Review

Hybrid solar cells

Serap Günes [a,b], Niyazi Serdar Sariciftci [a,*]

[a] Linz Institute for Organic Solar Cells (LIOS), Johannes Kepler University Linz, Physical Chemistry, Altenberger Strasse 69, A-4040, Linz, Austria
[b] Yildiz Technical University, Faculty of Arts and Science, Department of Physics, Davutpasa Campus, 34220 Davutpasa, Esenler, Istanbul, Turkey

Dedicated to Professor Michael Grätzel

Abstract

Historically, conventional solar cells were built from inorganic materials such as silicon. Although the efficiency of such conventional solar cells is high, very expensive materials and energy intensive processing techniques are required.

Hybrid and photoelectrochemical (dye sensitized) solar cells have been the cheap alternatives for conventional silicon solar cells. A hybrid solar cell consists of a combination of both organic and inorganic materials therefore, combines the unique properties of inorganic semiconductors with the film forming properties of the conjugated polymers. Organic materials are inexpensive, easily processable and their functionality can be tailored by molecular design and chemical synthesis. On the other hand, inorganic semiconductors can be manufactured as nanoparticles and inorganic semiconductor nanoparticles offer the advantage of having high absorption coefficients and size tunability. By varying the size of the nanoparticles the bandgap can be tuned therefore the absorption range can be tailored.

In this short review, we will focus on the concepts of organic/inorganic "hybrid" solar cells.

Serap Günes *was born in Istanbul, Turkey, in 1976. She obtained her Ph.D. at the Johannes Kepler University Linz in 2006 under the direction of Professor Niyazi Serdar Sariciftci. She moved to Yildiz Technical University, Istanbul/Turkey in September 2006 and serves as an Assistant Professor. Besides hybrid solar cells her research interests include microscopy techniques such as atomic force microscopy (AFM), near field scanning optical microscope (NSOM), and photovoltaic characterization of organic solar cells and also organic field effect transistors. She has been awarded "Turkish Physical Society Prof. Sevket Erk Young Scientist" prize in 2007.*

Niyazi Serdar Sariciftci *was selected as Fellow of the Royal Society of Chemistry (Great Britain), FRSC in 2005. In September 2003, he was elected to City Council of Linz (Gemeinderat der Landeshauptstadt Linz), GR. He became the Founding Director (Vorstand) of the Linzer Institut für organische Solarzellen (LIOS) at the Johannes Kepler University of Linz/Austria, in 2000. In 1996, he was Ordinarius Professor (Chair) and Director (Vorstand) of the Institute for Physical Chemistry at the Johannes Kepler University in Linz/Austria, and in 1992, was habilitation for the academic title of "docent" by the Central Interuniversity Commission in Ankara/Turkey. From 1992 to 1996 N.S. Sariciftci was a senior research associate at the Institute*

* Corresponding author. Tel.: +43 732 2468 8753; fax: +43 732 2468 8770.
  E-mail address: Serdar.Sariciftci@jku.at (N.S. Sariciftci).

for Polymers & Organic Solids at the University of California, Santa Barbara (Prof. Alan J. HEEGER, Nobel leaurate 2000 for Chemistry) in the fields of photoinduced optical, magnetic resonance and transport phenomena in semiconducting and metallic polymers and during 1989 – 1991, was a postdoctoral research associate at the 2nd. Physics Institute, of University of Stuttgart, Fed. Rep. of Germany, in the project "Molecular Electronics" SFB 329 of "Deutsche Forschungsgemeinschaft", c/o Prof. Michael MEHRING, with specialization on electron spin resonance (ESR), electron nuclear double resonance (ENDOR) and photoinduced electron transfer on supramolecular structures. He completed his PhD study at the University of Vienna, Austria with promotion for Doctor of Science in physics in 1989. In 1986, he studied Physics at the University of Vienna, Austria with sponsion for Master of Science in physics. He has to his credit over 350 scientific publications, 5 books and 8 patents. He is a member of the following societies: Fellow of the Royal Society of Chemistry, Member of the American Chemical Society, Member of the Materials Research Society, Member of the Austrian Chemical Society, Member of the SPIE.

