ROOM TEMPERATURE FABRICATION OF FLEXIBLE DSSCS USING ELECTROSPRAY METHOD

Horim Lee[1,2], Daesub Hwang[2], Yongsok Seo[1], Dong Young Kim[2*]
[1] Advanced Functional Polymeric Materials Lab, Seoul National University, Korea
[2] Optoelectronic Materials Lab, Korea Institute of Science and Technology, Korea
* Corresponding author(dykim@kist.re.kr)

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1. Abstract

Flexible plastic DSSCs are attractive commercial applications. But there are intrinsic problems with fabrication temperature for flexible plastic DSSCs because the two electrodes of DSSCs are consists of polymeric materials (ITO/PEN). So Making of Photo-electrode is allowed at only low temperature (~150°C). In this study, we introduced the binder-free TiO_2 electrodes using electrospray method for room temperature fabrication. Through this method, we obtained the TiO_2 nanospheres which show hierarchical structure. To make good adhesion between secondary particles and substrate/TiO_2 electrode, we apply the compression on photo electrode. As a result, we can find that electrical contact between TiO_2 secondary particles was enhanced after EIS measuring. By optimizing the thickness of the TiO_2 electrode, the cell shows conversion efficiency up to 6%.

2. Introtuction

Due to today's increased demands for energy supply, many people pay attention to alternatives which is cheap, clean source from the sun. In terms of this view, Dye-sensitized solar cells (DSSC) are regarded as one of best candidates because of its low fabrication cost. Since 1991 when the first DSSC was invented, the energy conversion efficiency was increased over 11% for glass substrates.[1] However, the current energy conversion efficiency for flexible DSSCs is lower than glass DSSCs.[2] In case of flexible DSSC, especially for ITO/PEN substrates , it is hard to make highly efficient DSSCs because the substrates can't be sintered.

The e-spray technique has recently been considered as a cheap and simple process to directly deposit thin films from their colloidal solutions. The techniques can be applied widely in modern material technologies, microelectronics, nanotechnology, and industries for the deposition of various ceramic powders, polymer powders, and TiO_2 electrodes for DSSCs, but have not previously been used to fabricate hierarchically-structured TiO_2 spheres. During e-spray deposition known as induction or conduction charging, the droplets can be charged of their atomization by mechanical forces in the presence of electric field between the solution and the depositing substrates. The electric field develops an electric charge on the liquid surface and the charge is carried out by the droplets detaching from the jet. The advantage of the e-spray is that

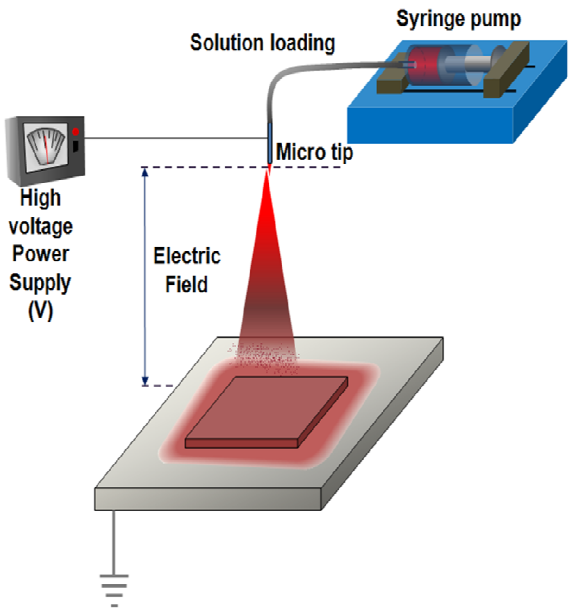


Fig 1. Electrospray Method

droplets are highly charged, up to a fraction of the Rayleigh limit. The Rayleigh limit is the magnitude of charge on a drop, which overcomes the surface tension force that leads to the drop fission. The magnitude of the charge on a droplet is given by the equation, $Q_R = 2\pi (16 \sigma_l \varepsilon_0 r^3)^{1/2}$, where σ_l is the liquid surface tension, ε_0 is the dielectric permittivity of the free space, and r is the droplet radius.

