



# Transparent platinum counter electrode for efficient semi-transparent dye-sensitized solar cells



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## ABSTRACT

A method for fabrication of highly transparent platinum counter electrodes (CEs) has been developed based on spray coating of Pt nanoparticles (NPs) on hot substrates. This method leads to 86% reduction in Pt consumption reducing the Pt cost per peak watt of counter electrode from \$0.79/Wp down to \$0.11/Wp compared to the conventional Pt counter electrodes made by sputter deposition. The simplicity and low cost of this method provide a basis for an up-scalable fabrication process. The Pt NP layer is over 88% transparent, leading to overall transparency of 80% when incorporated with indium tin oxide/glass substrates for functional counter electrodes. This counter electrode exhibits a large surface area and high catalytic activity, comparable to that of the conventional opaque CEs. Semi-transparent dye-sensitized solar cells fabricated based on this counter electrode showed 6.17% power conversion efficiency.

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## 1. Introduction

The low production cost and potential efficiency of dye-sensitized solar cells (DSSCs) are gaining attention in the renewable energy market [1]. These types of solar cells have a potential to be integrated into window panels in homes and office buildings. This constructive-technological idea can replace special absorptive and reflective tinted films applied on window glass surface to reduce the glare and bright light transmission while utilizing unwanted non-transmitted light for power generation. DSSCs are excellent candidates for highly efficient window-compatible photovoltaics. The optically translucent nature of the photoelectrode and a variety of organic dyes with diversity of colors, tuneable absorption spectrum and possibility of highly transparent counter electrode (CE) make them the technology of choice for producing windows that can generate electricity while reducing cooling cost and still allow a portion of sunlight to illuminate the interior of buildings. For window application of DSSCs, both power conversion efficiency and overall transparency of the device should be taken into account. The degree of DSSC transparency is affected by the transparency of photoanode, counter electrode as well as electrolyte. The impact of the thickness of TiO<sub>2</sub> [2], the structure of semiconducting oxide layer [3], the size of TiO<sub>2</sub> particles, and different dye combinations [4] on transparency of DSSCs has been addressed elsewhere. Nevertheless, the transparent DSSC technology has not been matured for large-scale utilization. One of the challenges is the fabrication of low cost and transparent counter

electrode that exhibits comparable catalytic activity, series resistance, and charge transfer resistance with those of sputtered Pt CEs.

Platinum is commonly used as the counter electrode material for efficient DSSCs. Extensive research has been conducted to replace the rare and expensive Pt for DSSC application. Nevertheless, platinum is still considered as the most effective CE candidate since the DSSCs with highest reported efficiency values all utilized platinum CE prepared by sputter deposition method [5,6]. This is because of platinum's high electrocatalytic affinity towards the reduction of triiodide to iodide, strong catalytic properties and high electric conductivity [7,8]. However, sputtering method for depositing thin films is known to be inefficient as it leads to deposition of target material over the entire chamber [9, 10]. It also uses rather expensive high vacuum equipment with long delays to achieve high vacuum ( $10^{-6}$  Torr). Since the two main aspects for large-scale window-integrated DSSCs are low cost and transparency for visible light, the standard platinum sputtering deposition method needs to be replaced or avoided in order to fabricate efficient window-integrated DSSCs.

The quest for making semi-transparent DSSCs has led to the development of counter electrodes based on carbon nanomaterials [11–15], conductive organic polymers [16,17], cobalt sulphide [18], and platinum based materials [9,19,20]. To fabricate transparent counter electrodes based on platinum, numerous alternative fabrication methods such as thermal decomposition of chloroplatinic acid [21,22], chemical deposition [23], direct and pulse current electrodeposition [20], casting followed by calcination [9], and dip-coating [19] methods have been investigated. Even though these methods either replace platinum or use little amounts of platinum loading compared to the sputter deposition method, the

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transparent CEs prepared by methods listed above exhibit high charge transfer and series resistances, resulting in sizable reduction of fill factor and overall power conversion efficiency of the semi-transparent device.