He has been awarded the Turkish National Science Prize 2006 (TUBITAK Bilim Ödülü), the "Energy Globe Oberösterreich" prize sponsored by the Oberösterreichische Nachrichten (October 2003) and the "Grünpreis 2001" sponsored by the Austrian Green Party, Upper Austria, which is given to innovative research in environmental protection (December 2001). He won a distinction prize with record average (9.92 out of 10) in Austrian Sankt Georg Kolleg graduation in 1980. In connection with this achivement, award of Ministery of Science and Technology in Austria with a full coverage fellowship for a whole University Study in Austria. He has also directly advised around 47 graduate students and postdoctoral assistants and has been a principal investigator in many projects. He was the founder of High Tech SPIN-OFF Companies: Konarka Austria (formerly QSEL created in 1998), engaged in plastic solar cells technology, www.konarka.com, NanoIdent AG, created in 2002 engaged in plastic photodetector and bio-photodetector technologies, www.nanoident.com and Plastic Electronic GesmbH, created in 2006, engaged in plastic analog and digital circuits www.plastic-electronic.com.

Contents

1. Introduction

The conversion of sunlight into electricity is a clean, abundant and renewable energy source. The efficiency of conventional solar cells made from inorganic materials reached up to 24% [1], using very expensive materials of high purity and energy intensive processing techniques. New ways of manufacturing solar cells that can scale up to large volumes and low cost are required. A broad range of solar cell technologies are currently being developed, including dye-sensitized nanocrystalline photoelectrochemical solar cells, polymer/fullerene bulk heterojunctions, small molecule thin films and organic–inorganic hybrid devices.

In this short review, we will mainly focus on the concepts of organic/inorganic "hybrid" solar cells. This review is organized as follows: First we will give a short description of photoelectrochemical solar cells using nanoporous TiO_2 electrodes. Next, we will discuss critical parameters in solid state dye-sensitized solar cells. Further, we will focus on p-type semiconductors as hole transporters. Then, we will explain quasi solid state dye-sensitized solar cells and nanoparticle-quantum dot sensitized solar cells and finally the conclusion.

1.1. Electrical characteristics of a solar cell

The current–voltage characteristics of a solar cell in the dark and under illumination are shown in Fig. 1. In the dark, there is almost no current flowing, until the contacts
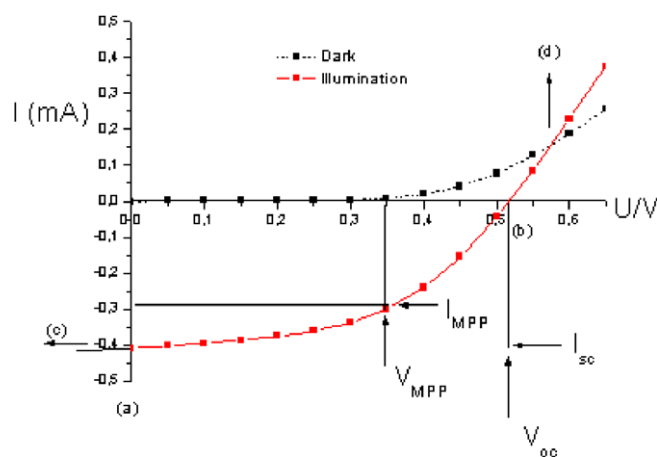

Fig. 1. Current–voltage (I–V) curves of an organic solar cell (dark, dashed; illuminated, full line). The characteristic intersections with the abscissa and ordinate are the open circuit voltage (V_{oc}) and the short-circuit current (I_{sc}), respectively. The largest power output (P_{max}) is determined by the point where the product of voltage and current is maximized. Division of P_{max} by the product of I_{sc} and V_{oc} yields the fill factor FF.

start to inject heavily at forward bias for voltages larger than the open circuit voltage. Under light illumination, (a) at short-circuit current condition the maximum generated photocurrent flows and (b) at flat band condition the photogenerated current is balanced to zero. In the fourth quandrant (between (a) and (b)) the device generates

power. At maximum power point (MPP), the product of current and voltage is the largest [2].

