Edited by
Gavin J. Conibeer
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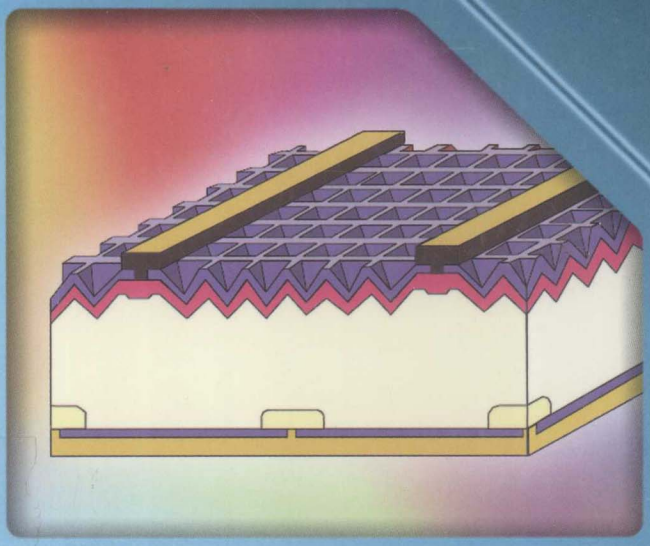
Solar Cell Materials

Developing Technologies



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in Materials for
Electronic
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Solar Cell Materials

Developing Technologies

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Series Preface

WILEY SERIES IN MATERIALS FOR ELECTRONIC AND OPTOELECTRONIC APPLICATIONS

This book series is devoted to the rapidly developing class of materials used for electronic and optoelectronic applications. It is designed to provide much-needed information on the fundamental scientific principles of these materials, together with how these are employed in technological applications. The books are aimed at (postgraduate) students, researchers and technologists, engaged in research, development and the study of materials in electronics and photonics, and industrial scientists developing new materials, devices and circuits for the electronic, optoelectronic and communications industries.

The development of new electronic and optoelectronic materials depends not only on materials engineering at a practical level, but also on a clear understanding of the properties of materials, and the fundamental science behind these properties. It is the properties of a material that eventually determine its usefulness in an application. The series therefore also includes such titles as electrical conduction in solids, optical properties, thermal properties, and so on, all with applications and examples of materials in electronics and optoelectronics. The characterization of materials is also covered within the series in as much as it is impossible to develop new materials without the proper characterization of their structure and properties. Structure-property relationships have always been fundamentally and intrinsically important to materials science and engineering.

Materials science is well known for being one of the most interdisciplinary sciences. It is the interdisciplinary aspect of materials science that has led to many exciting discoveries, new materials and new applications. It is not unusual to find scientists with a chemical engineering background working on materials projects with applications in electronics. In selecting titles for the series, we have tried to maintain the interdisciplinary aspect of the field, and hence its excitement to researchers in this field.

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1 Introduction

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1.1 INTRODUCTION

The environmental challenges to the world are now well known and publicised, and all but a small minority of scientists accept that a reduction on dependence on fossil fuels is essential for addressing the problems of the greenhouse effect and global warming. Everyone is aware of the limited nature of fossil-fuel resources, and the increasing cost and difficulty, as well as the environmental damage, of extracting the last remnants of oil, gas and other carbonaceous products from the earth's crust.

Photovoltaics, the conversion of sunlight into useful electrical energy, is accepted as an important part of any strategy to reduce this dependence on fossil fuels. All of us are now familiar with the appearance of solar cell modules on the roofs of houses, on public buildings, and more extensive solar generators. Recently, the world's PV capacity passed 100 GW, according to new market figures from the European Photovoltaic Industry Association (14 February 2013), which makes a substantial contribution to reducing the world's carbon emissions.

It is the aim of this book to discuss the latest developments in photovoltaic materials which are driving this technology forward, to extract the maximum amount of electrical power from the sun, at minimal cost both financially and environmentally.

1.2 THE SUN

The starting point of all this discussion is the sun itself. In his book 'Solar Electricity' (Wiley 1994), Tomas Markvart shows the various energy losses to the solar radiation that occur when it passes through the earth's atmosphere (Figure 1.1).

