

Invited Review

Electron injection efficiency in dye-sensitized solar cells

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ARTICLE INFO

Article history:

Received 31 October 2013

Received in revised form 17 February 2014

Accepted 19 February 2014

Available online 12 March 2014

Keywords:

Dye-sensitized solar cells

Electron injection efficiency

Transient absorption spectroscopy

Free energy change

Charge recombination

ABSTRACT

Electron injection processes in dye-sensitized solar cells (DSCs), which involve electron transfer from an excited dye to a semiconductor nanoparticle, have been discussed in many previously reported studies. In this review we discuss the working principles and primary processes of DSCs, as well as these processes' influence on basic properties of solar cells such as open-circuit voltage, short-circuit current, and incident photon-to-current conversion efficiency (IPCE). We focus our attention on the electron injection process, and we introduce methods to determine electron injection efficiency (Φ_{inj}) using time-resolved fluorescence and absorption spectroscopy techniques. We present difficulties associated with obtaining Φ_{inj} by means of such techniques, and we propose nanosecond time-resolved transient absorption spectroscopy as a reliable method. Then, Φ_{inj} values obtained are summarized. Factors limiting Φ_{inj} are discussed from the perspective of free energy changes for electron injection, the molecular structure of sensitizer dyes on the surface, and the presence of fast charge recombination pathways.

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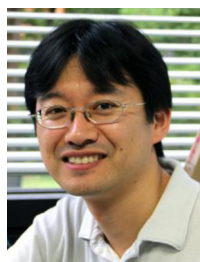
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1. Introduction

Dye-sensitized solar cells (DSCs) are attractive as a new class of solar cells, and an enormous number of studies have been carried out within the last two decades [1,2]. DSCs have received much attention from the research community owing to their attractive characteristics: (1) **Low cost:** The production cost of DSCs is expected to be relatively low because they are assembled from low-cost materials such as TiO_2 and because their preparation does not require high temperatures or high vacuum; (2) **Flexibility:** Because flexible polymer materials can be used as base substrates for DSCs, instead of the glass plates required for conventional solar cells, flexible and lightweight DSC devices can be prepared; (3) **Color availability:** The color of DSCs can be controlled by changing the molecular structure of the dyes contained within the DSCs, and optically transparent DSCs device can also be prepared; (4) **Potential for indoor application:** Under relatively low intensity light irradiation condition, DSCs maintain their performance, whereas solar cells based on inorganic materials exhibit dramatically suppressed performance under low light intensity. Thus, DSCs are suitable for indoor applications.

Owing to the above features, DSCs have potential to extend the range of practical solar cell applications. However, DSCs have not yet found widespread use because their performance characteristics, such as solar-light-to-energy conversion efficiency and long-term stability, have not yet been optimized. To improve such performance characteristics, numerous studies of DSCs' primary processes have been conducted. In this review, the mechanism of DSCs is discussed from a physical chemistry point of view, with particular emphasis on charge separation processes.

Fig. 1 shows the general structure of a DSC. A dye-sensitized electrode having photoelectric conversion properties consists of a nanoporous oxide semiconductor film, called a nanocrystalline film, and sensitizing dye molecules adsorbed on the surface by chemical bonds. TiO_2 has been widely used for the nanocrystalline films, and various dye molecules have been examined for the sensitizing dye [1,2]. The dye-sensitized electrode is immersed into an electrolyte solution containing a redox mediator, such as I^-/I_3^- , and a counter electrode. This electrochemical circuit (i.e., the DSC) operates as a photovoltaic cell. Under working conditions (i.e., when light impinges on the dye-sensitized electrode), the excited sensitizing dye injects electrons into the nanocrystalline film, and the generated conducting electrons can be collected and stored as electricity. The oxidized dye molecules are reduced by the redox

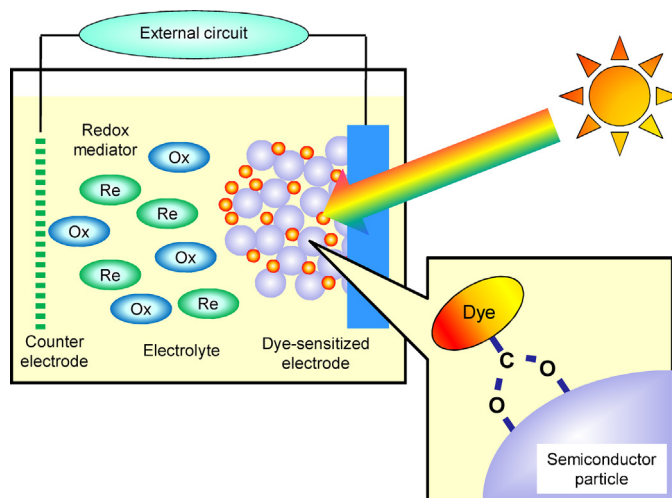


Fig. 1. General structure of dye-sensitized solar cells.

mediator through the counter electrode. Accordingly, DSCs can be considered to be the photovoltaic device based on chemical reactions in molecular level.

Dye-sensitized photocurrent is observed when dyes adsorbed on a semiconductor surface are excited by visible light [3]. In 1976, Tsubomura et al. created solar cells by adsorbing organic dye molecules on sintered ZnO [4], but the cells' solar-energy-to-electricity-conversion efficiency was not sufficiently high. In 1991, O'Regan and Grätzel first reported the basic concept of DSCs mentioned above [5]. Following that seminal report, various types of DSCs were prepared, and devices having high solar-energy-to-electricity-conversion efficiency ($\eta > 10\%$) were realized [2,6,7]. Reported values of η have been increasing gradually [2], and an η of 12.3% recently was reported for a DSC using cobalt complexes as the redox mediator [8]. Calculated thermodynamic efficiency limits in single junction solar cell is known to be 31% (Shockley-Queisser limit), whereas the current status of DSC's efficiency is much lower than the limit. To further improve the performance of DSCs, their primary processes need to be clarified. Some groups have already undertaken such studies [2,9–16].

2. Characterization of device performance for DSCs

2.1. Preparation of DSC devices

DSCs can be prepared relatively easily, as mentioned above. The many methods that have been used to prepare DSCs are too diverse to summarize briefly. Instead, we describe one basic procedure here, with further details available in the literature [14]. First, to prepare nanocrystalline films, a paste containing oxide semiconductor nanoparticles (10–20 nm in diameter) is painted on a conducting glass plate substrate by means of a screen printer, and the painted substrate is calcined at 400–500 °C. Although TiO_2 is the most commonly used nanocrystalline material, other oxide semiconductors such as ZnO and SnO_2 are also used, and nanocrystalline pastes of all of these materials are now commercially available. Next, the nanocrystalline semiconductor films are immersed in a solution of sensitizing dye for several hours to allow dye adsorption at the semiconductor surface. To form chemical bonds between the dye molecule and the semiconductor surface, the dye must contain an anchoring group such as a carboxyl group ($-\text{COOH}$). The nanocrystalline films have a high specific surface area (around 1000), and dye molecules are adsorbed on the surface densely. Thus, incident light can be absorbed sufficiently at the dye's peak absorption wavelength. The counter electrode consists of a conducting

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