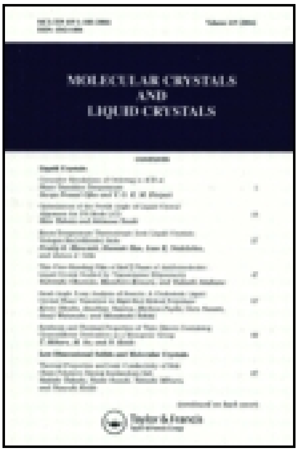

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Effects of the Multi-Layered Light Scattering Layers Dual-Coated by Al_2O_3 and SiO_2 on Dye-Sensitized Solar Cells

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Multi-layered light scattering layer has been designed to improve the energy conversion efficiency of dye-sensitized solar cells (DSSCs) by scattering of the incident light. To enhance the light scattering properties, the surface of light scattering TiO_2 particle was dual-coated with nano-sized Al_2O_3 and SiO_2 particles by using a modified sol-gel method. Four kinds of the TiO_2 electrodes with different thicknesses were fabricated and their photovoltaic effects on DSSCs were investigated. By increasing the thickness of light scattering layer, the total reflectance of working electrodes increased to 37%, resulting in an increase in the energy conversion efficiency up to 6.76%.

Keywords Dye-sensitized solar cell; light scattering layer; modified sol-gel method; dual-coating; energy conversion efficiency.

Introduction

Since Gratzel's group reported a low cost and high efficiency photovoltaic cell based on dye-sensitized colloidal TiO_2 films [1], dye-sensitized solar cells have been in the limelight as a promising alternative to conventional solar cells [2]. Previous studies have indicated that one of the key parameters to improve the efficiencies of DSSCs is the surface properties of the active layer [3,4]. To cut down the production cost of DSSC modules, we have to reduce the dye without sacrificing photovoltaic performances. An alternative about this issue was to introduce a light scattering layer on the nanoparticle TiO_2 layer [5,6]. Meanwhile, multi-layered light scattering layer was designed to improve the light harvesting efficiency of the dye-sensitized solar cells by scattering of the incident light. But, there is still a limitation on proving the effects of multi-layered light scattering layer without a modification of scattering layers.

In this study, to enhance the light scattering properties, the surface of light scattering TiO_2 particle was dual-coated with nano-sized Al_2O_3 and SiO_2 particles by a modified sol-gel method at room temperature. Four kinds of the TiO_2 electrodes with different thicknesses

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were fabricated and their photovoltaic effects on the DSSC devices were investigated as a function of thickness.

Experimental

To obtain the light scattering TiO_2 particles dual-coated with Al_2O_3 and SiO_2, to begin with, we prepared Al_2O_3-coated TiO_2 particles as follows. A commercial light scattering TiO_2 powder(Kojundo Chemical Laboratory Co. Ltd, 99.99%) for light scattering layer was coated with Al_2O_3 by a modified sol-gel method [7] where a colloidal Al_2O_3 (AL20, Nyacol Nano Technologies, Inc.) was used as a precursor. We prepared the colloidal Al_2O_3 of about 0.235 wt% concentration by diluting with de-ionized water at a pH 7. Light scattering TiO_2 of 4g was added in the diluted colloidal Al_2O_3 of 80ml and stirred with stirring using a magnetic bar for 3h. The Al_2O_3-coated light scattering TiO_2 particles were obtained by filtering and drying at 80°C for 12 h. Subsequently, we modified the surface of the Al_2O_3-coated TiO_2 particles by using a colloidal silica (LUDOX-AM 30, Sigma-Aldrich Chemical Co.) as a precursor materials. The single Al_2O_3-coated light scattering TiO_2 particles was stirred in the diluted colloidal SiO_2 of about 0.018 wt% concentration. The solution was stirred with magnetic bar for 3 h. The light scattering TiO_2 suspensions were washed three times in ethanol. Finally, dual-coated light scattering TiO_2 particles were obtained by filtering and drying at 80°C for 12 h.