The photovoltaic power conversion efficiency of a solar cell is determined by:

$$\eta_e = \frac{V_{oc} \times I_{sc} \times FF}{P_{in}}$$

$$FF = \frac{I_{mpp} \times V_{mpp}}{I_{sc} \times V_{oc}}$$

where V_{oc} is the open circuit voltage, I_{sc} is the short-circuit current, FF is the fill factor and P_{in} is the incident light power density, which is standardized at 1000 W/m^2 for solar cell testing with a spectral intensity distribution matching that of the sun on the earth's surface at an incident angle of 48.2°, which is called the AM 1.5 spectrum [3]. I_{mpp} and V_{mpp} are the current and voltage at the maximum power point in the fourth quadrant of the current–voltage characteristics.

2. Hybrid solar cells

Hybrid solar cells are a mix of nanostructures of both organic and inorganic materials. Therefore, they combine the unique properties of inorganic semiconductor nanoparticles with properties of organic/polymeric materials [4]. In addition to this, low cost synthesis, processability and versatile manufacturing of thin film devices make them attractive [5,6]. Also, inorganic semiconductor nanoparticles may have high absorption coefficients and particle size induced tunability of the optical band-gap. Thus, the organic/inorganic hybrid concept for photovoltaic solar cells is getting interesting and attractive in recent years.

In the literature, hybrid solar cells are manufactured using different concepts such as solid state dye-sensitized solar cells [7–10] and hybrid solar cells using the bulk heterojunction concept with different nanoparticles such as TiO$_x$ [11], ZnO [12], CdSe [13,14], CdS [15], PbS [16], and CuInS$_2$ [17,18].

A dye-sensitized solar cell of Graetzel type comprises of several different materials such as nanoporous TiO$_2$ electrodes, organic or inorganic dyes, inorganic salts and metallic catalysts [19–21]. After absorption of a photon, the excited electron within the sensitizer molecule is transferred to the conduction band of TiO$_2$, and diffuses through the porous TiO$_x$ network to the contact. The oxidized sensitizer molecule is reduced to the original state by supply of electrons through a liquid electrolyte redox couple within the pores (see Fig. 2) [21,87].

This photovoltaic conversion system is based on light harvesting by a molecular absorber attached to a wide band-gap semiconductor surface [20]. A monolayer of dye on a flat surface can only harvest a negligibly small fraction of incoming light. In this case it is useful to enlarge the interface between the semiconductor oxide and the dye. This is achieved by introducing a nanoparticle based electrode construction which enhances the photoactive interface by orders of magnitude [21]. The dye sensitization
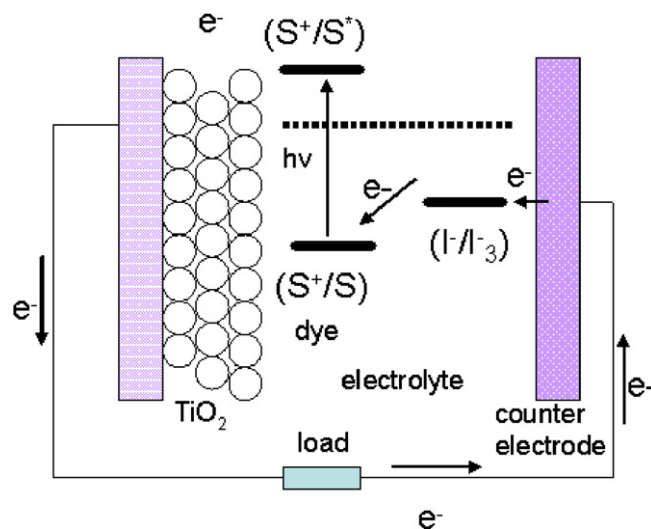

Fig. 2. Operation principle of a dye-sensitized solar cell.

of the large band-gap semiconductor electrodes is achieved by covering the internal surfaces of porous TiO$_2$ electrode with special dye molecules which absorb the incoming photons [22]. Sensitization effect can be seen in Fig. 3 [21] as a shift of the "incident photon to current efficiency (IPCE)" to higher wavelengths when coated with the dye [22].