This paper reports on the development of a high throughput, up-scalable and cost-effective counter electrode technology that can be utilized in window compatible DSSCs. The objective is to achieve CEs with high transparency, comparatively low charge transfer and series resistances, high fill factor and power conversion efficiency along with low Pt loadings. Our approach is based on spray deposition of Pt nanoparticles on hot substrates, which had not been applied before in semi-transparent DSSC fabrication and has a potential to replace conventional sputter deposition method. Compared to the aforementioned methods of CE fabrication, spray deposition on hot substrates is much more scalable and can be easily applied at window scales. Furthermore, it offers near 100% deposition yield on large substrates with no waste or residue to recycle. Losses at the edges can be minimized utilizing large area substrates or placing multiple substrates side by side during the spray deposition process.

## 2. Experimental details

Platinum nanoparticles (Particular GmbH) with maximum diameter of 70 nm with a 1 g/l concentration dissolved in acetone were spray-coated on a hot (80 °C) transparent conductive oxide (TCO) substrate such as fluorine tin oxide (FTO) (Hartford Glass Co. TEC-8, sheet resistance of ~8 Ω/sq and thickness of 2.3 mm) and indium tin oxide (ITO) (Delta Technologies, Ltd. sheet resistance of 5–15 Ω/sq and thickness of 1.1 mm) glass substrates. In addition to the spray coating, we also tried spin coating, dip coating, and drop casting methods to coat Platinum nanoparticles on the surface of TCO layers.

The spray deposition was conducted by a 0.2–0.3 mm airbrush spray gun enabled by compressed air. The acetone evaporated upon a contact with the hot substrate to leave Pt NPs without time for reorganization into agglomerates through capillary forces. This led to uniform deposition of the NPs over large areas. The duration of spray deposition for 1 ml of Pt NP/solvent was about 15 s with effective area coverage of 52 cm<sup>2</sup>. The spray deposited counter electrodes were annealed at 450 °C for 15 min in air. Besides the spray coating on hot surfaces, we also used spin coating, dip coating, and drop casting methods to prepare Pt NP counter electrodes. Conventional Pt counter electrodes were made by sputter deposition and used as the reference. The surface morphology of the prepared counter-electrode was analyzed by a scanning electron microscopy (SEM). Cyclic voltammetry was performed with a three-electrode system in an I/I<sub>3</sub><sup>-</sup> electrolyte solution with an Ag/Ag<sup>+</sup> reference electrode. The transmittance spectrum was measured by Shimadzu UV-VIS-2600 Spectrophotometer. Charge transfer and series resistances of a counter electrode were measured by electrochemical impedance spectroscopy (EIS) in the frequency range of 0.1 Hz to 100 kHz, and the probe voltage amplitude of 10 mV. For the EIS characterization, a symmetrical cell with an active area of 0.16 cm<sup>2</sup> was fabricated by assembling two identical counter electrodes, sealed with a 60 μm thick spacer and filled with redox electrolyte.

Two types of DSSC devices were fabricated utilizing the transparent Pt NP counter electrode and sputtered Pt counter electrode. The fabrication procedure for both types of cell was the same except for the fabrication technology of counter electrodes. DSSC with sputtered Pt counter electrode used as a reference cell. A thin layer of compact TiO<sub>2</sub> was coated over FTO glass substrate by spin coating a solution of titanium diisopropoxide bis-(acetylacetonate) in anhydrous ethanol, annealing at 115 °C for 15 min, and sintering at 475 °C for 45 min. 6 μm thick nanocrystalline TiO<sub>2</sub> (Ti-Nanoxide HT/SP, Solaronix) film was doctor-bladed over the compact layer and sintered following the above mentioned profile. A light scattering layer of TiO<sub>2</sub> (Ti-Nanoxide R/SP, Solaronix) was then doctor-bladed over the nanocrystalline layer and sintered as described above. The film was then treated with a 40 mM aqueous TiCl<sub>4</sub> solution at 70 °C for 30 min and sintered again [25]. The film was

then cooled down to 80 °C and soaked in 0.25 mM N-719 dye (Solaronix Ruthenizer 535-bisTBA) solution in acetonitrile/valeronitrile (1:1 volume ratio) for 24 h. The dye sensitized photoanode was then rinsed with acetonitrile to remove weakly bound dye molecules. The photoanode and the counter electrode were assembled using a thermoplastic sealant (Meltonix 1170-60, Solaronix). An electrolyte containing 0.03 M I<sub>2</sub>, 0.60 M 1-butyl-3-methylimidazolium, 0.10 M guanidinium thiocyanate, and 0.5 M tert-butylpyridine in acetonitrile and valeronitrile (85:15 volume ratio) was injected into the cell using a pipette.