On the other hand, we prepared the dual-coated light scattering TiO_2 pastes, composed of the mixture at a ratio of 25% TiO_2 powder, 2% Acetic acid, 10% Ethylcellulose, and 63% α-terpineol by weight percent. A commercial anatase TiO_2 nanoparticle paste (DSL 18NR-T, Dyesol) was used for the nanoparticle TiO_2 layer of the photoelectrode. Nanoparticle TiO_2 paste was printed on the transparent conducting glass substrate (F-doped tin oxide, FTO) with a size of 2 cm^2, and then the printed nanoparticle TiO_2 films were dried at 80°C. The light scattering TiO_2 paste was printed on nanoparticle TiO_2 films/FTO glass, and then the printed light scattering TiO_2 films were dried at 80°C. Subsequently, the printed TiO_2 films were annealed at 450°C for 15 min, yielding a working electrode, namely, two layered TiO_2 films consisting of the nanoparticle TiO_2 layer (9 μm thick) and the light scattering TiO_2 layer (4 μm thick per one layer). To obtain the two layers and three light scattering layers, the light scattering TiO_2 paste was printed two and three times, respectively, on nanoparticle TiO_2 films/FTO glass. The active area of two layered TiO_2 films was 0.5×0.5 cm^2. Two layered TiO_2 films were immersed in 0.5 mmol ethanol solution of N719 dye (Dyesol) for 24 h and dried in an oven.

The DSSC device was assembled by placing a platinum-coated counter electrode, fabricated using Pt-paste (PT-1, Dyesol), on the dye-sensitized working electrodes, and clipping two electrodes together. Two electrodes were separated by a Surlyn spacer (25μm thick) and sealed by heating. A redox electrolyte solution (Idololyte AN-50, Solaronix) was introduced between two electrodes.

An investigation on the effects of multi-layered dual-coated light scattering layer was done by the help of FESEM(Field Emission Scanning Electron Microscopy, HITACHI S-4800), XRF(X-Ray Fluorescence spectrometer, PHILIPS PW2400), UV-VIS-NIR(Ultraviolet-Visible-NearInfraRed, Cary 5 spectrophotometer). IPCE(Incident to Photon-to-Current Efficiency) characteristics of DSSC devices were measured by an action spectrum measurement system(PEC-S20, Peccell). I-V characteristics of DSSC devices were measured under 100 mW/cm^2 of simulated AM 1.5 solar light by using Solar Simulator(PEC-L12, Peccell).

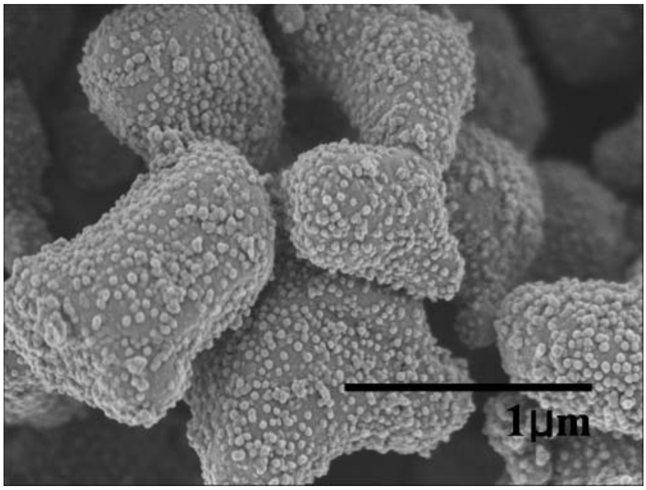

Figure 1. SEM images of dual (Al_2O_3, SiO_2)-coated light scattering TiO_2 powder.

Results and Discussion

Figure 1 represents SEM images of the dual-coated TiO_2 powder. The surface of the dual-coated TiO_2 being 300∼800 nm in size can be clearly seen. One can find the uniform distribution of nano-sized oxide particles on the surface of TiO_2 powder. The large TiO_2 particles used in this paper increase the light scattering yield of incident solar light in the wavelength range $\lambda = 550$–800 nm.

Figure 2 shows the XRF spectra of the dual-coated light scattering TiO_2 powder. The XRF spectra reveal that AlKα, SiKα, TiKα, and TiKβ peaks are well observed. This result confirms in a roundabout way that the nano-sized oxide materials dual-coated on the surface of the light scattering TiO_2 powder are Al_2O_3 and SiO_2.