The atmosphere also affects the solar spectrum, as shown in Figure 1.2.

A concept that characterises the effect of a clear atmosphere on sunlight, is the 'air mass', equal to the relative length of the direct beam path through the atmosphere. The extraterrestrial spectrum, denoted by AM0 (air mass 0) is important for satellite applications of solar cells. At its zenith, the radiation from the sun corresponds to AM1, while AM1.5 is a typical solar spectrum on the earth's surface on a clear day that, with total irradiance of 1 kW/m^2, is used for the calibration of solar cells and modules. Also shown in Figure 1.2 are the principal absorption bands of the molecules in the air. AM1.5 is referred to frequently in a number of the chapters in this book, and readers should be aware of its meaning.

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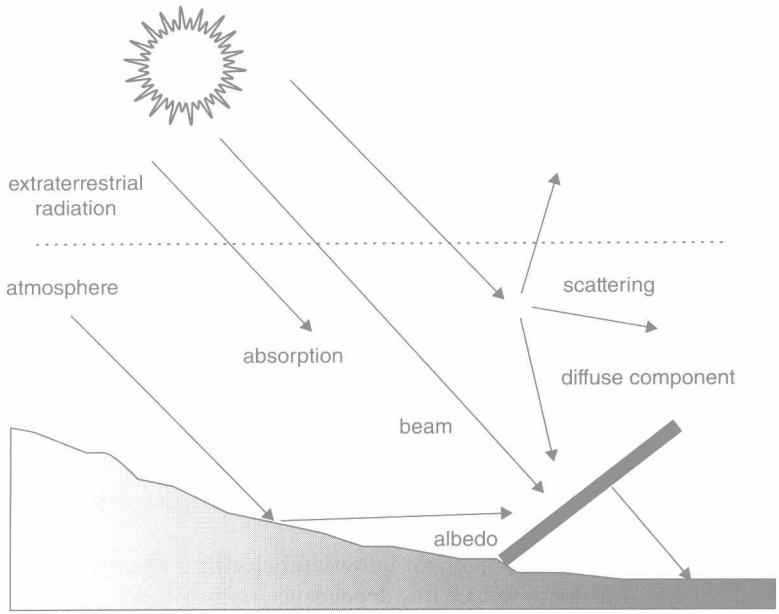

Figure 1.1 Solar radiation in the atmosphere. (Reproduced with permission from Markvart, 2000. Copyright © 2000, John Wiley & Sons, Ltd.)

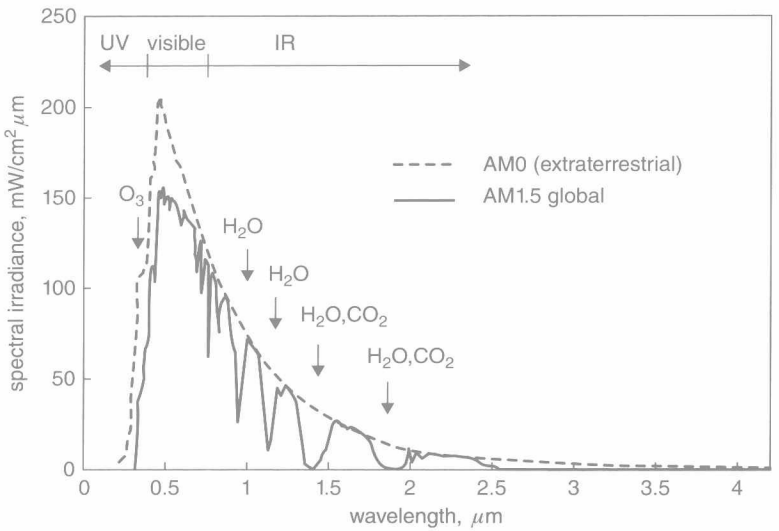

Figure 1.2 The solar spectrum. (Reproduced with permission from Markvart, 2000. Copyright © 2000, John Wiley & Sons, Ltd.)