The ideal sensitizer dye for a single junction solar cell converting global AM 1.5 sunlight to electricity should attach to the semiconductor oxide surface, absorb all light below a threshold wavelength and inject photoexcited electrons into the conduction band of the oxide [20,21,23].
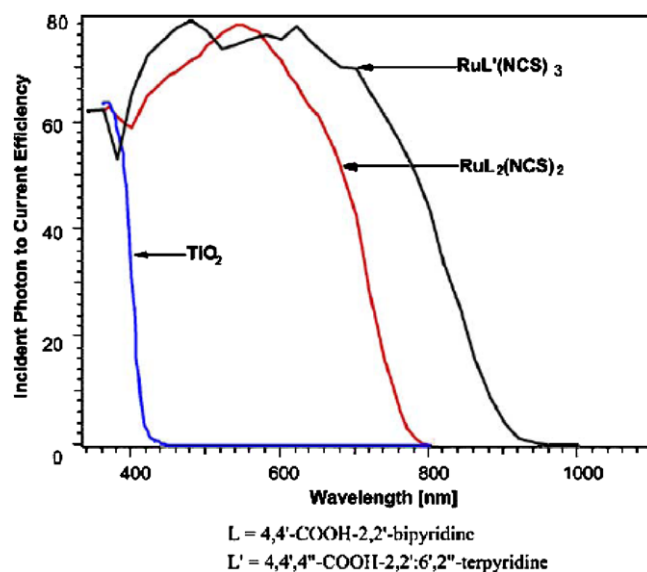

Fig. 3. Sensitization effect can be seen as the shift of incident photon to current efficiency curves to higher wavelengths when coated with the dye as compared with that of naked TiO$_2$. Reprinted with permission from "Conversion of Sunlight by Nanocrystalline Dye Sensitized Solar Cells", Journal of Photochemistry Photobiology A: Chem. 164, (1–3) (2004) 3. Copyright Elsevier, 2004.

Many different compounds have been investigated for semiconductor sensitization, such as porphyrins [24–26], phthalocyanines [27–29], transition metal complexes [30,31], and coumarin [32].

Metal complex sensitizers usually have anchoring (carboxylated) ligands (see Fig. 4) for adsorption onto the semiconductor surface [23].

The dyes having the general structure of $ML_2(X)_2$, where L stands for 2,2′-bipyridyl-4-4′-dicarboxylic acid, M for ruthenium or osmium and X for halide, cyanide, thiocynate, or water have been found promising [22,33,34]. The excitation of Ru complexes via photon absorption is of metal to ligand charge transfer (MLCT) type. This means that the highest occupied molecular orbital (HOMO) of the dye is localized near the metal atom, Ru in this case, whereas the lowest unoccupied molecular orbital (LUMO) is localized at the ligand species, in this case at the bipyridyl rings. At the excitation, an electron is lifted from the HOMO level to the LUMO level. Furthermore, the LUMO level, extending even to the COOH anchoring groups [22], is spatially close to the TiO_2 surface, which means that there is significant overlap between electron wavefunctions of the LUMO level of the dye and the conduction band of TiO_2. This directionality of the excitation is proposed as one of the reasons for the fast electron transfer process at the dye–TiO_2 interface [22].

Cells based on this concept show energy conversion efficiencies up to 11% on small-area cells [35], module efficiencies between 5% and 7%.

2.1. Solid state dye-sensitized solar cells

In a solid state dye-sensitized solar cell the electrolyte is replaced with a p-type semiconductor or organic hole conductor materials [36–38] avoiding problems such as leakage of liquid electrolytes [39].

A solid state dye-sensitized solar cell is schematically shown in Fig. 5. The mesoporous metal oxide electrode, commonly, TiO_2 is placed in contact with a solid state hole conductor. Attached to the surface of the nanocrystalline electrode film is a monolayer of the sensitizing dye. After the excitation of the dye an electron is injected into the conduction band of the semiconductor oxide electrode. The sensitizer dye is regenerated by the electron donation from the hole conductor [40]. In the solid state cell, the charge transport is electronic whereas when using liquid or polymer electrolyte, ionic transportation takes place [19].

