energy on earth. More than 35 GW of photovoltaic power (W_p) has been installed until 2010. This corresponds to a yearly generation of electricity of above 35 TWh. The growth is very fast with an increase in capacity during 2010 of 12 GW_p corresponding to a 50% increase as compared to end of 2009 according to the European Photovoltaic Industry Association.

The vision of solar cell research is for photovoltaic electricity generation to become competitive with the conventional sources of electricity. Evidently, the ideal photovoltaic system would employ solar panels with very low production costs, high efficiency and a good design, enabling easy and low-cost installation. Some of these

requirements may be interchangeable as, for example, very low production cost and limited lifetime may give similar results as higher production cost and very long lifetime. The bottom line to all this is the cost per energy unit, or kWh. This cost will include not only the solar panels, but also the balance of system (BOS) costs, which consist of all other components required to get electricity as well as costs for installation. Depending on the site for the photovoltaic installation, different amounts of energy will be produced. The amount of electric energy and its distribution in time over the year will depend both on the latitude of the installation as well as on the microclimate, tilt angle, azimuth angle (S, SE or SW) and possible shadowing problems.

The thin film solar cell technology is increasing in capacity and market share and is now accounting for about 20% of the PV market. CdTe solar cells represent about half and the rest is divided between thin film silicon and Cu(In,Ga)Se_2 where the latter is increasing rapidly. The primary advantage for Cu(In,Ga)Se_2 -based solar cells is the high efficiency, which has now exceeded the 20% level (Jackson et al. 2011). Much of this improvement is due to improvements in the semiconductor material quality. However, as the material quality of the semiconductors improves, other parts of the solar cell are becoming the new bottlenecks to increase the efficiency further towards the theoretical limit for non-concentrated light, the so-called Shockley–Queisser limit, which is 30% for single junction solar cells, such as the Cu(In,Ga)Se_2 (CIGS) and $\text{Cu}_2\text{ZnSn(S,Se)}_4$ (CZTS)-based solar cells discussed in this contribution (Henry 1980). In order to overcome the 30% limit, it is possible to combine these materials to form tandem junction solar cells, for example by stacking a CuGaSe_2 cell with a bandgap of 1.7 eV on top of a CuInSe_2 solar cell with a 1.0-eV bandgap. However, up to

now such efforts have not yielded solar cells exceeding the present 20% efficiency for the single junction devices and therefore this approach will not be discussed here.

In this review, research on these two related low-cost photovoltaic materials for thin film solar cell applications will be discussed in a perspective of near future use for very large-scale implementation as generators for electricity.

THE THIN FILM SOLAR CELL

The basic design of a thin film solar cell is a sunlight-absorbing layer squeezed in between two contact layers. One or more extra layers may be necessary to passivate the boundaries between the layers, otherwise these interfaces may act as recombination centres that reduce the number of generated electrons and thereby reduce the conversion efficiency. An electron microscopy image of a thin film solar cell based on the material $\text{Cu}(\text{In,Ga})\text{Se}_2$ (CIGS) is shown in Fig. 1. The outline for a thin film $\text{Cu}_2\text{ZnSn}(\text{S,Se})_4$ (CZTS) solar cell is similar. In addition to the absorber and contact layers, electrical contacting is needed, either to contact individual cells or to form monolithically integrated solar cell modules.

By absorption of photons in the sunlight, electron hole pairs are created. These electron hole pairs are separated by the action of an electric field in the p–n junction, which is formed between the p-type absorber layer and the n-type

buffer and window layers. In the process of separation, electrons are swept over the p–n junction and collected at the surface of the solar cell on the other side of the p–n junction, which becomes negatively charged. Consequently, the backside of the solar cell becomes positively charged since it now has a deficiency of electrons. The difference in the amount of electric charge carriers between the negative and positive contacts is the reason for the difference in electric potential. Upon contacting an external load, electrons will flow from the negative contact through the load to the positive contact. The electric load can be adjusted to optimize the output from the solar cell to the maximum power point. The amount of power dissipated in the external load at the maximum power point is divided with the power of the incident light and the result is the solar to electric conversion efficiency of the solar cell according to Eq. 1.

$$\eta = \frac{P_{\text{mp}}}{P_{\text{in}}}, \quad (1)$$

where η is the conversion efficiency, P_{mp} is the power dissipated in the external load at the maximum power point and P_{in} is the power of the incident light.