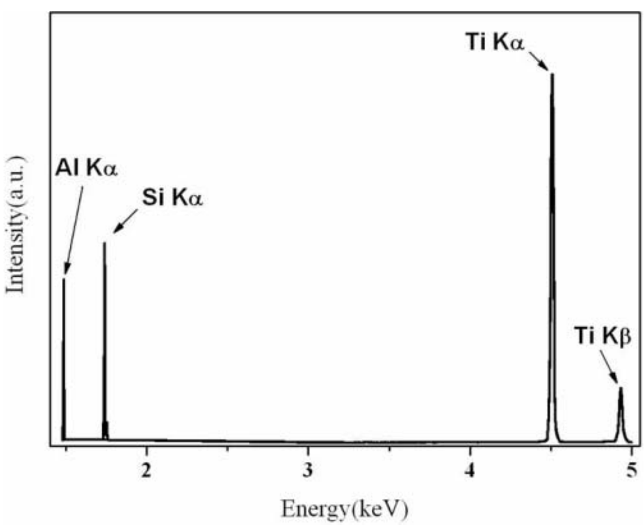

Figure 2. XRF results of dual (Al_2O_3, SiO_2)-coated light scattering TiO_2 powder.

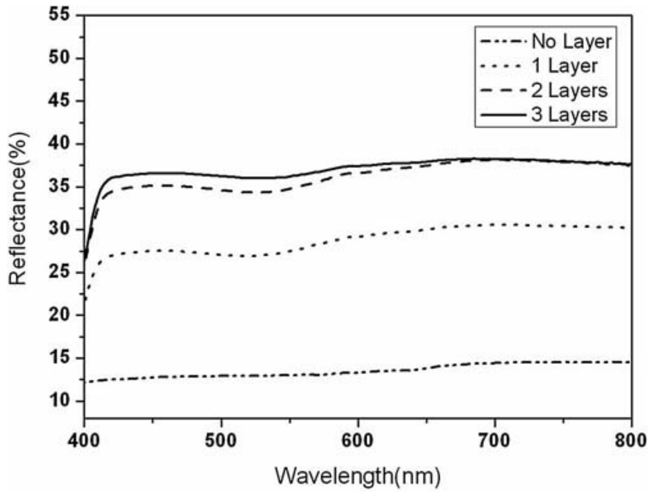

Figure 3. Total reflectance of the working electrodes with and without the light scattering layers.

Figure 3 shows the total reflectance of the dual-coated light scattering layer to investigate on the light scattering abilities as a function of thickness(the number of layers). Reflectance data implies the intensity of the scattered light toward the nanoparticle TiO_2 layer by the light scattering TiO_2 particles, since an increase of the light scattering abilities yields an increase of the reflectance. By increasing the thickness of the light scattering layer, the total reflectance of the light scattering layer was increased to 37%, as shown in Fig. 3.

Figure 4 shows IPCE result of DSSC devices with the dual-coated light scattering layer as a function of thickness. We found that the IPCE data increases as increases the thickness of the light scattering layer. Because IPCE describes the photocurrent collected per incident photon flux as a function of illumination wavelength [8] and means the maximum

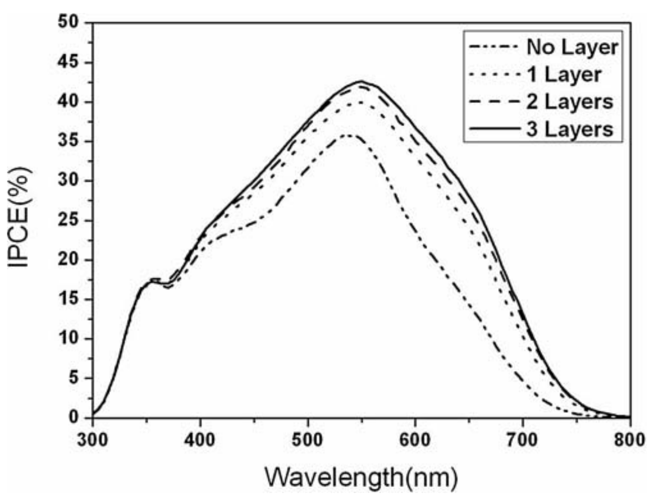

Figure 4. IPCE results of the DSSC devices with and without the light scattering layers.