1.3 BOOK OUTLINE

The book starts with a clear exposition of the fundamental physical limits to photovoltaic conversion, by Jean-Francois Guillemoles. This covers the thermodynamic limits, the limitations of classical devices, and develops this theme for more advanced devices. The identification of device parameters used in other chapters can also be found in this chapter.

Material parameters, of course, also require a thorough understanding of characterisation tools, and the second chapter, by Daniel Bellet and Edith Bellet-Amalric, outlines the main material characterisation techniques of special interest in solar cell science. X-ray analysis, electron microscopy, ion-beam techniques and spectroscopy characterisation methods are discussed, including Raman, X-ray photoelectron and UV/Visible spectroscopy, which are rarely detailed in such a materials book.

The next chapter, by Martin A Green, concentrates on developments in crystalline silicon solar cells. Despite the fact that silicon is an indirect-bandgap semiconductor, and therefore is a much less efficient absorber of above-bandgap light than direct-gap semiconductors (such as GaAs), silicon is still the overwhelming choice for solar cell manufacture. As the second most abundant element in the earth's crust, with a well-established technology, the chapter explores recent developments that have produced low-cost devices with efficiencies approaching the maximum physically possible.

Amorphous and microcrystalline silicon solar cells, are next reviewed by Ruud E I Schropp. These thin-film technologies are finding many exploitable applications with their lower usage of absorber materials and use of foreign substrates.

Turning next to direct-bandgap semiconductors, Nicholas J Ekins-Daukes outlines recent developments in III-V solar cells. III-Vs give the highest efficiencies of any solar cell materials. But despite their large absorption coefficients for above-bandgap light, the materials are relatively expensive, and often difficult and rare to extract from the earth's crust. Their place in the technology is assessed, together with recent advances.

Chalcogenide thin-film solar cells are next reviewed by Miriam Paire, Sebastian Delbois, Julien Vidal, Nagar Naghavi and Jean-Francois Guillemoles. Cu(In Ga)Se_2 or CIGS cells have made impressive progress in recent years with the highest efficiencies for thin-film cells, while Cu_2ZnSn $(S,Se)_4$ or CZTS or kesterite uses less-rare elements than CIGS, and so has significant potential for large-scale production.

The field of organic photovoltaics (OPV) has become of great interest since the efficiency achieved rapidly increased from around 1% in 1999, to more than 10% in 2012 (Green 2013). The chapter by Claudia Hoth, Andrea Seemann, Roland Steim, Tayebeh Amin, Hamed Azimi and Christoph Brabec reviews this novel technology, concentrating on the state-of-the-art in realising a photovoltaic product.

Lastly, one of us (Gavin Conibeer) looks to the future, by outlining third-generation strategies that aim to provide high conversion efficiency of photon energy at low manufacturing cost. Approaches covered include multiple energy level cells (such as tandem cells and multiple exciton generation), modification of the solar spectrum (such as by down- and upconversion), and thermal approaches (such as thermophotovoltaics and hot-carrier cells). The emphasis in all these approaches is on efficiency, spectral robustness, and low-cost processes using abundant nontoxic materials. The book ends with some concluding remarks by the editors, looking to the future in this rapidly developing field.

Finally, no book in this very extensive field can claim to be complete. To explore the field further, readers are recommended to consult 'Thin Film Solar Cells' by Jef Poortmans

and Vladimir Arkipov (Wiley 2006), a companion volume in this Wiley Series on Materials for Electronic and Optoelectronic Applications, which includes such areas as dye-sensitised solar cells (DSSCs), in the chapter by Michael Gratzel. We hope that this book, with its emphasis on technological materials, will be of use to all who are interested in this field.

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Poortmans, J., and Arkipov, V., '*Thin Film Solar Cells*' Wiley, Chichester 2006.

Green, M.A., Emery, K, Hishikawa, Y., Warta, W., and Dunlop, E.D., Solar cell efficiency tables (version 41), Progress in Photovoltaics: *Research and Applications*, 21 (2013) p. 1–11.

2 Fundamental Physical Limits to Photovoltaic Conversion

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2.1 INTRODUCTION

Where to stop the quest for better devices? What does better mean? The conversion efficiency arises prominently in this respect.