The main advantages with both CIGS and CZTS are their respective high absorption coefficients for the solar spectrum. This enables the thickness of the light absorbing layers to be between 1 and 2 μm as opposed to crystalline silicon, which needs to be 50–100 times thicker. The low thickness releases some of the constraints on material quality, since electrons hole pairs are generated close to the charge separating p–n junction. Another advantage is the ability to tune the bandgap to make an optimum match to the solar spectrum. In CIGS this is made by changing the concentrations of indium and gallium, in CZTS it is possible to mix sulphur and selenium.

Status of CIGS and CZTS

CIGS is in the process of large-scale commercialization, in contrast to CZTS, which is still in the early research phase. Some of the challenges in research and commercialization will be discussed in the following.

Using the basic device layout presented in Fig. 1, efficiency highlights are presented in Table 1. As can be observed, there is a gap between record cell efficiencies and the values for modules from research and additionally to modules from regular production. There are two fundamental reasons for the discrepancy between cell and module efficiencies. The first is the differences in active area, where the dead area in the cell interconnects in a module is included in the aperture area. The second difference is caused by parasitic absorption in the transparent conductive window layer, which needs to be thicker and

Fig. 1 Outline of a thin film solar cell based on $\text{Cu}(\text{In,Ga})\text{Se}_2$. The different layers are indicated from top to bottom in the figure as window layers, absorber layer and back contact. The back contact consists of molybdenum metal, the absorber of $\text{Cu}(\text{In,Ga})\text{Se}_2$ and the window layer of two buffer layers and a transparent conducting oxide layer. SEM image: Marika Edoff

Table 1 Thin film device efficiency records

Type of device	Efficiency (%)	Manufacturer
Small CIGS lab cell on glass	20.3	ZSW, Germany (Jackson et al. 2011)
Small CZTS lab cell on glass	9.7	IBM, USA (Todorov et al. 2010)
Small CIGS lab cell on flexible polymer	18.7	EMPA, Switzerland (Chirila et al. 2011)
CIGS Submodule 30 × 30 cm ²	17.2	Solar Frontier, Japan
Full size CIGS module, interconnected large cells	15.7	Miasolé, USA
Full size CIGS monolithic module, regular production	14.6	Solibro, Germany

thereby more absorbing in a module than in a small area solar cell. These two reasons alone allow for a difference in module to cell efficiency of ~ 2 absolute%, i.e., that a solar cell module made from a 20% efficiency record cell material would have about 18% efficiency. The rest of the differences between cell and module can be overcome by optimization of the module production processes improving uniformity and material properties over large areas.

Efficiency values are from measurements performed at standard test conditions, i.e., at a light intensity of 1000 W/m², at 25°C cell temperature and at a solar spectrum corresponding to AM1.5, which means that the sunlight has travelled 1.5 times through the atmosphere before it reaches the device. The best efficiency values obtained for this type of devices are comparable to those obtained for solar cells and modules made from multicrystalline silicon, the technology which dominates the present market. To reach similar efficiencies compared to the main competitor is, however, not enough. In order for a new product to compete with an established one on a very tough market, there must be more benefits to the new technology. One of the most important competitive advantages with thin film is the potential for low-cost production, partly by low use of active materials and partly by highly automated processing and handling during production.

In total, the industrial production of CIGS-based solar modules during 2010 was around 400 MW_p. This number should be compared to the total number of solar modules, which was about 17.5 GW according to GTM market research and PV News. The CIGS module production is predicted to expand to beyond 1 GW_p capacity during 2011. Even if the material use for the active components of the thin film modules is low, expansion to the multigigawatt scale may be limited by the availability of indium, gallium and selenium. The material constraints for different PV technologies are reviewed by Zuser and Rechberger (2011); they find that indium may be a constraint at a cumulative installed capacity of a few 100 GW_p, i.e., much higher than the predicted production in the near future, but far from enough for supplying the world with PV electricity generated from CIGS PV modules. For the CdTe

technology the availability of Te is even more limited. This is the main reason for the interest in photovoltaic materials, such as CZTS, made solely from abundant elements.

