

Chapter 1

Introduction

1.1 Energy Supply and Climate Change

Photovoltaics and in particular organic photovoltaics are fields of applied research whose support by the society is based on the insight that this technology can significantly contribute to worldwide electricity generation by conversion of solar energy into electrical energy. Apart from wind energy, photovoltaic energy conversion is the most promising among the emerging “renewable” energy technologies due to its large theoretical potential, which is given by the solar energy flux hitting the earth surface. This potential is estimated to be 6,000 times larger than the global annual energy consumption of $\approx 12,000$ million tons of oil equivalent (≈ 15 TW mean power) in 2010. Note that the global energy demand roughly doubled within the last thirty years and keeps growing as shown in Fig. 1.1a.

The technical potential of solar energy conversion is very large as well: A rough estimation demonstrates that an area of less than 6 % of the Sahara¹ would be enough to satisfy the world energy demand with today’s technologies. However, this estimation neglects distribution and storage issues. Currently, energy is provided to a large extent (> 80 %) by fossil fuels due to economic and traditional technical reasons [2].

Mainly two issues arise from fossil-powered energy conversion. First, fossil resources are finite and their distribution on the earth surface is very unbalanced. Second, burning fossil fuels is accompanied by carbon dioxide (CO_2) emission which results in climate change, because CO_2 acts as a greenhouse gas. The greenhouse effect is caused by the atmosphere reflecting (infrared) heat radiation from the surface of the earth back to it. This effect is essential for the development of life on earth, because the radiation balance between the sun, atmosphere, and the earth surface yields a mean earth’s surface temperature of $\approx 14^\circ\text{C}$ which would be -15°C [3] without the greenhouse effect. The temperature was very stable in the last millennia and the global temperature distribution created climate zones with their characteristic

¹ Assumptions: area $9 \times 10^6 \text{ km}^2$, mean irradiation 285 W/m^2 [1], power-conversion efficiency 10 %.

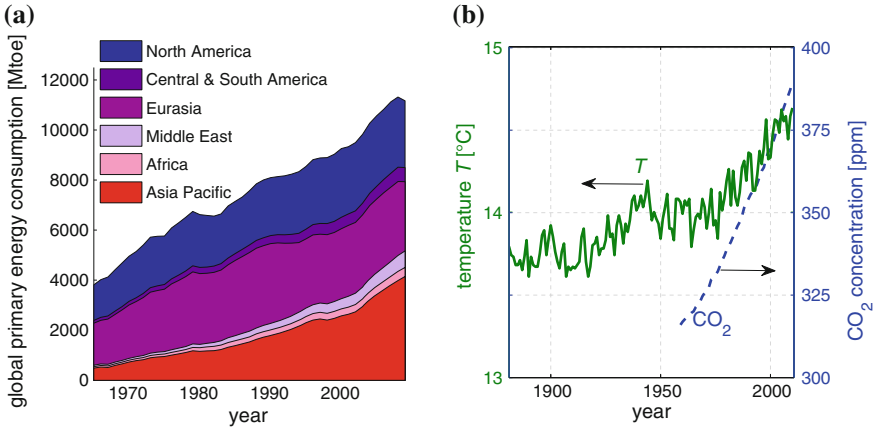


Fig. 1.1 **a** Development of the regional distribution of primary energy demand in million tons of oil equivalent (1 Mtoe \approx 42 PJ). *Source* BP [5]. **b** Annual mean CO_2 concentration in the atmosphere from measurements at the Mauna Loa Observatory, Hawaii [6]. In pre-industrial times the CO_2 concentration was 280 ppm [4]. The development of the mean global temperature in the last century is plotted as well [7]. Both curves describe a significant increase

flora and fauna, which are very sensitive to temperature. An increased greenhouse gas concentration in the atmosphere disturbs this equilibrium and leads to a higher global temperature, which has been observed within the past decades (Fig. 1.1b). In recent years it became broad consensus in science and politics that this rise in temperature is anthropogenic and correlates with the amount of CO_2 in the atmosphere, which has increased significantly compared to pre-industrial times (Fig. 1.1b) [4].