The hole conductor must be able to transfer holes from the sensitizing dye after the dye has injected electrons into the TiO_2; that is, the upper edge of the valence band of p-type semiconductors must be located above the ground state level of the dye (see Fig. 6).

Furthermore, hole conductors have to be deposited within the porous nanocrystalline layer penetrating into the pores of the nanoparticle and finally it must be transparent in the visible spectrum, or, if it absorbs light, it must be as efficient in electron injection as the dye. CuI, CuBr or CuSCN were found to be the successful candidates to replace the liquid electrolyte [37,41–43]. The energy conversion efficiency of the fully solid state solar cell of nanoporous n-TiO_2/cyanidin/p-CuI was found to be 1% [41]. The efficiency of the solid state device was further improved by Tennakone et al. [36] employing CuI as hole transporter and ruthenium bipyridyl dye complex as a sensitizer instead of cyanidin. The cells based on this structure gave
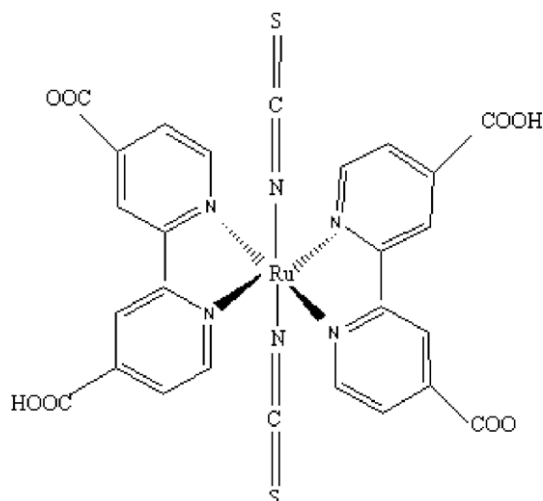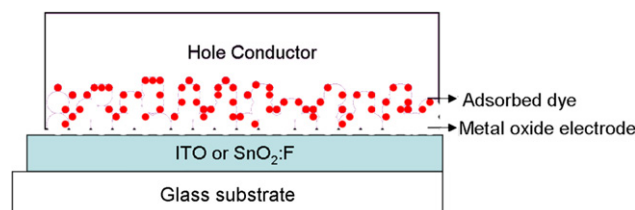

Fig. 5. Schematic description of a solid state dye-sensitized solar cell.

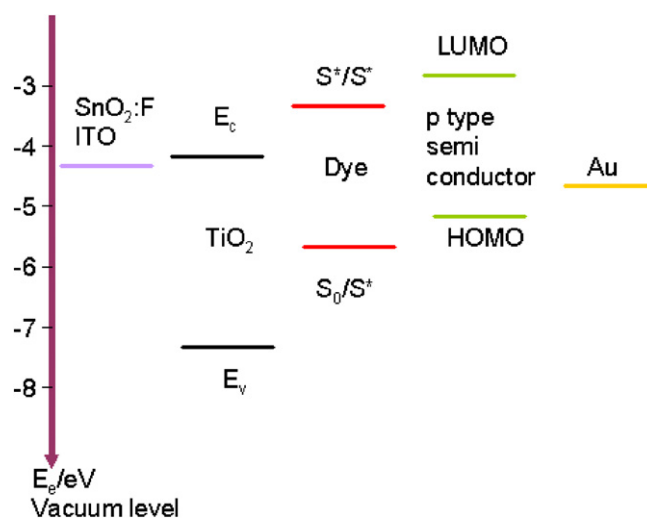Fig. 4. Chemical structure of a ruthenium dye.

Fig. 6. Energy diagram for an efficient charge transfer between solid state dye-sensitized solar cell components.

a maximum power conversion efficiency of 6% corresponding to a fill factor of about 45%. When light intensities were higher than 100 mW/cm^2, the efficiency reduced to about 4.5%.