CHALLENGES FOR CIGS RESEARCH

The CIGS material system is still not very well known in spite of high device efficiencies. In order to push the efficiency values further from the present 20% towards the theoretical limit of 30% for a single junction device, a number of issues have to be solved. Two of these are discussed more in detail below.

Doping

CIGS is a semiconductor that is doped by defects and for thin film the material is always a p-type semiconductor. The exact nature of the doping is debated but some of the defects will add to the p-type doping such as Cu vacancies and some may be compensating donors, such as Se vacancies and In_{Cu} antisite defects. Theoretical calculations have explained the large tolerance to compositional variations with clustering of Cu vacancies and In at copper sites ($2 V_{Cu} + 1 In_{Cu}$). These clusters are neutral and do not contribute to the doping, but can take up deficiency of copper with respect to indium and gallium in the crystal structure.

It is also known that a much higher carrier concentration is obtained if Na is added to the CIGS material. Na may come from the glass substrate commonly used for this type of devices or can be added in the processing. Na is reported by Wei et al. (1999) to suppress the formation of the defect cluster and thereby increase the effective doping of the CIGS material.

BUFFER LAYERS AND INTERFACES

The n-type partner in the p–n junction in CIGS solar cells consists of a buffer layer followed by a non-doped layer of

ZnO. On top of these buffer layers a doped ZnO layer is deposited to carry the current. A commonly used buffer layer is CdS, but since this layer contains Cd it is preferred to use alternative materials. In addition to the environmental concerns, another disadvantage with the CdS buffer layer is that it absorbs light in the UV region of the solar spectrum. If this parasitic absorption can be avoided, the conversion efficiency increases by increased current density. A number of alternative buffer layers have been tested and shown promising results (Naghavi et al. 2010). Best results are for Zn(O,OH,S) and (ZnMg)O layers, but also ZnSnO and In₂S₃ buffer layers are good alternatives. Recently, CIGS solar cells with high efficiency were obtained using a ZnSnO buffer layer (Hultqvist et al. 2011). The suitability of a new material as buffer layer in CIGS-based solar cells is partly dependent on the nature of the interface between the CIGS material and the buffer layer. If the CIGS/buffer interface is blocking the electrons from getting into the junction, then the efficiency decreases drastically.

The other important interface is found at the back contact. The back contact is made of molybdenum metal, which selenizes slightly upon exposure to selenium. The MoSe₂ layer is reported to be beneficial for the back contact properties, but is still a region of high recombination velocity. A perfectly passivated back contact would allow for thinning down of the CIGS layer and still obtaining highly efficient devices.

Bandgap Engineering

One important asset of CIGS (as well as for CZTS) is its ability to use compositional variations to adjust the bandgap. This property can be used in two ways: the first is to adjust the bandgap to the optimum obtained in calculations of theoretical maximum efficiency, the other to use in-depth bandgap grading to improve electronic transport in the material. With a good design of in-depth gallium profiles the combination of high absorption (caused by a low bandgap where maximum absorption takes place) and high voltage (caused by a high bandgap in the near junction region) can be obtained.

Modelling, both of the electronic structure of the materials as well as electrical modelling of devices, is becoming increasingly important to penetrate deeper into understanding where the new bottlenecks in efficiency can be found.

CHALLENGES FOR CZTS RESEARCH

The CZTS is even less understood in terms of doping properties and defects, but preliminary calculations predict

that this material may have similar properties as CIGS (Persson 2010; Siebentritt et al. 2010).

