



Polymer electrolytes and perovskites: lights and shadows in photovoltaic devices



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ABSTRACT

Photoelectrochemical devices for the conversion of sunlight into electricity are experiencing a period of fervour due to the use of new solid-state materials capable of ensuring a durability higher than that of liquid electrolytes-based cells. Dye-sensitized solar cells (DSSCs) and perovskite solar cells (PSCs) are currently contending for the role of leader in the field of third-generation photovoltaic technologies. In particular, such success is attributed to the introduction of polymer electrolytes and perovskites, which have recently contributed to obtaining high conversion efficiencies. This review summarizes the most important results in this field and the strategies developed to maximize DSSCs and PSCs performance. In addition, limitations and disadvantages of polymer electrolytes and perovskites are presented, together with possible strategies aimed at improving the photovoltaic efficiency, reducing the cost and limiting the use of toxic and rare materials.

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1. Introduction

The gradual depletion of fossil fuels and the increasing energy demand to support the current model of economic growth are forcing human society to face a global energy problem [1,2]. Indeed, the world population exceeded 7 billion at the end of 2011 and consumes a quantity of energy estimated in $5.6 \cdot 10^{20}$ J per year, which corresponds to a continuous demand of 17.8 TW. Moreover, the world primary energy consumption continuously increases by 1.6% per year and it has recently been estimated that will be around $8.1 \cdot 10^{20}$ J per year (25.8 TW) in 2035 [3]. On the other hand, it has been clearly demonstrated that Sun can represent a powerful solution to this issue. Indeed, the amount of energy that Sun provides to Earth is enormous, the latter receiving 174 PW of incoming radiation at the upper atmosphere. To give an idea, the total solar energy absorbed by Earth in one hour is higher than the energy the world uses in one year [4]; in other words, the amount of solar energy reaching the surface of the planet is so high that in one year it is about twice as much as will ever be obtained from all the non-renewable resources of coal, oil, natural gas and mined uranium [5].

In the last two decades, the scientific community has intensively worked with the aim of proposing new photovoltaic (PV) and photoelectrochemical devices capable of converting

sunlight into electricity. In particular, much attention has been focused on third-generation devices, namely those based on organic or hybrid organic-inorganic materials [6]. The aim of these technologies is to provide good performance through the use of cheap, widely available and non-toxic materials. In this context, dye-sensitized solar cells (DSSCs) have proven to be one of the most valuable third generation PV technologies for the conversion of solar energy into electricity [7]. This is also proven by the fact that the seminal work of O'Regan and Grätzel is the most cited article in the whole energy field [8] and more than 17,000 papers (SciFinder database) have been written to propose new materials, characterization techniques and large-scale implementation of DSSCs [9]. At present, efficiencies higher than 13% have been obtained [10], and the current research efforts are focused on the improvement of the long-term stability of these devices [11–13].

The presence of an organic solvent within the DSSC device has always been seen as a problem, both from technological (effective sealing issues) and safety (flammability) viewpoints. Therefore, in the last decade the scientific community has worked with the aim of replacing the liquid electrolyte with a polymeric one, and very interesting results were obtained [14–16]. On the other hand, other scientific groups have focused their attention on molecular and inorganic solid conductors [17]. This line of research culminated in the investigation of perovskites as solid electrolytes for DSSCs [18]. This leads to another, two years ago it was discovered that certain perovskites can act either as dyes and charge carriers, and therefore a new class of third-generation PV devices – “perovskite solar cells” (PSCs) – was introduced [19].

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The purpose of this review article is to describe the state of the art of polymer electrolytes and perovskites for solar energy conversion, evaluating positive and negative aspects of these two classes of materials born for the same purpose, but which have led to PV devices totally different as regards both the principle of operation and performance (efficiency and stability upon time).

2. Dye-sensitized solar cells: device, operating principle and performance records

A schematic of the interior of a standard DSSC is shown in Fig. 1. At the heart of the device is a mesoporous semiconducting oxide film composed of a network of titanium dioxide nanoparticles (TiO_2 NPs) that have been sintered together to establish electronic conduction. The film thickness is typically around $10\ \mu\text{m}$, the porosity being 50–60% and the NPs diameter 10–30 nm. The TiO_2 layer is deposited on a transparent conducting oxide (TCO) on a glass (or plastic) substrate. The most commonly used substrate is glass coated with fluorine-doped tin oxide (FTO). A monolayer of a molecular sensitizer is chemisorbed on the surface of the nanocrystalline film: the set which includes the conductive substrate, the semiconductor NPs and the adsorbed dye molecules is called “photoanode”. Cell also includes the electrolyte, usually an organic solvent containing the iodide/triiodide (I^-/I_3^-) redox system, and a counter electrode, which is usually a FTO glass coated with a thin layer of a platinum (Pt) catalyst. An inexpensive semiconductor, a dye, a redox couple, traces of a metal catalyst and two conductive glasses: these simple, cheap and abundant materials made the DSSC a PV device markedly different from those of first and second generation [9].