The impacts of global warming are very severe and potential consequences are a rise in sea level due to a thermal expansion of ocean water and the melting of on-shore ice shields. Furthermore, desertification and water shortages are becoming more likely and even collapses of whole ecosystems and changes of ocean currents being essential for the global balance of living nature are expected. The last point represents one of the many non-linear effects which are hard to predict, but contain a high risk of a rapid change. Although nature is not a static system, but continuously in change, development, and assimilation, the survival of a certain species is in danger upon a fast change of the environment. How fast, to what extent, and at which sacrifice modern man is capable to adapt is unknown. On the one hand, besides biological adaption, he has technological and medical means to support him. On the other hand, he is very vulnerable. His economic wealth and the survival of societies and dense populations heavily rely on technological large-scale food production and globalized markets.

Based on the elaborations of several researchers, summarized in IPCC reports [4], the issues of global change are continuously discussed by policy makers on international summits on climate change. In 1992 during the Earth Summit in Rio de Janeiro, the United Nations Framework Convention on Climate Change (UNFCCC) was initiated with the aim of reducing the impacts of global warming. On a succeeding summit the Kyoto Protocol (1997) was adopted which contains reduction targets

for CO₂ emissions. However, it is not ratified on a global scale. Also the common 2-degree-target was only recently officially agreed on in Cancun (2010). This target demands for a maximum global mean temperature rise of 2 °C compared to pre-industrial times. Two degrees is a number that was chosen because the consequences of a rise of mean earth surface temperature by this value are supposed to be in a manageable range. For a higher increase in temperature non-linear effects become more likely.

To fulfill this 2 °C target (which is not very likely given the current trajectory) the energy sector has to be decarbonized completely within the next three decades. Renewable energies are the most sophisticated and most developed kind of technology which can contribute to a quick decarbonization. The main obstacle are their electricity-generation costs which are currently higher than those of conventional energy technologies. This is, however, to a large extent due to the fact that the external costs of the conventional technologies are not internalized. Consequently, the long-term damages caused by these technologies have to be carried by the broad world population.

This cost problem was recognized by policy makers. Here, Germany is mentioned as an example, as it was one of the first countries with a legislation that actively supported renewable energies. In 2000 the German parliament initiated a feed-in tariff, where in their electricity bill all customers support the introduction of renewable energies. In the meantime many other countries followed this model. On the one hand, supporting photovoltaics in Germany is a very expensive way of reducing carbon-dioxide emissions, and the feed-in tariff has shown the problem of over subsidizing and the risk of favoring non-economic concepts also on the long run. On the other hand, this concept was very successful in bringing costs down, which significantly reduced the time until photovoltaics will be an economic and CO₂-saving way of electricity generation in southern countries. It lead to installed capacities of 31 GW (wind) [8] and 32 GW (photovoltaics) [9] at a total installed electrical capacity of ≈170 GW [10] in Germany in 2012. These data are nominal capacities, which represent peak values (W_p) in the case of the renewable technologies. They show that in Germany solar and wind energy can already now provide from 0 to 50 % of the instantaneous power² dependent on the weather, season, and demand. It is obvious that grid extensions and/or an increase in the electricity-storage capacity will become very important for a further extension of renewable energy technologies. As these measures introduce additional costs and drops in the overall energy-conversion efficiency, they require very cost-effective solar and wind energy harvesting units.

In 2012 renewable energy technologies had a share of 26 % (5.3 % photovoltaics) in Germany's electricity generation [9]. The costs, especially of photovoltaics, have decreased significantly, following a learning curve due to economy of scale and technological improvements. Today, photovoltaic electricity generation is already economical in off-grid systems and attractive for self-consumption in southern countries (grid parity) [12]. Consequently, large power plants are built all over the world, with

² In Germany the typical Sunday peak load around noon is approximately 60 GW [11].

the first ones operating economically without subsidies. Therefore, the cumulated, globally installed capacity rose to 100 GW at the beginning of 2013 [9].