Obtaining Single-Phase Kesterite

A major challenge in the making of CZTS as well as CZTSe material is to obtain single-phase kesterite, which is the material with the best photovoltaic properties. The single-phase region is rather narrow as compared to CIGS. During fabrication of CZTS and CZTSe it seems important to avoid the combination of high temperatures and the presence of secondary phases, such as tin sulphide and tin selenide which both are highly volatile. Loss of tin leads to loss of stoichiometry and precipitation of other secondary phases, such as Cu₂S(Cu₂Se) and ZnS(ZnSe), which are detrimental to device performance. In Fig. 2, a cross-section electron microscopy image shows precipitation of ZnSe in a Cu₂ZnSnSe₄ thin film. The best result up to now has been obtained by dissolving nanoparticles in a hydrazine solvent followed by annealing (Todorov et al. 2010).

The CZTS Cell Structure

The first generation of CZTS solar cells still use the same cell structure as the CIGS cells, with CdS buffer layer and TCO and with a back contact made of molybdenum metal. Depending on deposition method various amounts of MoSe₂ or MoS₂ are formed at the back contact. The use of

Fig. 2 A cross-section transmission electron microscopy image showing a CZTSe film. The large grains consist of CZTSe, whereas the small grains found to the right in the upper part of the CZTSe film (in the rectangle) are ZnSe precipitates. TEM image: Timo Wätjen

alternative buffer layers to CdS has up to now not yielded solar cells with the same efficiency as when CdS has been used.

Challenges with Upscaling to Product Size Modules

A common size for photovoltaic modules is about 1 m², sometimes slightly less, sometimes more. The advantage of using large modules is that it takes fewer modules to make a system and thereby less mounting material, the disadvantage is that they may be difficult to handle for one person. Another argument for large modules is that by increasing the module area the surface/edge ratio increases, which leads to an increase of the total area efficiency. Since one junction box is required for each module, the cost per installed capacity will also decrease if fewer boxes are needed. An overview of module sizes and efficiencies for different CIGS manufacturers are given in Fig. 3.

Several deposition methods are used in manufacturing of CIGS for photovoltaic modules, for example selenization of sputtered metal layers (Solar Frontier and Honda Soltec), reaction of sputtered and evaporated stacked elemental layers in reactive atmosphere (Miasolé, Solteure and Avancis) and co-evaporation (Solibro, Global and Würth). Best results up to now on the cell level have been obtained for co-evaporation, but on the module level sputtering and selenization have both yielded high efficiency.

As was briefly discussed in the “Introduction”, the basic unit that has to be considered when calculating electricity cost is the kWh or produced amount of electricity. Cost per kWh will be decided mainly by photovoltaic system cost, system lifetime and amount of solar radiation at the installation site. The photovoltaic system cost is divided in module cost, installation cost and BOS cost. The two latter components will partly be dependent on the size of the system and partly on the power produced by the system.

Fig. 3 Total area efficiency (blue bars) and module power (red squares), as measured under standard test conditions for a number of commercial CIGS-based photovoltaic modules on the market. Data taken from IEA-PVPS, from the company webpages and product data sheets

Installation of larger modules or a larger amount of modules will cost more. Therefore, module efficiency will come into the cost structure for installation and BOS. Thus, even if the cost per square metre module is low, the system cost for a specific installed power may be high. To give specific numbers for costs is very difficult in view of the sharp decline in prices for installed systems during the last year, but PV modules at the end of 2011 were sold in large volumes at prices at below 1 Euro/Wp according to Solarbuzz (www.solarbuzz.com), which regularly lists both module prices and other components.

RESEARCH FOR HIGH EFFICIENCY SOLAR CELLS

We work on complete in-house fabrication of solar cells based on co-evaporation of Cu(In,Ga)Se_2 films and more recently also cells based on the light absorbing material $\text{Cu}_2\text{ZnSn(S,Se)}_4$. We perform extensive characterization of solar cells including temperature-dependent measurements as well as all relevant material characterization.

The research is divided into five research areas.

Synthesis and Characterization of CIGS Materials by Co-Evaporation

Co-evaporation gives a high flexibility to design bandgap grading for optimum solar cell device performance, therefore part of this area has been dedicated to work with this task (Schleussner et al. 2011), which also is supported by 1D computer modelling using the latest version of SCAPS (Decock et al. 2011). Another important issue is influence of alkaline elements. Na increases the carrier density in this type of solar cells. In order to use other substrate materials than soda-lime glass, such as plastic foil or metal foil, Na has to be added by other means. One successful way of adding Na is by precursor layers made of NaF. This was a pioneering work by our group, but is still highly relevant.

