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Research paper

Efficiency enhancement in dye sensitized solar cells using dual function mesoporous silica as scatterer and back recombination inhibitor

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ABSTRACT

In the present work, we report the usage of mesoporous silica for improving light harvesting as well as for suppression of back recombination without affecting the extent of dye loading on TiO_2 films. Synthesized mesoporous SiO_2 was characterized by X-ray photoelectron spectroscopy, X-ray diffraction, Brunauer Emmett and Teller measurement, Scanning electron microscopy and Transmission electron microscopy. DSSCs were fabricated by incorporating different wt% of mesoporous SiO_2 in TiO_2 paste. An improvement of 50% was observed for devices fabricated using 0.75 wt% of mesoporous SiO_2. The mechanism behind the improvement was investigated using electrochemical impedance spectroscopy and UV–Vis spectroscopy.

1. Introduction

Owing to their eco-friendly attributes and cost effectiveness, dye sensitized solar cells (DSSCs) have emerged as promising candidates among different generation solar cells [1]. So far the maximum reported efficiency for DSSCs is approximately 13%, which is below the theoretical estimate by approximately 20% [2]. Efforts are therefore underway to enhance DSSC efficiency through different approaches. So far improvement in efficiency has been achieved either via light harvesting or by reduced recombination. In order to improve DSSC efficiency via light harvesting, various approaches like co-sensitization of TiO_2 electrode with different dyes [3], plasmonic enhancement [4,5] synthesis of new sensitizers with higher molar extinction coefficients [6] and incorporation of scattering particles have been implemented [7]. On the other hand, reduced back electron transfer (or recombination) was achieved using post and pre-treatment of mesoporous TiO_2, blocking layer on FTO, insulating layer on mesoporous TiO_2 or co-adsorbents in the electrolyte [8–12].

Of late the usage of large sized nanoparticles as well as core–shell structures as scatterers in TiO_2 have attracted attention since the incorporation of scatterers within TiO_2 layer results in enhancement of interaction of photons with dye molecules. This enhancement is attributable to the scattering of light by large sized particles, which in turn results in enhancing the device efficiency [13,14]. This approach has generated considerable interest due to the availability of a variety of nanoparticles. It has been reported that silica acts as an efficient scattering center for improving light harvesting owing to higher difference in refractive index from surrounding medium, (the refractive index of TiO_2 is 2.56 while that of silica is 1.46) [15]. Rho et al. had observed 22% enhancement in efficiency with silica beads as scatterers [16]. On the other hand, 20% improvement in photovoltaic performance was reported when SiO_2/TiO_2 core/shell nanoparticles were used [14]. Park et al. had observed 46% improvement with hybrid silica conjugated TiO_2 nanostructures [17]. However, these approaches also resulted in reduced dye loading and in turn reduced light harvesting. To address this drawback, a two layered approach, each having different sized nanoparticles have been explored. In this approach, the first layer consists of small sized mesoporous TiO_2 nanoparticles (10–15 nm) while second layer contains large sized scattering particles. Wang et al. had reported 18% improvement in efficiency by using multilayer approach as compared to single layer approach [18]. Niaki et al. had observed 7% enhancement in photovoltaic performance [19] using double layer film doped with Zn ions [20]. However, single reflection of light limits the improvement that is attainable in photovoltaic current density and therefore restricts the device efficiency [18].

We have earlier proposed that a significant improvement in efficiency can be achieved by improving both light harvesting and reduced recombination through a novel strategy of co-sensitization [3,21].

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In the present case, mesoporous SiO_2 was chosen since it not only acts as good scatterer but also provides efficient barrier between TiO_2 and electrolyte. In our approach, mesoporous SiO_2 particles with controlled pore size were synthesized so that TiO_2 sites remain available for dye loading (Fig. 1). We have thus demonstrated that an improvement of 50% in device efficiency can be achieved by proper choice of mesoporous SiO_2 owing to cumulative effect of enhanced light harvesting and reduced back recombination.

2. Materials and methods

2.1. Materials

Polyethylene block polymer, tetraethyl orthosilicate (TEOS) and di-tetrabutylammoniumcis-bis(isothiocyanato)bis(2,2-8bipyridyl-4,4dicarboxylato) ruthenium(II) (N719) dye were purchased from Sigma Aldrich. Fluorine-doped tin oxide coated substrates (FTO), TiO_2 paste (Ti-Nanoxide T) and platinum catalyst precursor (Platisol-T) were procured from Solarnix and electrolyte containing I^-/I_3^- as redox couple in 3-methoxypropionitrile (EL-HSE) was received from Dyesol.