1.2 Development of (Organic) Photovoltaics

The first silicon solar cell with an efficiency of around 4 % was invented in the Bell Laboratories in 1953, six years after the discovery of the p-n junction by William B. Shockley, Walter H. Brattain, and John Bardeen, who were awarded by the Nobel price in physics for the discovery of the transistor effect in 1956. Five years after the invention of the silicon solar cell, the first solar module was used in space. The term “module” describes a set of solar cells that are electrically connected and packaged. In the 1960s commercial modules were available for terrestrial use and showed cell efficiencies of 14 %. At the end of the seventies an annual production of 500 kW_p was reached. The modules powered remote telecommunication systems (e.g. in the Australian Outback) with an estimated cost of 100 EUR/W_p. Prices decreased due to incentives during the oil crisis in the seventies, the German 1000 and 100,000 “Dächer (roofs) Programm” in the 1990s, and the feed-in tariff starting in the year 2000. The prices are still decreasing, e.g. on the German market from 5 EUR/W_p in 2006 to 1.7 EUR/W_p in 2013 [13] (system price for roof top installation). This drop in price together with an increase of module efficiency beyond 15 % (cell efficiency beyond 20 %) made the tremendous growth of the installed capacity possible and maintained a high market share of silicon solar cells (80–90 %).

The working principle of every solar cell is based on the (inner) photoelectric (“photovoltaic”) effect, first discovered by the physicist Alexandre E. Becquerel (1839) with electrolytic cells [14]. Photoconductivity was shown for selenium by Smith [15] in 1873, and the outer photoelectric effect was systematically investigated by Hertz and Hallwachs in 1886 [16]. In 1904 the physicist and Nobel Prize laureate Lenard [17] discovered the role of the frequency of light regarding the energy of the emitted electrons. His results were theoretically explained by Einstein [18] who was awarded by the Nobel Prize for this work in 1921. The photoelectric effect is very interesting from a technological point of view, as it allows for the most direct conversion of sun light into electricity. This process simply requires a planar solid-state device (i.e. solar module) aligned to the sun. A solar module does not contain any chemical liquids or mechanical wear parts like conventional generators with rotating elements. Therefore, long-term stability and low maintenance costs are inherent and big advantages of solar modules compared to conventional means of electricity generation.

From the findings at the end of the nineteenth century it was still a long way to solar cells based on silicon (1953), which is nowadays the most common industrial semiconductor material for several kinds of applications. In the meantime, a variety of materials was investigated and electronically characterized. Amongst those were organic materials, which are based on hydrocarbon molecules. First investigations of the electronic properties of organic molecular solids were reported for anthracene at

the beginning of the 20th century [19–21]. In the 1970s (semi)conducting polymers were discovered [22]. Allan J. Heeger, Alan G. MacDiarmid, and Hideki Shirakawa received the Nobel Prize in chemistry in 2000 for the discovery and development of these conductive polymers. In the 1980s high electroluminescence efficiencies were reached in organic materials. This achievement led to the development of organic light emitting diodes (OLEDs) [23, 24] as first broad application of organic electronics. Today, OLEDs can be found in efficient flat panel displays [25] and are about to become an alternative large-area light source with a high efficiency and a pleasant irradiation spectrum [26].