More efficient devices, everything kept equal, would first translate into cheaper solar electricity. Are there limits to reducing the cost of PV electricity? In 2012, modules were sold 0.5–0.7 €/W and the cost of solar electricity is around 20 cts/kWh. In the longer term, development of photovoltaics (PV) has to be based on a major technological breakthrough regarding the use of processes and materials at very low cost, or/and on the engineering of devices offering far higher performance, harvesting most of the available solar energy. Two approaches are targeted at this issue today: the first aims at low-cost materials and low-cost processes to reduce the surface cost of PV devices, possibly sacrificing some of the device efficiency, and the second, aiming at the maximal possible efficiency, at the same cost as today's modules (see Figure 2.1). There is a major difference between these two approaches: the conversion concepts, the materials and the processes.

If we think in terms of the manufacturing costs of PV modules, the target aimed at requires that the system needs to produce 1 MWh (comprising about 0.2 m^2 of high-end c-Si modules lasting 25 yr) cost less than €30 for parity with the base load or €120 for grid parity. For a very low-cost device, for instance based on polymers or organic–inorganic hybrids, with an expectation for conversion efficiencies on a par with those achieved by the amorphous Si line (on the grounds of similar structural disorder and a low carrier mobility) and shorter life durations, the budget is €7.5/m^2 (5-year life duration with 5% efficiency, including power electronics and installation), closer to the cost of structural materials than of functional electronic materials. Finally, for profitable electricity production, we need to pay attention to the system costs. Thus, one sees that it might be extremely difficult to attempt to reduce production costs far beyond what is currently being obtained with inorganic thin-film systems.

This chapter will deal with the scientific issues behind the photovoltaic conversion process, keeping in mind what would make a difference to having this technology more widely used.

The first of these questions is of course the efficiency of the processes. Since the appearance of the first PV devices, the question of the conversion efficiency limits arose, and for a good reason: not only does it have high scientific and technological visibility, it is also

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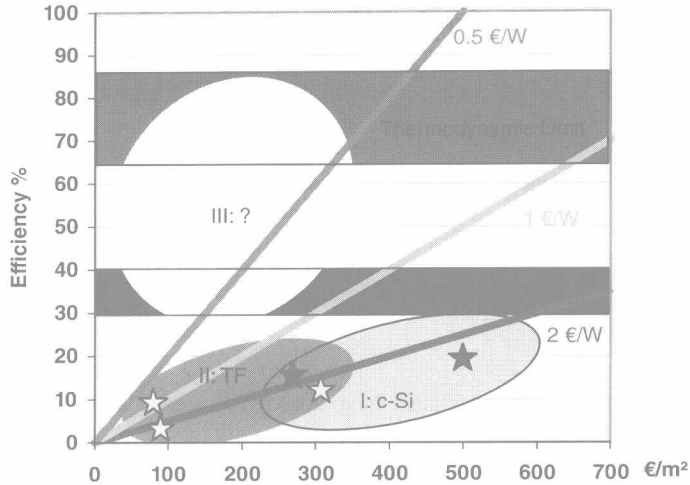

Figure 2.1 Relation of the cost per watt of solar energy to the surface cost of manufacturing solar devices (modules) and the device efficiency. The light gray, dark gray, and white oval regions represent the ranges found for crystalline silicon (first-), thin-film (second-), and third-generation solar technologies, respectively. The white zone marks the anticipated range for very high efficiency devices. For comparison, limiting efficiencies derived from thermodynamic constraints are also indicated as horizontal bands (low range: no concentration, high range: maximal concentration). Stars indicate industrial production costs as they could be estimated in 2010 from available data: filled stars for c-Si modules and hollow stars for thin-film technologies. (Adapted with permission from Green, 2003. Copyright © 2003, Springer.)

one of the major factors in lowering the cost of generating solar electricity. Interestingly, this question of efficiency limit took quite a bit of time before being settled [Landsberg and Badescu, 1998].

The paper of Schockley and Queisser, devising an approach based on a detailed balance approach of photovoltaic conversion is still one of the most quoted papers on PV, yielding the limit of single-junction, standard PV devices.