Although CuI attracted very much attention as a p-type semiconductor, it was observed in 2003 by Sirimanne et al. [44] that the interface of TiO$_2$/CuI degrades due to the release of iodine and the formation of a trace amount of Cu$_2$O and/or CuO for the degradation of the cell. A better stability was observed by covering the TiO$_2$ electrode by a thin MgO layer [45]. Kumara et al. used 1-methyl-3-ethyl-imidazoliumthiocyanate (MEISCN) which is a molten salt, resulting in a power conversion efficiency of 3.75% and a peak IPCE of 59% [46,47]. CuSCN is an alternative to replace CuI with a more stable performance. The solar cells prepared by dissolving CuSCN in *n*-propyl sulphide gave an efficiency of 1.25% [42]. Later, the efficiency of a similar device employing CuSCN was improved to 2% by O'Reagan et al. [48].

Compared to inorganic p-type semiconductors, organic p-type semiconductors possess the advantage of low cost processability. Among organic p-type semiconductors, 2,2′,7,7′-tetranis(*N*,*N*-di-*p*-methoxyphenyl-amine)9,9′-spirobifluorene (OMETAD) has been one of the very first to be investigated. Bach et al. [38] reported a dye-sensitized heterojunction between TiO$_2$ with the amorphous OMETAD as an organic hole transport material. The performance of solid state dye-sensitized solar cells based on spiro-OMETAD was further improved by Krüger et al. [49]. By blending the hole conductor matrix with a combination of 4-*tert*-butylpyridine (*t*BP) and Li[CF$_3$SO$_2$]$_2$N, an efficiency of 2.56% under AM 1.5 illumination was achieved. The efficiency of a similar device was further improved to 3.2% by Krüger et al. [50] by performing the dye adsorption in the presence of silver ions in the dye solution. Mende and Grätzel [51] demonstrated 4% efficiency and also showed that if the pores of nanoporous TiO$_2$ are not completely filled this leads to lower current densities and poorer performance of the cell under sunlight.

Conjugated polymers have also found application in dye-sensitized solar cells since they are well known as polymeric hole transporting semiconductors and metals materials [52]. However, high molecular weight polymers cast from solution, do not penetrate into the pores of the nanoparticles [19]. Poly (3 alkylthiophenes) were used to replace the liquid electrolyte by Sicot et al. [9] and Gebeyehu et al. [8,10].

A polymeric gel electrolyte is considered as a compromise between liquid electrolytes and hole conductors in quasi solid state dye-sensitized solar cells [19,53,54]. A mixture of NaI, ethylene carbonate, propylene carbonate and polyacrylonitrile was reported by Cao et al. [55]. Poly (vinylidenefluoride-co-hexafluoropropylene) (PVDF-HFP) used to solidify 3-methoxypropionitrile (MPN) was utilized by Wang et al. [56] with conversion efficiencies under full sunlight of over 6%, showing high stability under thermal stress at 80 °C as well as under prolonged soaking with light.

Polymer electrolytes are composed of alkaline salts dissolved in a high molecular mass polyether host or polypropylene oxide host [19,57]. In polymer electrolytes, the polymer matrix should be an efficient solvent for the salt, capable of dissociating it and minimizing the formation of ion pairs [19]. Nogueira et al. reported solid state dye-sensitized solar cells by employing a copolymer, poly(epichlorohydrin-co-ethylene oxide), Epichlomer-16 as electrolyte with power conversion efficiencies up to 2.6% [58,59]. Haque et al. reported flexible solid state dye-sensitized solar cells using Al$_2$O$_3$ coated TiO$_2$ electrodes and an I$_2$/NaI-doped solid state Epichlomer-16 electrolyte with an efficiency of 5.3% [60]. Kaneko and Hoshi reported efficiency of 7% with a solid state dye-sensitized solar cell incorporating polysaccharide involving redox electrolytes [61].

2.2. Nanoparticle sensitized TiO$_x$ solar cells

Nanoparticle sensitized solar cells are prepared by replacing the dye with inorganic nanoparticles or quantum dots. They can be adsorbed from a colloidal quantum dot solution [62–64] or produced in situ [65–67]. Inorganic nanocrystals instead of organic dyes could imply tunability of the band-gap and thereby the absorption range [67]. Nanocrystals have large extinction coefficients due to quantum confinement and intrinsic dipole moments, leading to rapid charge separation and are relatively stable inorganic materials [13].