The first photocurrent in an organic semiconductor was observed also in anthracene by Kalman and Pope in 1959 [27]. In the following two decades, several organic photovoltaic devices were developed. They consisted of a metal-organic junction which showed efficiencies of less than 0.1 % (for a contemporary review, see [28]). The first major breakthrough in the deployment of organic semiconductors in solar cells has been made by Ching Tang who developed the donor-acceptor solar cell and reported an efficiency of 1 % [29] in 1986. Tang's cell comprised a junction of two materials, one electron and the other hole accepting and transporting (a perylene tetracarboxylic derivative and copper phthalocyanine). The second major step was the invention of the bulk heterojunction which is a mixed layer of donor and acceptor, fabricated by a co-deposition of the two molecules. This approach was firstly reported in [30, 31]. A detailed overview of the development of organic photovoltaics can be found in [32].

After these achievements the amount of publications rose nearly exponentially in the last decade. Efficiencies recently reached 12 % (Fig. 1.2) [33, 34], also pushed by several spin-offs and established companies turning focus on this topic. The reason for this boom is found in the expected high potential of organic semiconductors, which

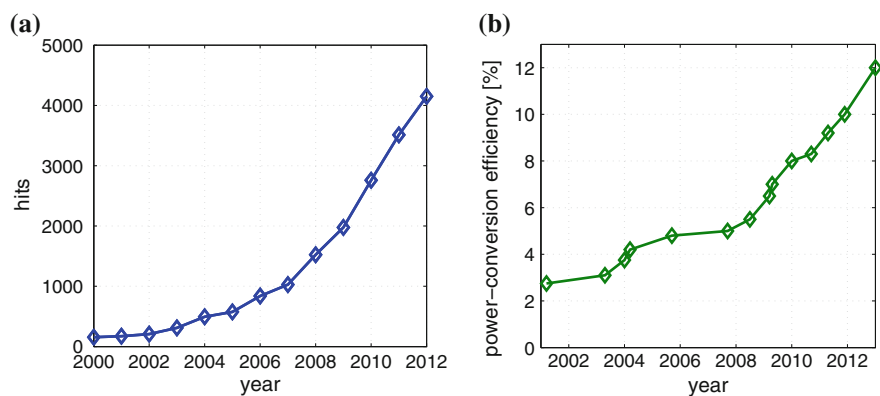


Fig. 1.2 **a** Hits of a search in the database “Web of Science” regarding organic photovoltaics (search string: Topic = (“organic photovoltaic” OR “organic photovoltaics” OR “organic solar cell” OR “organic solar cells” OR “polymer solar cell” OR “polymer solar cells”). Search performed on 26.08.2013). **b** Development of the maximum power-conversion efficiency for organic solar cells on the laboratory scale during the last decade

are either (vacuum or solution processed) small molecules or (casted or printed) polymers.

The main advantages of organic solar cells are:

- Potentially cheap production by high-throughput roll-to-roll printing or other low-temperature deposition techniques
- Low energy payback times due to a fabrication process avoiding expensive purification methods or energy-intensive steps
- High versatility and efficiencies due to the toolbox of organic chemistry
- High energy yield due to a good low-light performance and a positive temperature coefficient of the power-conversion efficiency
- Non-toxicity and low consumption of abundant absorber materials (a few grams per square meter)
- New products containing and merging with photovoltaics, e.g. in architecture due to a tunability of color and (semi-)transparency
- Application in novel and mobile devices (e.g. consumer electronics) due to mechanical flexibility and low weight (Fig. 1.3)

One major challenge from the economic point of view is a realization of these potential properties within the next few years. Here, the main difficulty is the competition with established technologies. As already mentioned, crystalline silicon photovoltaics showed a tremendous reduction of production costs in the last decade. Furthermore, the developments in inorganic thin-film photovoltaics reveal the potential of these technologies to cover several of the mentioned advantages of organic photovoltaics as well, e.g. mechanical flexibility. Regarding organic solar cells, first