This question has also been approached on a more general basis, using thermodynamics (Landsberg and Tonge, 1980, Parrott 1992, De Vos 1992) to give device-independent or even process-independent limits (Section 2.1). These limits are essentially related to the source (the sun) characteristics and to the conditions of use (e.g. ambient temperature). Perhaps more useful, and practical, limits have been proposed for defined processes.

In very general terms, photovoltaic conversion in its simpler form supposes several steps:

1. solar photon transferred to the active part of the system;
2. absorption of the photons and energy transfer to the electronic system;
3. selective extraction of electrons to contacts (2 at least);
4. channelling of e-free energy to useful load whose impedance is adjusted.

These steps are illustrated in Figure 2.2 and describe PV process as it is working in all working devices, with nonessential modifications for organic PV (in which electron and holes are coupled as excitons) and multijunction cells (where the incident spectrum on a cell is a part of the total solar spectrum).

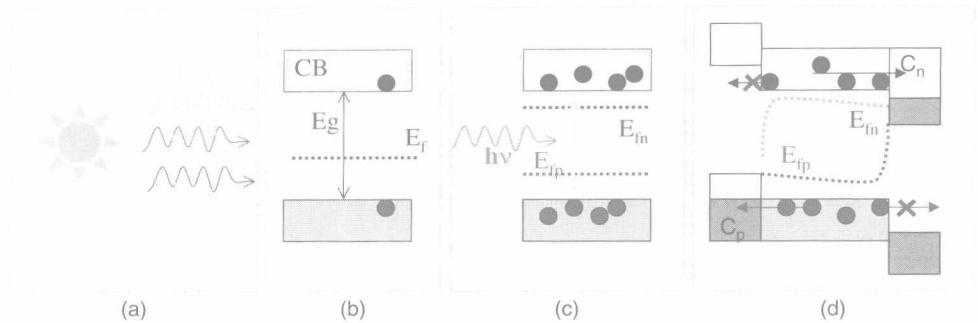

Figure 2.2 Photovoltaic action proceeds in 3 steps. (a) photons have to be collected and coupled to the converting system; (b) the converting systems contains occupied (VB) and empty states (CB) separated by a gap between which light induces transitions are allowed; (c) upon photon absorption, two populations of charge carriers are created; (d).

In this process, the work per absorbed photon is equal to the electromotive force between the electrodes (i.e. the voltage) times the elementary charge, that is the difference in the quasi-Fermi levels in the two contacts. In the absence of current extraction and when the mobility is high enough this quasi Fermi level (QFL) separation is that of electrons and holes in the absorber (see Section 2.4 for a more complete discussion).

Because the QFL is generated by the incoming flux, it increases with the light flux. This can also be understood as a larger generation rate per unit volume will create a larger density of electron–holes pairs, and therefore a higher conduction-band electron QFL and a lower valence-band electron QFL (that is a larger chemical potential of holes in the VB). This large QFL separation can be obtained in different ways, everything else being equal:

- by decreasing the recombination pathways for photogenerated carriers, for instance increasing the carrier lifetime;
- by increasing the generation of electron–hole pairs, for instance by concentration of the incident solar influx;
- by decreasing the generation volume, for instance by thinning down the cells, which requires light trapping to keep the total generation constant.

A large chemical potential can be seen as a large partial pressure: this helps the extraction of generated carriers and therefore a larger free energy per carrier can be collected, whereas, whatever the concentration of electron–hole pairs, their potential energy is always the same, near E_g. The collection of carriers depends on the chemical potential of the carriers in the contacts, that is, *in fine*, of the external conditions, and for instance the load into which the solar cell will deliver power.

The maximum power is delivered when the load impedance matches the differential impedance of the generator (as is true by the way for any generator).

Indeed, if the device has a current–voltage characteristic $I(V)$, the power $P = I \cdot V$ is maximum for

$$\frac{\mathrm{d}P}{\mathrm{d}V} = 0 \text{ and, as } \frac{\mathrm{d}P}{\mathrm{d}V} = I + \frac{\mathrm{d}I}{\mathrm{d}V} \cdot V \tag{2.1}$$

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