In this study, we can make binder free photoelectrodes by using electrospray. For this reason, there is no need to sinter the photoelectrode anymore. So the conversion efficiency of the DSSCs is quiet higher than other flexible DSSCs.

3. Experimental

The 10 wt % P-25(Degussa) nc-TiO_2 was dispersed in ethanol by using an ultra apex mill (Model UAM-015, Kotobuki). The dispersed solution was loaded into a plastic syringe which was connected to a high voltage power supply (BERTAN SERIES 205B). Then, the dispersed P25 solution was electrosprayed directly onto the conducting ITO-PEN substrates (10 cm x 10 cm). To prepare the hierarchically-structured TiO_2 sphere with a diameter of about 640nm, the electric field of 15 kV was applied between the metal orifice and the conducting substrate. The feed rate was controlled by a syringe pump at 35-30 μl/min. In order to form a uniform thickness in a large area, the nozzle and the substrate were placed on the motion control system with a microprocessor.

the TiO_2 electrodes were immersed into the purified 3×10^{-4} M *cis*-di (thiocyanato)-*N,N′*-bis (2,2′-bipyridyl-4-carboxylic acid-4′-tetrabutylammonium carboxylate) ruthenium(II) (N719, Solaronix) solution for 15h at room temperature.

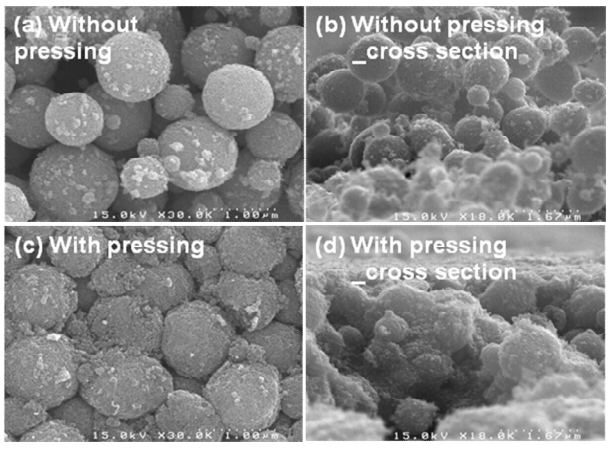
For the counter electrode, the FTO plates were drilled by microdrill, washed with 0.1M HCl solution in ethanol, and then subsequently cleaned in an ultrasonic bath with water and ethanol for 15min. A Pt counter electrode was prepared by drop casting of 5mM H_2PtCl_6 in isopropyl alcohol onto the washed FTO plates and then sintered at 400°C for 20 min under air condition. For flexible DSSCs, the Pt sputtered ITO-PEN(Peccell, Japan) were used for flexible counter electrodes. The dye-adsorbed TiO_2 electrodes were rinsed with ethanol and dried under nitrogen flow. The dye-adsorbed TiO_2 electrodes were assembled and sealed with the counter electrode using the thermal adhesive films (Surlyn, Dupont 1702, 25-μm-thick) as a spacer to produce sandwich-type cells.

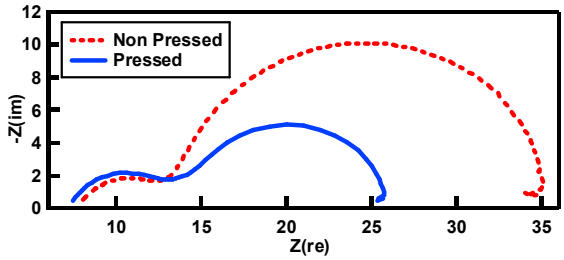
4. Results and Discussions

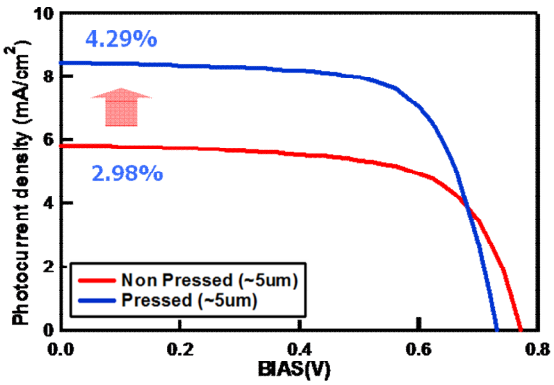
First, we can get the TiO_2 secondary sphere from electrospray process. In these secondary particles, there is no any binder or surfactant so we don't have to sinter the photoelectrode.