To embed the particles into porous TiO$_2$ films and to use those modified layers as light converting electrodes [68], the incorporated nanoparticles need to be much smaller than the pore sizes of the nanoporous TiO$_2$ electrodes.

2.3. Extremely thin absorber (ETA) solar cells

Extremely thin absorber (ETA) solar cells are conceptually close to the solid state dye-sensitized solar cells [69]. In the ETA solar cells, an extremely thin layer of a semiconductor such as CuInS$_2$ or CdTe or CuSCN replaces the dye in TiO$_2$ based solar cells [70]. The ETA solar cell has the advantage of enhanced light harvesting due to the surface enlargement and multiple scattering. Similar to the solid state dye sensitized solar cells, the operation of the ETA solar cell is also based on a heterojunction with an extremely large interface [71].

Nanu et al. [71] fabricated TiO$_2$/CuInS$_2$ ETA solar cell using atomic layer chemical vapour deposition technique. A 2 nm Al$_2$O$_3$ tunnel barrier and a 10 nm thick In$_2$S$_3$ buffer layer were inserted between TiO$_2$ and CuInS$_2$ to overcome the interfacial recombination problem. This kind of cell gave an energy conversion efficiency of 4%.

2.4. Hybrid solar cells based on bulk heterojunction concept

Another strategy for hybrid solar cells is to use blends of inorganic nanocrystals with semiconductive polymers as a

photovoltaic layer. The basis of this is the bulk heterojunction concept [14,15,17,18]. Bulk heterojunction concept in inorganic/organic hybrid solar cells is similar to that used in organic/organic solar cells. Excitons created upon photoexcitation are separated into free charge carriers at interfaces between two semiconductors in a composite thin film such as a conjugated polymer and fullerene mixtures [72]. Electrons will then be accepted by the material with the higher electron affinity (electron acceptor, usually fullerene or a derivative), and the hole by the material with the lower ionization potential, which also acts as the electron donor. The solubility of the n-type and p-type components is an important parameter of the construction of hybrid solar cells processed from solutions.

Bulk heterojunction hybrid solar cells have been demonstrated in various semiconducting polymer blends containing CdSe [13,14,73], $CuInS_2$ [17], CdS [15] or PbS [74] nanocrystals. This strategy is promising for several reasons [4]:

1. Inorganic semiconductor materials can have high absorption coefficients and photoconductivity as many organic semiconductor materials.
2. The n- or p- type doping level of the nanocrystalline materials can easily be varied by synthetic routes so that charge transfer in composites of n- or p- type organic semiconducting materials with corresponding inorganic counterparts can be studied.
3. If the inorganic nanoparticles become smaller than the size of the exciton in the bulk semiconductor (typically about 10 nm), their electronic structure changes. The electronic structure of such small particles are more like those of a giant molecule than an extended solid. The electronic and optical properties of such small particles depend not only on the material, of which they are composed but also on their size [13,76–79]. Band-gap tuning in inorganic nanoparticles with different nanoparticle sizes can be used for realization of device architectures, such as tandem solar cells in which the different band-gaps can be obtained by modifying only one chemical compound [1,75]. A substantial interfacial area for charge separation is provided by nanocrystals, which have high surface area to volume ratios [73].

Photovoltaic devices from a composite of 8×13 nm, elongated CdSe nanocrystals and regioregular poly(3-hexylthiophene) (P3HT) have been reported by Huynh et al. [73]. Under 4.8 W/m^2 monochromatic illumination at 514 nm such devices with 80% (vol) CdSe had an I_{sc} of 0.031 mA/cm^2 and a V_{oc} of 0.57 V. For a similar device, Huynh et al. [14] achieved a power conversion efficiency of 1.7% under simulated AM 1.5 illumination with CdSe nanocrystals of 7×60 nm size.

