Fabrication of high-efficiency dye-sensitized plastic photocells by low-temperature electrode preparation using a binder-free nanocrystalline TiO₂ coating paste

Yujiro Kijitori, Takurou N. Murakami, Norimichi Kawashima, and Tsutomu Miyasaka Graduate School of Engineering, Toin University of Yokohama, Yokohama, Kanagawa 225-8502, Japan

preparation Low-temperature of mesoporous semiconductor electrodes for dye sensitization has been a subject of recent intense study for realizing full-plastic thin flexible photocells. We have devised high-efficiency plastic photocells by two approaches, electrophoretic deposition of TiO₂ nanocrystalls¹⁻³ and preparation of binder-free viscous TiO_2 paste for doctor blade coating. Both methods need completion of inter-particle binding (necking) that enables efficient diffusion of conductionband electrons through particles. In this paper binder-free pastes for doctor-blade coating were prepared by using TiO₂ precursor sol, which causes efficient chemical necking at temperatures below 200°C. We could achieve energy conversion efficiency of 4.6-5.4%, which is comparable with the top efficiency obtained by electrophoretic technique combined with post chemical treatments and with the efficiency obtained by hightemperature sintering of the same TiO₂ particle on a glass electrode.

Nanocrystalline TiO2 (Showa Denko F-5, average size, 20 nm) was mixed with light-scattering large TiO₂ particle (G2, 500 nm) and acidic TiO₂ precursor sol in a mixture solvent of tert-butanol and ethanol to give a viscous paste free of binder materials. The viscosity of the paste was variable and was controlled by the composition of solvents. The paste (TiO₂ content, 14wt%) was coated on a transparent ITO-PET or ITO-PEN film (10-15 ohm/ \Box) by the doctor blade method, dried, and heat-treated at 150-200°C for 5 min to form a mesoporous TiO₂ layer. TiO₂-coated films showed high adhesion strength for TiO₂ layer (about 10 µm thick) against bending (see Fig. 1). The film electrode was dye-sensitized with a Ru complex (N719) and made into a thin cell (effective electrode area, 8×8 mm) with Pt-coated counterelectrode and methoxyacetonitrile-based I^{-}/I_{3}^{-} electrolyte composition.

Figure 2 shows I-V characteristics for the TiO₂ layer according to the present method and a reference layer prepared by high-temperature sintering at 550°C on glass electrodes. Photocurrent density (J_{sc}) proved to reach the same level as that of sintered electrode. Open-circuit voltage $(V_{\rm oc})$ was lower than the sintered one suggesting that there is a wide distribution in conduction-band potential of TiO₂ layer as a result of low temperature preparation process. By optimization of the large particle (G2) content and thickness of TiO2 layer, energy conversion efficiency under 100 mW/cm² reached around 4.6% at 11-12g TiO₂/m² and G2 content of 30 wt%, as shown in Fig. 3. Non-volatile molten salt (imidazolium iodide) was also used as electrolyte, with which efficiency level was decreased to 3-3.5%. With this method, we are fabricating large-size full plastic photocells (>50 cm²) by introducing metal grid pattern electrodes on the ITO surface for current collection.



Fig. 1 Flexible plastic film photoelectrode bearing dye-sensitized mesoporous TiO₂ layer



Fig. 2 Photocurrent-voltage characteristics of the low-temperature TiO_2 coating in comparison the same coating having been treated by high-temperature sintering at 550°C



Fig. 3 Photocurrent-voltage characteristics of the low-temperature coated binder-free TiO₂ layer at incident intensities of 1 sun (100 mW/cm²) and 1/4 sun.

References

1. T. N. Murakami, Y. Kijitori, N. Kawashima, and T. Miyasaka, *Chem. Lett.*, **32**, 1076(2003).

2. T. Miyasaka, Y. Kijitori, T. N. Murakami, M. Kimura, and S. Uegusa, *Chem. Lett.*, **31**, 1250(2002).

3. Takurou N. Murakami, Yujiro Kijitori, Norimichi Kawashima, and Tsutomu Miyasaka, *J. Photochem. Photobiol. A*, 2004, in press.