Through electrospray method, hierarchically structured TiO_2 particle were formed. The average diameter of HS-TiO_2 is about 600nm. But film adhesion of as-sprayed TiO_2 electrode was very pool because the secondary particles were stacked on charged particles so there is some repulsive force between TiO_2 spheres. In this case, the as-sprayed cell shows poor power conversion efficiency.

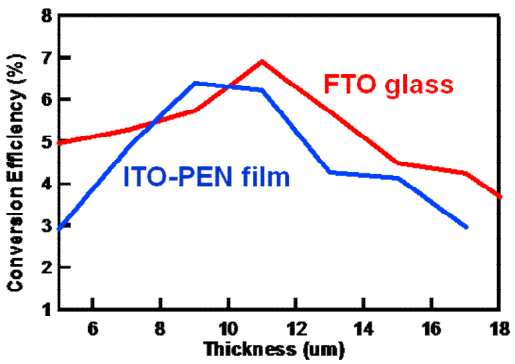
To make better electrical contact between TiO_2 spheres, compression was applied. Using lamination machine, the as-sprayed electrode was pressed at 10MPa for 10min. Through compression method, HS-TiO2 particle shape was changed and pore volume was decreased. But the photocurrent density was increased and physical adhesion was enhanced. This enhancement is due to reduction of resistance between TiO_2 particles or substrates. That result was confirmed by measuring the EIS under 1sun.



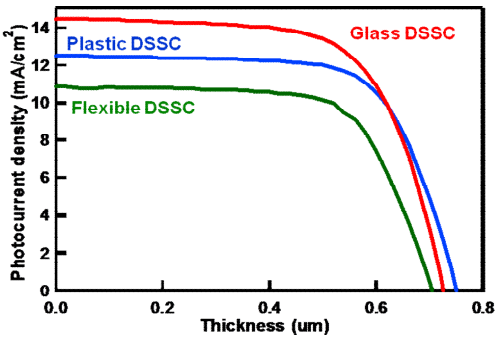




Second, we prepared various TiO_2 electrodes which have different thickness and measured the energy conversion efficiency. From this experiment, we determined that the optimum thickness of TiO_2 photoelectrode at room temperature fabrication is about 10~11μm. This value is shorter than conventional glass DSSCs. Because thermal annealing process was skipped, interconnection of HS-TiO_2 particles isn't good so diffusion length ma be reduced. Also, in case of ITO-PEN substrates, maximum efficiency appeal at 9μm. This is due to the higher resistance of ITO-PEN substrate.



By optimizing the TiO_2 photoelectrodes, the cell showed maximum efficiency at 9~11um thickness. The maximum conversion efficiency ~5% for flexible base and ~7% for glass base DSSCs.



	WE : ITO/PEN CE : Pt-ITO/PEN	WE : ITO/PEN CE : Pt-FTO glass	WE : FTO glass CE : Pt-FTO glass
Area (cm^2)	0.31	0.30	0.36
V_{oc} (V)	0.71	0.75	0.73
J_{sc} (mA/cm^2)	10.87	12.54	14.46
FF (%)	67.7	68.3	68.5
EFF (%)	5.18	6.42	6.91

5. Summary

In our study, highly efficient and binder-free TiO_2 photoelectrode for DSSCs were made using electrospray method. The conversion efficiency at room temperature fabrication was improved compression and by optimizing the thickness. Especially for flexible plastic DSSCs, the cell shows conversion efficiency ~ 5%.

References

[1] M. Gratzel. *J.Photochem.Photobiol.A Chem*. **164**, 3(2004)

[2] M. Toivola. *International Journal of Energy Research.* **30**.1145(2009)

[3] S. Uchida, *J. Photochem. Photobiol. A* **164**, 93(2004)

[4] T. Yamaguchi, H. Arakawa, *Chem. Commum.* 4767(2007)

[5] O'Regan, B. *Nature* **1991**, 353, 737-740