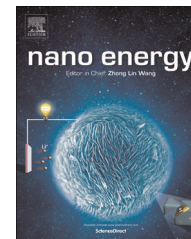


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SHORT COMMUNICATION

Size effects of platinum nanoparticles on the electrocatalytic ability of the counter electrode in dye-sensitized solar cells

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Abstract

The size effects of the Pt nanoparticles (PtNPs) on the electrocatalytic ability for the reduction of triiodide ions (I_3^-) were studied and further applied to of the counter electrode (CE) of a dye-sensitized solar cell (DSSC). First, well dispersed carbon black-supported Pt nanoparticles (CB/PtNPs) with various sizes of the PtNPs (6-2 nm) were synthesized by a CO-assisted method. The particle size and size distribution of the CB/PtNPs with various sizes of the PtNPs were verified by XRD and HR-TEM. Electrochemical surface areas (ECSA) of the PtNPs with various sizes were estimated by using CO anodic stripping voltammetry. Slurries of these catalysts were deposited as the catalytic films on ITO glasses to prepare CE for DSSCs. The size effect of the PtNPs on the electrocatalytic ability of the CEs on DSSC for the reduction of the I_3^- was studied from the perspectives of the photovoltaic performance of the cell. Cyclic voltammetry (CV) was performed to understand the reaction kinetics and electrocatalytic ability of the CEs containing the films of CB/PtNPs with various sizes of the PtNPs. The charge transfer resistances (R_{ct}) for the reduction of I_3^- at the interfaces of the CB/PtNPs films with the electrolyte were investigated by electrochemical impedance spectroscopy (EIS) and Tafel polarization plots. Intrinsic heterogeneous rate constant (k_0) and effective electroactive surface area (A_e) of the catalytic films were determined by using a rotating disk electrode (RDE). The density functional

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theory (DFT) simulation results suggested that the tri-iodine molecule (I_3) had the stronger adsorption energy (E_{ads}) among the PtNPs. Adsorption energies of I_3 on the PtNPs were estimated through CO stripping voltammetry, and the corresponding binding energies were obtained through XPS. Apparent Activation energies (E_a) of the films of CB/PtNPs were calculated from Arrhenius plots. With an optimized size of PtNPs (4 nm), the pertinent DSSC showed a solar-to-electricity conversion efficiency (η) of $9.32 \pm 0.08\%$ (Pt usage: $44.4 \mu\text{g cm}^{-2}$), which was found to be higher than that of the cell with sputtered Pt (s-Pt) on its CE ($\eta=8.43\%$, Pt usage: $64.4 \mu\text{g cm}^{-2}$).

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Introduction

In a dye-sensitized solar cell (DSSC), the counter electrode (CE) plays the roles of collecting electrons from the photoanode and reducing the triiodide ions (I_3^-) to iodide ions (I^-) in the electrolyte [1-3]. High electrocatalytic ability, excellent electrical conductivity, and high corrosion resistance are indispensable for the functioning of a good CE [4]. Pt, an expensive noble metal, has been widely used as the catalytic layer on the CE of a DSSC, because of its outstanding chemical durability and superior electrocatalytic activity for the redox reaction of I^-/I_3^- [5]. The Pt film on the CE catalyzes the reduction of I_3^- that are produced by the oxidation of I^- in the electrolyte by the oxidized dye molecules. However, Pt is expensive, rarely available, and already in demand for various chemical and electrochemical industries. By replacing the Pt or reducing the amount of Pt usage in DSSCs, the cost of fabricating the cells in industrial scale can be greatly reduced. Although alternative materials, such as conducting polymers [6-8], carbonaceous materials [9-12], and transition metallic compounds [13-16] have been used in the place of Pt, the solar-to-electricity conversion efficiency (η) of their cells is mostly less than that of the cell with Pt. On the other hand, reducing the amount of Pt usage also seems to be a choice in this direction of research. A vast number of technological applications involving catalytic and electrocatalytic processes utilize supported noble metal nanoparticles, because of their useful physical and chemical properties [17,18]. Among the noble metal nanoparticles, Pt nanoparticles (PtNPs) have attracted much attention for applications in energy conversion devices [19,20] and electrochemical sensors [21-23]. Some of literature also proposed that electrocatalytic ability of carbonaceous materials [24] and transition metallic compounds [25] can be further improved by decorating with PtNPs. Li et al. have reported a DSSC with carbon black-supported Pt on its CE, wherein they achieved an efficiency of 6.72%, which is comparable to that of a cell with a typical Pt-CE (6.63%) [26]. Poudel et al. have introduced a carbon nanoparticles with larger effective surface area as the support for PtNPs as CEs in DSSCs [27]. Xiao et al. proposed the low temperature preparation of a high performance Pt/single-wall carbon nanotube CE for flexible DSSCs [28]. Yen et al. have improved the electrocatalytic ability of a graphene-based CE in a DSSC through the attachment of PtNPs on the surface of the graphene; they achieved a higher η of 6.35% for the cell with graphene and PtNPs, compared to that of the cell with bare graphene

[29]. Guai et al. also reported that graphene-coated Pt composite film provides large conductive interface for significant improvement of Pt utilization efficiency, which in turn leads to higher conversion efficiency of the DSSC [30].

The synthesis of nanoparticles with controlled size and morphology are of fundamental and technological interest. The efforts to understand the science of nanomaterials and to exploit their beneficial properties are of equal concern. Effective control of size of PtNPs is very important to enhance their beneficial role in their catalytic applications [31,32]. Increased surface area and tailored surface of PtNPs have long been used to optimize their catalytic ability [31,33]. Song et al. have provided a simple and effective urea-assisted homogenous deposition method for preparing a uniform particle size distribution of PtNPs thin layer on conducting glass and further used as a CEs in DSSCs [34]. Lin et al. introduced 3-(2-aminoethylamino)propyl-methyldimethoxysilane as an additive to inhibit the growth of semicircle-like Pt grains on conducting glass by electrodeposition [35]. Fang et al. have compared the thickness of Pt-based catalytic layer, from 25 to 415 nm, on the conducting substrate and further applied to the DSSCs [5]. Lee et al. prepared the high transmittance Pt nanoparticles coated flexible substrate at an ambient temperature (40 °C) and employed as CE for DSSCs [36]. Chang et al. proposed that the structure of Pt with nanospheres is more suitable than that with nanosquare or nanospindle for catalyzing the I_3^- reduction, thus further influence related cell efficiency [37]. Zhang et al. proposed that electrocatalytic ability of Pt for reduction triiodide can be influenced by its mainly crystalline facets [38].

Though there have been several reports on the thickness effect and on the structural effects of Pt on its catalytic ability for the reduction of I_3^- in a DSSC, there is hardly any report that focused on the effect of its particle size on this ability. Some of the reports have indicated that the intrinsic electrocatalytic abilities of Pt for oxygen reduction [19,39], methanol oxidation [40], and CO oxidation [17] were dramatically influenced by its particle size. Maillard et al. reported that significant size effects of PtNPs had occurred in the particle size range of 1-4 nm; the effects are, a positive shift of the CO stripping peak with decreasing particle size and a pronounced asymmetry of the current transients at a constant potential [17]. These results suggested us that the electrocatalytic activity of Pt for the reduction of I_3^- in a DSSC would be affected, if the size of its nanoparticles was varied. Nevertheless, previous reports focused on the interactions of I^- and I_3^- with Pt

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