The illuminated current density/voltage (JV) characteristics of the DSSCs were measured using simulated sunlight calibrated at 100 mW/cm<sup>2</sup> using a 300 W Newport 67005 Xe Arc lamp equipped with AM 1.5 filter. The power calibration was conducted through NREL calibrated S1133 Hamamatsu photodetectors, with an absorption window of 350–750 nm, similar to that of the fabricated DSSCs. The transmittance of the DSSCs was recorded by a Shimadzu UV-VIS-2600 Spectrophotometer.

## 3. Results and discussion

Our experimental results showed that spray deposition method on hot substrates was advantageous over the spin coating, dip coating, and drop casting methods. Spin coating suffered from very low concentration of Pt particle left on TCO surface. Dip-coating and drop-casting both resulted in non-uniform distribution of Pt nanoparticles due to agglomeration process during the solvent evaporation. Based on these experiments, only the results associated with the spray deposition process are presented.

The spray deposition of 1 mL of Pt NP solvent coats approximately 52 cm<sup>2</sup> without waste, and without need for post-processing recycling. This leads to Pt consumption of  $1.9 \times 10^{-5}$  g/cm<sup>2</sup>, compared to about 7× the Pt ( $1.38 \times 10^{-4}$  g/cm<sup>2</sup>) for a 50 nm thick sputter deposited Pt counter electrode, a typical thickness utilized for Pt CE in literature [26,27]. Assuming that RF sputtering has a 100% deposition/recycling yield, the Pt NP layer in our study uses 86% less Pt. Spray coating is also highly advantageous over spin coating and dip coating methods. Typical deposition yield for spin coating is very low, 5% to 10% [28], which require multiple post-processing recycling steps, and dip coating uses hundreds to thousands of times larger solution volumes compared to the solution volume adsorbed into the substrate making it vulnerable to cross contamination among substrates.

Fig. 1 shows photographs of a Pt counter electrode prepared by RF sputtering of Pt on ITO coated glass (a) and a Pt NP counter electrode prepared by spray deposition (b). The counter electrode prepared by sputter deposition method is completely opaque while the Pt NP counter electrode appears highly transparent in the visible light spectrum.

Fig. 2 shows the transmittance spectrum of the Pt NP counter electrode in UV/VIS/NIR regime prepared on both FTO/glass and ITO/glass substrates versus that of a sputter-coated Pt CE. The sputter-coated Pt substrate demonstrates zero transmittance in the entire spectrum. The figure also includes the transmittance spectrum of bare ITO/glass and FTO/glass substrates. The transmittance spectrum for the Pt NP samples shows a typical interference pattern due to reflection of light at the air/TCO and TCO/glass interfaces. The results suggest that deposition of Pt NP layer does not affect the interference pattern of the Pt-free substrates as the maxima and minima of the transmittance spectrum do not exhibit a wavelength shift, which indicates that Pt NP film partially covers the surface. The maximum transmittance of the Pt NP counter electrode prepared on ITO glass is around 80% at 550 nm compared to 89% maximum transparency of the ITO/glass substrate at the same wavelength. Assuming the loss of transmittance is directly related to surface coverage, the Pt NP film itself is 90% transparent and hence the surface coverage of Pt on ITO-coated glass substrate is about 10%. The maximum transmittance of Pt NP counter electrode on FTO-coated glass substrate is around 73% at 550 nm compared to 83% FTO-coated glass substrate. This means that the Pt NP film itself in this case is 88% transparent and hence the surface

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