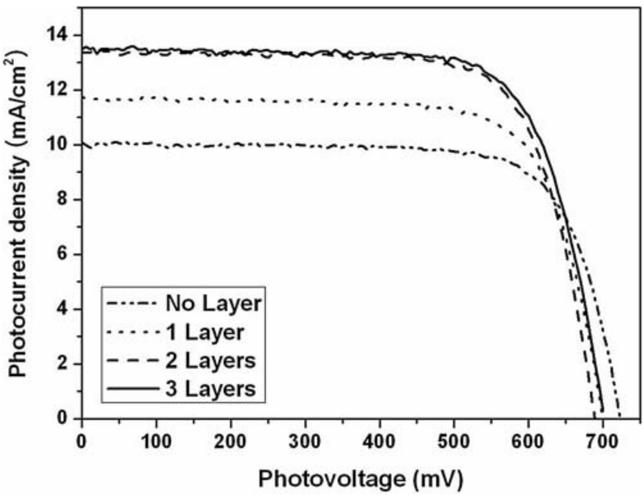

Figure 5. Photocurrent-Photovoltage (I-V) curves of DSSC devices with and without the light scattering layers.

possible efficiency with which incoming radiation can convert to electrons by the dye [9], one can expect that DSSC with multi-layered light scattering layer exhibits better device performances.

Figure 5 shows the photocurrent-photovoltage(I-V) characteristics curve of the DSSC device. The detailed photovoltaic characteristics are listed in Table 1. As shown in Table 1, DSSC with three-layered dual-coated light scattering layer exhibits the highest device performance such as open-circuit photovoltage (V_{oc}), short-circuit photocurrent (J_{sc}), fill factor (FF). It is of interest that the energy conversion efficiency gradually increases with the thickness of the light scattering layer up to 6.76% in the case of the three-layers. From the results of Table 1, J_{sc} value gradually increases with the thickness of the light scattering despite decreasing the fill factor. These results can be explained that the DSSC with the multi-layers gradually increases the light scattering yield with the thickness of the light scattering in spite of the losses due to the series and parallel resistances. The results indicate that an increase in the light scattering ability with increasing thickness of layers, probably due to increasing in reflectance and IPCE results as seen in Figs 3 and 4, leads to an

Table 1. I-V characteristics of DSSC devices with and without the scattering layers. "No Layer" specimen is DSSC without a scattering layer. "1 Layer" specimen is DSSC with a scattering layer printed on the active layer. "2 Layers" and "3 Layers" specimens are DSSC with two-layered and three-layered light scattering layers printed on the active layer, respectively

Specimens	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF (%)	Efficiency (%)
No Layer	723.3	9.99	73.69	5.33
1 Layer	699.8	11.64	72.68	5.92
2 Layers	688.4	13.38	71.97	6.63
3 Layers	700.1	13.48	71.65	6.76

enhanced performance of the dye-covered TiO_2 nanoparticles. But there is no difference between two-layers and three-layers in reflectance of photoanode and efficiency of DSSC, which are due to the limitation of the path lengths of incident solar radiation by up to two-layers. One can find that the overall characteristics of DSSC were enhanced enough in the case of the two-layers. Thus, multi-printing of the dual-coated TiO_2 light scattering layer is a way for improving the device characteristic of DSSC.

Conclusions

The light scattering TiO_2 particles were dual-coated with Al_2O_3 and SiO_2 nanoparticles by a modified sol-gel method. The reflectance of the light scattering TiO_2 layer consisting of the working electrodes increases with increasing the thickness of the light scattering layer, indicating that the thickness of the light scattering layer affects the quantities of light scattered toward the active layer to large amount. DSSC showing higher reflectance of the working electrode revealed higher J_{sc}, resulting in higher IPCE and the energy conversion efficiency up to 6.63%, especially in the case of DSSC with two-layered dual-coated light scattering layer.

Acknowledgment

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