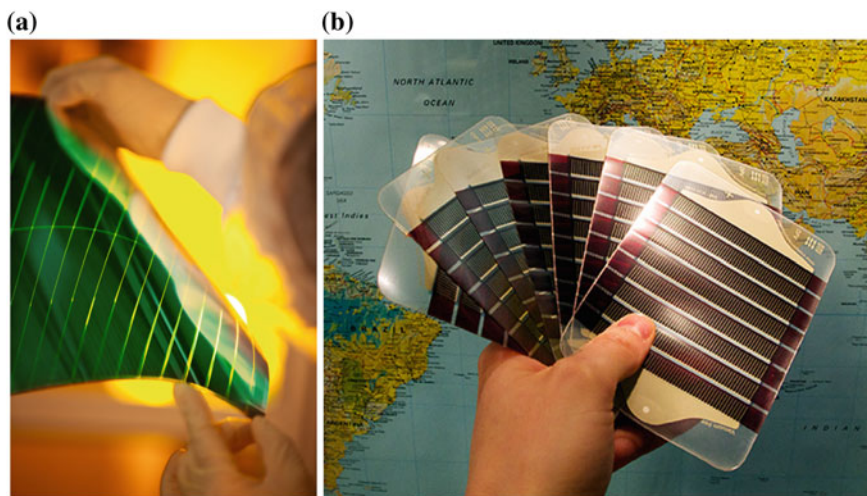


Fig. 1.3 **a** Example of a flexible solar foil (picture Heliatek GmbH). **b** Demonstrator modules with an efficiency of 1.8 % currently shipped worldwide [35] (picture DTU Energy Conversion)

pilot lines are installed in order to demonstrate a feasible transition from product innovation to process innovation. Currently, printed demonstrator modules are shipped worldwide for free (Fig. 1.3b) [35]. Product innovation and development are still crucial as an increase in solar-cell efficiencies beyond 12–15 % will enhance the ability of organic photovoltaics to compete with the established technologies. However, process development is very important at the current stage. It is required to verify that the performance and the lifetime, which have already been achieved, can be transferred to large-scale devices with ≈ 5 % efficiency at the anticipated costs. On the long run and on the terawatt scale, the criteria of material abundance favor mainly two technologies, namely silicon and carbon-based photovoltaics [36]. The success of organic photovoltaics depends strongly on the current investments, making this technology capable of finding its market a side of silicon and possibly in the future even in competition.

This book focuses on the research and development of organic solar cells, as the physics of these devices is far from being completely understood. However, a further improvement of efficiencies and stability, increasingly demands for a detailed understanding of the processes limiting the device performance. Whereas in the first years, research was mainly based on trial-and-error experiments and simple material screening, in recent years modeling and simulations became more important (e.g. large-scale computational screening projects [37]). On the one hand, many results obtained from microscopic theories are far from experimental results. On the other hand, theoretical considerations in particular on the device level considerably improved the understanding of the working principle of the complete solar cell. That is why the working principle of organic solar cells is the topic of the prevailing book, giving insights into theory, simulations, and experiments.

1.3 Scope and Structure of This Book

The scope of this book is to give the interested reader a comprehensive insight into the device physics of organic solar cells. It is an attempt to bridge the gap between experimentalists and theoreticians or computational physicists. It is suited for readers with background in physics, electrical engineering, materials science, and chemistry. The book covers a broad spectrum from fundamental theory to applied methods and technology to the latest results in the research field of organic photovoltaics. Many topics go beyond organic solar cells and are, therefore, interesting for readers working with different kinds of solar cells. The strategy of the book is to repeatedly pick up important characteristics shifting the perspective. Therefore, many topics once introduced based on very fundamental theories are revisited in the context of organic solar cells and subsequently applied to explain experimental data in a further chapter.

This book focuses on teaching on organic solar cells. Its scope is not to provide a complete review on the field of organic photovoltaics. That is why in particular

the bibliography is not intended to be an exhaustive list of the major publications on organic solar cells.