Hybrid solar cells based on nanoparticles of $CuInS_2$ in organic matrices were reported by Elif Arici et al. [4,17,18]. Nanocrystalline $CuInS_2$ was used with fullerene derivatives to form interpenetrating interface donor–acceptor heterojunction solar cells. Also bulk heterojunctions blending of $CuInS_2$ and a p-type polymer (PEDOT:PSS); poly(3,4-ethylenedioxythiophene):poly(styrene sulfonic acid) in the same cell configuration showed better photovoltaic response with external quantum efficiencies up to 20% [17,18].

Van Hal et al. [80] reported on hybrid devices based on blends of TiO_2 with MDMO-PPV. To prepare bulk heterojunctions they blended MDMO-PPV with titanium(iv)-isopropoxide, a precursor for preparation of TiO_2 nanocrystals. Subsequent conversion of titanium(iv)isopropoxide precursor via hydrolysis in air in the dark resulted in the formation of a TiO_2 phase in the polymer film. Such a device exhibited an I_{sc} of 0.6 mA/cm^2 and a V_{oc} of 520 mV with a fill factor of 0.42. An external quantum efficiency up to 11% has been achieved.

In a study, Zhang et al. [82] demonstrated hybrid solar cells from blends of MEH-PPV and PbS nanocrystals. They investigated the effect of different surfactants on the photovoltaic performance of the hybrid devices using PbS nanoparticles exhibiting 250 nA short-circuit current and an open circuit voltage of 0.47 V.

Beek et al. [83] reported on hybrid solar cells from regioregular polythiophene and ZnO nanoparticles. They used blends of nanocrystalline zincoxide (nc-ZnO) and regioregular P3HT from solution to construct hybrid polymer–metal oxide bulk heterojunction solar cells. Thermal annealing of the spincast films significantly improved the solar energy conversion efficiency up to 0.9% with I_{sc} of 2.4 mA/cm^2 and a V_{oc} of 685 mV.

Olson et al. [84] fabricated ZnO nanofiber/P3HT composite PV devices. The best performance with this cell structure gave a conversion efficiency of 0.53%. Incorporation of a blend of P3HT and (6,6)-phenyl C_{61} butyric acid methyl ester (PCBM) into the ZnO nanofibers produced an efficiency of 2.03%. The power conversion efficiency was limited by the large fiber spacing and the reduced V_{oc}.

Gur et al. [81] reported air stable all-inorganic nanocrystal solar cells processed from solution. Although the investigated cells completely consisted of inorganic nanoparticles the study is interesting since the solution processed bilayers of inorganic nanoparticles which are also donor acceptor heterojunctions with large interfaces in volume (quasi bulk heterojunctions). By sequentially spincasting films of CdTe and CdSe and sintering the films at 200 °C the cells gave a 2.1% power conversion efficiency under simulated AM 1.5 illumination. The cells employing a Ca top contact capped with Al had an AM1.5 power conversion efficiency of 2.9% with I_{sc} of 13.2 mA/cm^2, V_{oc} of 0.45 V and a fill factor of 0.49.

Recently, Gorbach et al. [85] investigated the photoeffect of an organic layer on Si solar cell performance. The organic layer was grown from aqueous solutions at room temperature. After the organic layer deposition, the improvement in the solar cell performance was observed. I_{sc} was doubled and V_{oc} increased up to 50 mV and FF increased by 20%. This could be important to achieve bet-

ter performances in commercial silicon based solar cells. Similar increase in performances is observed by Kelting et al. [86] upon the sensitization of thin film silicon by a phthalocyanine as strong absorber.

3. Conclusion

Hybrid solar cell research shall combine the advantages of organic semiconductors and nanoparticles with the properties of the inorganic semiconductors and nanoparticles. The parameter space to choose from is large and only a fraction of possible combinations has been realized. Even such limited efforts have attracted much attention due to the simple processability and low cost processing. Their power conversion efficiencies are still low compared with the conventional inorganic solar cells. Further research and development for optimization is required for different types of hybrid solar cell devices.

Hybrid solar cells based on inorganic semiconductor nanoparticles are dependent on the synthesis routes and the reproducibility of such nanoparticle synthesis routes. The surfactant which prevents the particles from further growth is, on the other hand, an insulating layer which blocks the electrical transport between nanoparticles. For the future development of nanoparticles for hybrid solar cells such surfactants should be tailored considering the device requirements.

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