The operating principle of the DSSC is relatively simple and is typical of a photoelectrochemical regenerative cell. With reference to the different reactions reported in Fig. 2 and the kinetic data for the electron transfer processes listed in Fig. 3, the following steps can be distinguished:

- Dye excitation (1): upon absorption of a photon with frequency ν , the sensitizer passes from its ground state (S) to the excited one (S^*), accordingly with the reaction:



- Electron injection (2): the ultrafast (on the femtosecond scale) injection from the excited dye (usually a ruthenium, Ru, complex) in the TiO_2 conduction band (CB) represents one of the most astounding findings in the DSSC research [20]:



Of course, for good device performances the duration of the injection process should be compared with the decay of the excited

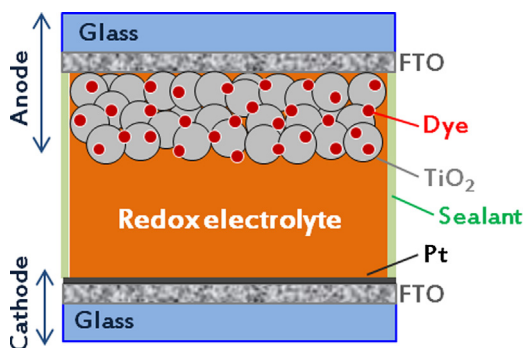


Fig. 1. DSSC device schematic.

state of the dye to the ground state. This phenomenon is ruled by the excited state lifetime of the dye (20–60 ns for Ru complexes, [21]). Semiconductors different from TiO_2 have also been proposed [22,23].

- Regeneration of the oxidized dye (3): the interception of the oxidized dye by the electron donor (I^-) is in the microsecond time domain:



Considering a turnover number (i.e., the cycle life of the sensitizer in the device) of 10^8 , the lifetime of the oxidized dye must therefore be higher than 100 s if the regeneration time is $1\ \mu\text{s}$ [24].

- Electron transport through the mesoporous oxide film (4): the highly efficient charge transport (in the millisecond range) through the nanocrystalline TiO_2 layer represented one of the most puzzling phenomenon when DSSCs were proposed:



Indeed, the mesoporous electrodes are very different from their compact analogues, because the inherent conductivity of the film is very low and a built-in electrical field is not supported by the small size of the individual colloidal particles. TiO_2 films may be viewed as an ensemble of individual NPs through which electrons can percolate by hopping from one crystallite to the next, by means of a charge transport mechanisms that is still under keen debate today.

- Recombination of electron in the semiconductor with oxidized dye (5): it is an undesired process that follows a multiexponential time law, occurring on a microsecond to millisecond time scale, depending on the electron concentration in the semiconductor (and thus on the light intensity):



- Recombination of electrons in the semiconductor with electrolyte species (6): this process influences the electron lifetime (τ_n). Lifetimes observed with the I^-/I_3^- mediator are very long (1–20 ms under 1 sun irradiation), thus explaining the reason behind the choice of this redox couple. The unwanted recombination reaction can be written as:



- Reduction of electron acceptor in the electrolyte at the counter electrode (7): likewise the dye, also the redox mediator must be regenerated. Such a process is carried out through a Pt-catalyzed reduction, easily obtained when nanoscale Pt clusters are used:



Transport of the redox mediator between the electrodes is mainly driven by diffusion, and it is characterized by an electrolyte diffusion impedance, which contributes to the series resistance (R_s) of the DSSC. Typical values are around $2\ \Omega\ \text{cm}^{-2}$, and lead to characteristic times above 1 s [25]. The electrolyte diffusion impedance depends on the diffusion coefficient and concentration of the redox couple, and on the distance between the electrodes. Also the charge transfer reaction at the counter electrode contributes to the R_s of the DSSC, by means of the so-called “counter electrode charge transfer resistance” (R_{CE}). It should be lower than $10\ \Omega\ \text{cm}^{-2}$ to avoid significant losses [26]; otherwise,

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