The chapters of this book are grouped in three main parts I–III distinguishing the main scope of the chapters belonging to a respective part. Part I containing Chaps. 2–5 gives a broad introduction to the topic of organic solar cells in a textbook like format. It is well suited as starting point for students who are about to work in the field of organic solar cells. It presupposes some basic knowledge in solid state physics and describes the general ideas of solar thermal energy conversion independent of the chosen material system. In some cases intuitive explanations are preferred to rigorous mathematical derivations for the sake of comprehension. However, the major physics relevant for solar cells is introduced in detail acquainting the reader with the basic concepts. The main focus is the working principle of solar-cell devices. It is elaborated starting from general considerations of solar-thermal energy conversion and semiconductor physics in the first chapter. Here, the practically oriented reader can skip the detailed derivations based on thermodynamics. Important are the main principles, which will be applied to organic solar cells in the subsequent chapter. There, the basics of organic chemistry are briefly reviewed in an intuitive way to provide the reader with the background necessary to understand the semiconductive properties of organic molecules.

Chapter 3 continues with the working principle of donor-acceptor heterojunction solar cells, focusing on the open-circuit voltage, the photocurrent, metal-organic interfaces, and the charge-transfer state. In particular, ongoing discussions on the latter demonstrate that the understanding of photocurrent generation in organic materials is not completely settled. Hence, some parts of this chapter give a review on the most recent theoretical and experimental approaches for characterizing the photophysics of organic solar cells without giving a final conclusion. The chapter closes with more practical aspects regarding materials and the experimental realization and characterization of organic solar cells.

In Chap. 4 an electrical model based on drift-diffusion and an optical model based on the transfer-matrix approach are presented. The content covers several descriptions of charge carrier transport and recombination. One part of the chapter focuses on a possible realization of a drift-diffusion simulation. This part is dedicated to computational physicists and might be skipped by other readers who may wish to go back to it if necessary when digesting explanations based on these simulations in subsequent chapters.

The last chapter (5) of Part I contains a detailed theoretical study based on drift-diffusion simulations. Showing the interplay between properties of the absorber and the contacts, it provides explanations on major processes and parameters influencing photovoltaic energy conversion.

Part II (Chaps. 6–8) focuses on systematic experimental studies which have been performed mainly during the PhD work of the author. Each chapter of this part is organized in a way that the reader can follow it without having read all previous chapters. Although elaborated on experimental and simulation data of selected material systems, most of the conclusions of this part are of general nature. Thus, they may be helpful for scientists who are dealing with the interpretation of current-voltage

data of (organic) solar cells in general. The first chapters (6–7) of this part contain discussions on the origin and limiting processes regarding the open-circuit voltage and give explanations for distorted J - V curves which follow S-shaped characteristics. The last chapter (8) contains detailed investigations on a selected model system, which is the most common small-molecule solar-cell system zinc phthalocyanine (ZnPc)—fullerene C₆₀. In particular, the effect of the mixing ratio, a gradient in the active layer, the role of the absorption profile, and dominating recombination mechanisms are discussed. The results provide a detailed picture of the interplay between energy levels, charge carrier mobilities, and recombination mechanisms in organic solar cells in general.

Part III (Chaps. 9 and 10) contains a summary and wrap-up of the general conclusions drawn in the second part. This summary is arranged in a catalog of guidelines on how to interpret experimental current-voltage curves from the drift-diffusion point of view. A variety of schematic drawings based on simulation results visualize the influence of several material parameters and physical mechanisms on the current-voltage characteristics of solar cells. The last chapter gives an outlook with a survey on the major challenges of organic solar cells regarding efficiency, stability, ecology, and economics.

Each chapter begins with a brief abstract containing a list of questions. The intended purpose of these questions is twofold: First, they provide a table of contents, which is oriented at problems. Therefore, the reader gets a quick idea which problems are addressed in the prevailing chapter. Second, they serve as means of control to the reader allowing him or her to check whether he or she has grasped the main messages of the chapter. That is why short versions of the answers are provided at the end of each chapter including a list of literature as suggestion for further reading.

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