High quality CIGS layers is a prerequisite for advancing the technology, both in terms of other components of the solar cell structure, but also in the work with alternative absorber layers and alternative processing, where it serves as a solid reference.

Process Development of Absorber Layers by Sputtering

Sputtering is a high throughput deposition method, which is easy to employ to coat large areas. In several production facilities sputtering is used to fabricate metal layers for subsequent reaction to CIGS. Our approach is to use reactive sputtering in a specially designed reactor to fabricate high quality CIGS layers. The reactor has been built and is in the final stage of installation.

Buffer Layer Development

A versatile method of depositing thin layers of various kinds of materials is to use atomic layer deposition (ALD). Using ALD, excellent thickness control, composition control and perfect coverage of the thin layers are obtained. In Table 2, which lists the best devices using various buffer layers, shows that good results have been obtained with a variety of materials. Buffer layers of CdS are commonly used as a reference process. CdS has up to now been used by industry for CIGS devices.

New Absorber Materials

The research at Uppsala University on $\text{Cu}_2\text{ZnSn(S,Se)}_4$ has been built up during the last 1.5 years. The equipment consists of a sputter system for reactive sputtering as well as a furnace for post-treatment of the layers. In the present build-up phase the emphasis of the work is on understanding fundamental reaction processes (Scragg et al. 2011), but first devices showing up to 3% efficiency have been produced.

Environmental Evaluation and Testing of Solar Cell Modules

With a magnificent view of the Uppsala landscape a number of commercial as well as research solar cell modules are installed. In order to be able to predict electricity production from different kinds of module technologies and to relate yearly output to measurable parameters already before the installation, frequent measurements are done and evaluated. One measurement every 10 s is

Table 2 Small cell efficiency of various buffer layers produced at Uppsala University. The Zn(O,S) result has been confirmed by NREL (Zimmermann et al. 2006)

Buffer layer	Small cell efficiency (%)	Deposition method
Zn(O,S)	18.5	ALD (Zimmermann et al. 2006)
$(\text{Zn,Mg})\text{O}$	18.1	ALD (Hultqvist et al. 2007)
ZnSnO	18.0	ALD (unpublished results)
CdS (reference)	18.8	CBD (unpublished results)

collected both from the modules and from a calibrated pyranometer. A maximum power point tracker dedicated for this application has been constructed and is since 1 year tested in real conditions (Zimmermann and Edoff 2011).

SUMMARY

Thin film solar cell industry is taking a larger share of the total photovoltaic market. The reasons can be found in reduced production costs and increased efficiency for this type of solar cells. Limitations in efficiency for the thin film solar cells are found in parasitic absorption and limited carrier lifetime as well as recombination in the contact and junction regions. All these issues are addressed by research with the objective of getting closer to the theoretical limit for single junction devices, which is 30%. Currently, the best thin film solar cells have above 20% efficiency and are based on the semiconductor alloy material $\text{Cu}(\text{In,Ga})\text{Se}_2$. A new material, which earns an increasing interest, due to the fact that it is made by abundant elements, is $\text{Cu}_2\text{ZnSn}(\text{S,Se})_4$. This material is believed to have many similarities to $\text{Cu}(\text{In,Ga})\text{Se}_2$, but is still in an early research phase.

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Marika Edoff (✉) received her Ph.D. degree from the Royal Institute of Technology in Stockholm, Sweden in 1997. In 2003, she became group leader for the Thin Film Solar Cell group at Uppsala University, and in 2012 she was promoted to professor. During her leadership, the research of the Thin Film Solar Cell group has broadened to also include new absorber materials, such as CZTS. The research also comprises Cd-free buffer layers for solar cells and research on up-scaling issues and module technology. She is one of four founders of the spin-off company Solibro AB, which today is a full-scale manufacturer of CIGS-based solar cell modules in Thalheim in eastern Germany.

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