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Laser spectroscopic assessment of a phthalocyanine-sensitized solar cell as a function of dye loading



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ABSTRACT

Silicon phthalocyanines with bulky axial ligands are interesting materials for dye-sensitized solar cells (DSSCs) because they possess a strong absorption in the red region (~ 670 nm) and their ligands hinder the stacking of their rings. This suppresses the formation of aggregates, a common tendency of dyes on the surface of dye-sensitized metal-oxide layers. Herein, the novel silicon phthalocyanine, Pc 61, which has ligands terminating in carboxylic acid groups has been synthesized, characterized and used in studies on the harnessing of photons in the long-wavelength visible light range. To investigate the relation between dye loading and performance of DSSCs, a series of Pc 61-sensitized TiO₂ films and DSSC devices with different dye immersion times have been prepared and characterized through optical, electrical and laser spectroscopic studies. To our knowledge, this is the first time that injection rates of phthalocyanine-based DSSC performance have been assessed as a function of dye immersion time. The results of the incident photocurrent conversion efficiency (IPCE) and power conversion efficiency (η_{eff}) studies reveal that solar cells fabricated with a 120 min Pc 61 immersion time have the best performance among the tested ones. This is corroborated by time-resolved emission lifetime data. Our studies demonstrate that the short photoluminescence lifetime of Pc 61-TiO₂ films results from efficient electron injection by excited-state Pc 61 into the TiO₂ film. This agreed with both experimentally obtained higher IPCE and higher η_{eff} .

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1. Introduction

Dye-sensitized solar cells (DSSCs) [1,2] have been extensively studied because of their low cost of fabrication and relatively good efficiency [3–5]. A typical DSSC has a sandwich structure. This structure consists of a light-harvesting dye-sensitized photoanode, a counter electrode, and a redox electrolyte. The active part of the photoanode is a mesoporous nanocrystalline wide band gap metal oxide, such as TiO₂ as shown in Fig. 1. Since the excited state of the dye lies above the conduction band of the metal oxide, an energy difference exists. This difference provides a driving force for photo-induced electron injection (2). In an optimized cell, a percentage of the injected electrons can perform work in the external circuit and complete the circuit, replenishing of the

oxidized dye via the redox electrolyte, as show in paths (4) and (5) of Fig. 1.

However, not all of the photoexcited electrons produced in a DSSC perform work in the external circuit. For example, Listorti [6] and Watson [7] have shown that relaxation of the excited dye by fluorescence (6) or by recombination with oxidized electrolyte (7) can significantly reduce the overall efficiency of a DSSC. Furthermore, the injected electrons can also undergo back electron transfer from the metal oxide anode to the dye (8) [8,9].

The time scales of these pathways and the kinetic competition between them play an important role in electron injection and collection efficiency and affect the overall power conversion performance of a DSSC [6]. The efficiency of the electron injection process also plays an important role because it competes kinetically with the decay of the excited state of the dye [9–11]. This is highly dependent on the distance between the dye and the metal-oxide surface [12].

To date, the most promising DSSCs have been based on TiO₂/Ru (II) polypyridyl complexes. These DSSCs have efficiencies exceeding 11% [13,14]. However, the promise of such cells is reduced by

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the high cost of the Ru complexes and their lack of absorption in the red/near infrared (IR) spectral region. In fact, many potentially useful dyes, while absorbing in the higher energy region of the visible spectrum (400–600 nm) do not absorb in the red region. However, one group of potential dyes, the phthalocyanines (Pcs), has a strong absorption in this region and has high chemical, thermal and photostability. This makes them attractive for DSSCs [15–22]. Thus the combination of phthalocyanines and higher energy absorbing dyes has the potential to yield panchromatic sensitization [5,23,24]. Due to the planar structure of the phthalocyanine ring, those phthalocyanines which do not carry bulky ligands or substituents have a strong tendency to aggregate. When they are on a semiconductor surface, this tendency leads to fast deactivation of the dye excited state and a very low incident photon to current efficiency (IPCE) of the cells [17,25]. To alleviate the problem of aggregation, the addition of phthalocyanines with bulky equatorial or axial substituents can be used [26–29]. The axially ligated silicon phthalocyanines are interesting because not

only can the axial substituents prevent undesired dye aggregation on the semiconductor surface, but they also can increase the solubility of the phthalocyanines in common organic solvents and thus ease electrode construction [30,31]. Martin-Gomis et al. showed a promising photovoltaic response from a silicon phthalocyanine axially ligated with carboxylic acid groups as a dye sensitizer, (although the reported power efficiency was below 1%) [32]. Further, in related work Macor and co-workers found that near-IR sensitization can be achieved by utilizing axially ligated silicon naphthalocyanine dyes on TiO₂ surfaces, and observed a photocurrent in a DSSC constructed from such sensitized electrodes [33].