2.2. Synthesis of mesoporous silica and fabrication of DSSCs

Mesoporous SiO_2 was synthesized using Polyethylene block polymer and tetraethyl orthosilicate as precursors using the method reported earlier (S1) [22,23]. For fabrication of DSSCs, FTO substrates were first treated with 40 mM aqueous solution of $TiCl_4$ at 70°C for 30 min and then annealed at 450°C for 30 min to form a compact layer of TiO_2 (c-TiO_2). Mesoporous SiO_2 particles were added to the TiO_2 paste in different weight% 0.5 wt%, 0.75 wt% and 1 wt% under continuous magnetic stirring for homogeneous mixing of these particles into TiO_2 paste. The resultant paste was further used to prepare films using earlier procedure [3]. The electrodes thus prepared using 0.5 wt%, 0.75 wt% and 1 wt%, of SiO_2 are referred as 0.5 S, 0.75 S and 1 S, respectively, the reference electrode (i.e. TiO_2) is referred as T hereafter. These electrodes were sensitized with N719 dye by dipping electrodes in 0.3 mM dye solution in ethanol for 24 h. The thickness of photoanodes was estimated to be 2.5 μm from their cross-sectional view in SEM image. Photocathodes were prepared by drop casting platinum catalyst precursor onto the cleaned FTO followed by annealing the same at 450°C. Finally, the cells were assembled by using parafilm and electrolyte containing I^-/I_3^- as redox couple in 3-methoxypropionitrile.

2.3. Characterization

Brunauer Emmett and Teller (BET) analysis was performed using Micromeritics porosity analyser ASAP 1020. The X-ray photoelectron (XPS) spectra was recorded using Mac-2 electron analyser with K_α as radiation source, Scanning electron microscopic images were taken with Carl Zeiss Supra 55 while Transmission electron microscopic images were obtained using JEOL JEM-2100. UV–Vis spectra were recorded with Shimazu, UV–VIS–NIR 3600. X-ray diffraction (XRD) pattern was obtained from D8 Focus, Bruker Ettlingen, Germany with Cu Kα as source. The photovoltaic properties of DSSCs were carried out using Sciencetech Solar Simulator equipped with 150 W Xenon lamp and Autolab ECO Chemie PGSTAT 30 Potentiostat/Galvanostat. The illumination intensity was 100 mW/cm^2 at AM 1.5 G. Prior to the characterization, intensity of lamp was calibrated using standard silicon cell.

3. Results and discussions

Synthesized mesoporous SiO_2 was characterized using XPS and BET measurements. As shown in Fig. 2(a), the nitrogen adsorption–desorption isotherm exhibited a type IV shape with type H1 hysteresis, suggesting well-ordered mesoporous SiO_2 with a narrow and uniformly distributed cylindrical pores [24]. The BET surface area of mesoporous structure of SiO_2 was estimated to be

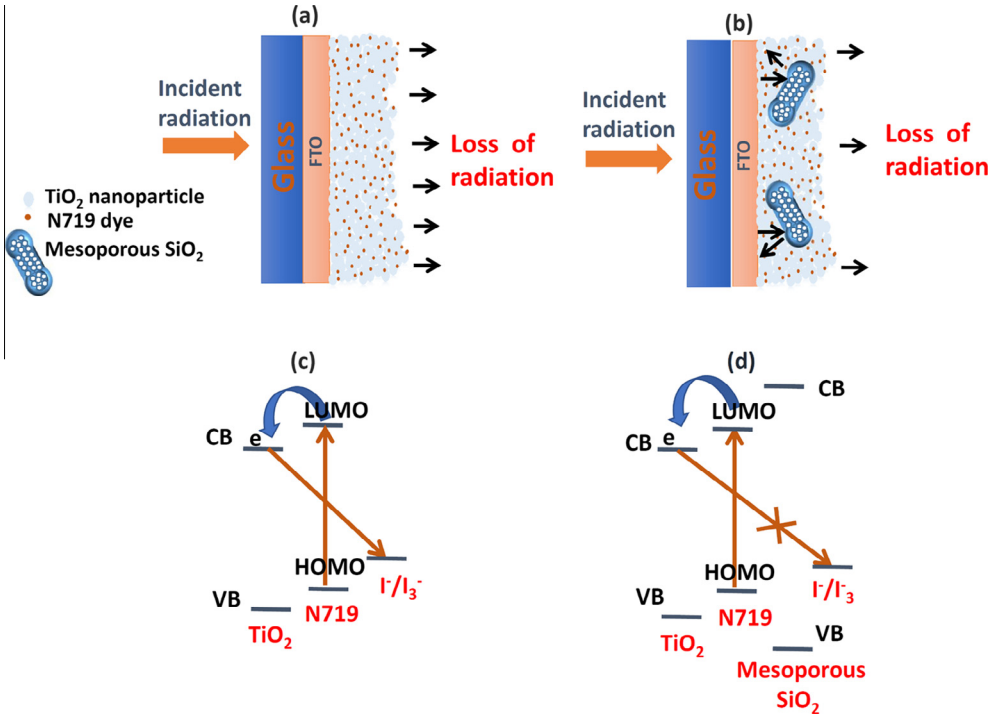

Fig. 1. Schematic representing absorption of photons (a–b) and recombination of charge carriers (c–d) before and after incorporating mesoporous SiO_2 respectively.