Because improvement of the electron-transfer dynamics in DSSCs is needed for further optimization of DSSCs [34], an investigation of the interaction between the dye sensitizer and the semiconductor surface is desirable. This interaction is directly influenced by dye coverage and orientation. These in turn are influenced by electrode immersion time (during photoanode preparation). However, little work utilizing femtosecond laser spectroscopy has been focused on immersion time [13,35].

In this study, we report the synthesis and use of a novel silicon phthalocyanine, Pc 61 (Fig. 2), a symmetric sensitizer dye with two trans-axial ligands terminating in carboxyl groups. This dye and nanoparticle TiO₂ have been used to make DSSC anodes. Following the lead of others [36–39], we have used time-resolved spectroscopies to monitor the interfacial electron injection process, the relationship of Pc 61 coverage and the overall DSSC efficiency. In addition, a series of sandwich-type DSSCs were prepared to investigate the relationship between immersion time, injection rate and power conversion efficiency.

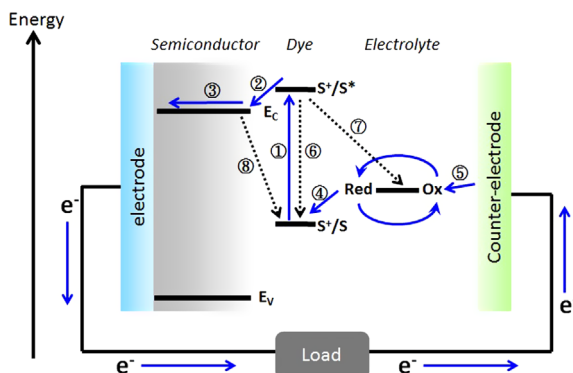


Fig. 1. A schematic representation of the mechanisms of a dye-sensitized solar cell. The electron transfer processes considered here are: (1) light absorption, (2) electron injection, (3) electron diffusion, (4) reduction of the oxidized dye by the redox electrolyte, (5) reduction of the oxidized redox electrolyte by the cathode, (6) luminescence, (7) reduction of the electrolyte by the photoexcited sensitizer dye, and (8) back electron transfer.

2. Materials and methods

2.1. Synthesis

The first step of the synthesis of Pc 61 is an ordinary axial ligand exchange. The second is an ester hydrolysis using mild

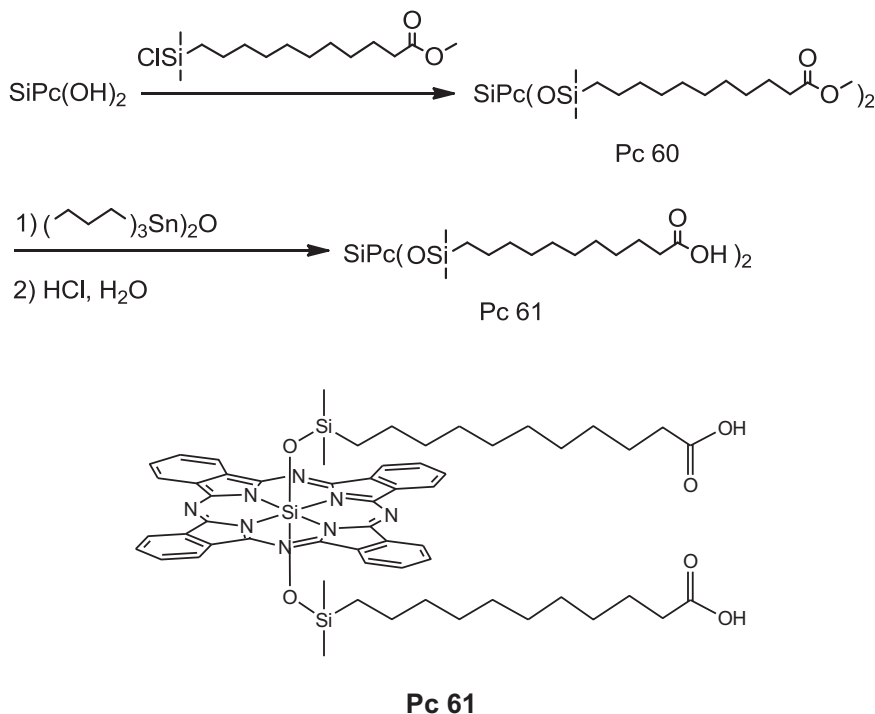


Fig. 2. Synthesis and structure of SiPc[OSi(CH₃)₂(CH₂)₁₀COOH]₂, Pc 61.

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