



Cost-effective alloy counter electrodes as a new avenue for high-efficiency dye-sensitized solar cells



Benlin He ^{a,b}, Qunwei Tang ^{a,b,*}, Liangmin Yu ^{a,*}, Peizhi Yang ^c

^a Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, Qingdao 266100, PR China

^b Institute of Materials Science and Engineering, Ocean University of China, Qingdao 266100, PR China

^c Key Laboratory of Advanced Technique & Preparation for Renewable Energy Materials, Ministry of Education, Yunnan Normal University, Kunming 650092, PR China

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ABSTRACT

Pursuit of cost-effective and efficient counter electrodes (CEs) has been a persistent objective for dye-sensitized solar cells (DSSCs). Aiming at reducing fabrication cost without sacrificing power conversion efficiency of DSSCs, here we report the successful design of binary Pt–Ni alloy CEs by a simple cyclic voltammetry technique. Due to the rapid charge transfer ability and electrocatalytic activity, the power conversion efficiency of the DSSC employing binary PtNi_{0.75} alloy CE has been elevated to 8.59% in comparison with 6.98% from Pt-based solar cell. The impressive results along with simple synthesis highlight the potential application of low-Pt alloys in robust DSSCs.

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1. Introduction

Fossil fuels such as coal, oil, and natural gas have occupied energy market for hundreds of years. However, the combustion of these non-renewable fuels can release sulfides, carbides and dusts which will damage ecology and pollute environment. By addressing these issues, it is a prerequisite to develop renewable, green, and environment-friendly energy resources to resolve energy and environment crisis [1]. Among various energy candidates, dye-sensitized solar cells (DSSCs) [2–5], electrochemical devices converting solar energy into electrical power honored by a high efficiency and no environmental impact, are promising solutions to global energy and environmental problems because of clean, high efficiency, good durability, and relatively simple fabrication. Since the first prototype reported by Grätzel in 1991 [6], DSSCs have attracted growing interests and great achievements have been made. However, it is still premature for their commercialization. Until now, the most limiting factor in the development of commercial DSSCs has been their cost [7,8]. The historically high prices for Pt feedstock, a traditional counter electrode (CE) material, have meant that a cell could not be fabricated at a cost low enough to compete with conventional silicon solar cells. The

task of a CE is to collect electrons from external circuit and to reduce triiodide (I₃[−]) into iodide (I[−]), therefore, an efficient CE electrocatalyst should display good charge-transfer ability and excellent electrocatalytic activity. Other candidates such as carbonaceous materials [9,10], conducting polymers [11,12], or their composites [13–15] present either modest electrocatalysis, unsatisfactory electron-conduction, or large interfacial resistance. More importantly, such CEs have a fast attenuation in electrocatalytic activity and long-term stability [16]. Therefore, it is a prerequisite to develop cost-effective but stable CEs before DSSCs are becoming a commercial reality.

Alloy materials have established themselves as the alternative electrocatalysts for fuel cell applications in the past two decades [17,18]. Aiming to increase the catalytic activity of the electrode kinetics and to lower the cost of electrocatalysts, it has been shown that alloying of Pt metal with transition metals could be an efficient route to meet the cell requirements. However, the employment of Pt-free or low-Pt alloys in DSSCs is still at preliminary stage. Previous studies in our group have revealed that binary M–Pt (where M = Co, Ru) alloys have exceptional electrocatalysis toward I[−]/I₃[−] redox couples [19,20]. However, Co species in Co–Pt alloys are easily oxidized and Ru sources are relative price high. In searching for other robust M–Pt alloys, here we report the synthesis and characterization of a new class binary Ni–Pt alloy CEs. The resultant Pt–Ni alloys show supercatalytic behavior toward I[−]/I₃[−] redox couples, allowing the rapid interconversion

* Corresponding authors. Tel.: +86 532 66781690; fax: +86 532 66782533.

E-mail addresses: tangqunwei@ouc.edu.cn (Q. Tang), yuyan@ouc.edu.cn (L. Yu).

between $I_3^- \rightarrow I^-$. The Pt–Ni alloy CEs are expected to significantly enhance the electron conduction, charge-transfer ability, and therefore power conversion efficiency of the DSSCs.

2. Experimental

2.1. Preparation of binary Pt–Ni alloy CEs

The feasibility of this strategy was confirmed by a cyclic voltammetry (CV) method: A cleaned FTO glass substrate was used as a working electrode, and the CV curves were recorded from -0.4 to $+0.3$ V and back to -0.4 V for 10 cycles. Before the measurement, the supporting electrolyte consisting of 2 mM H_2PtCl_6 , 1 mM of $Ni(NO_3)_2$, and 5 mM of HCl aqueous solution was degassed using nitrogen for 10 min.

2.2. Assembly of DSSCs

A layer of TiO_2 nanocrystal anode film with a thickness of $10 \mu m$ was prepared by a sol-hydrothermal method [21] and a layer of TiO_2 nanocrystal anode film with a thickness of $10 \mu m$ and an active area of 0.25 cm^2 was prepared by coating TiO_2 colloid onto conducting glass using a doctor blade technique, followed by sintering in air at 450°C for 30 min. Resultant anodes were further sensitized by immersing into a 0.50 mM ethanol solution of N719 dye ([cis-di(thiocyanato)–N,N'–bis(2,2'–bipyridyl–4–carboxylic acid)–4–tetrabutylammonium carboxylate]). The DSSC was fabricated by sandwiching redox electrolyte between dye-sensitized TiO_2 anode and FTO supported Pt–Ni alloy CEs. A redox electrolyte consisted of 100 mM of tetraethylammonium iodide, 100 mM of tetramethylammonium iodide, 100 mM of tetrabutylammonium iodide, 100 mM of NaI, 100 mM of KI, 100 mM of LiI, 50 mM of I_2 , and 500 mM of 4–tert–butyl–pyridine in 50 ml acetonitrile.

2.3. Electrochemical characterizations

The electrochemical performances were recorded on a conventional CHI660E setup comprising an Ag/AgCl reference electrode, a CE of platinum sheet, and a working electrode of FTO glass supported Pt–Ni alloy. The CV curves were recorded from -1.0 to $+1.4$ V and back to -1.0 V. Before the measurement, the supporting electrolyte consisting of 50 mM M LiI, 10 mM I_2 , and 500 mM $LiClO_4$ in acetonitrile was degassed using nitrogen for 10 min. Electrochemical impedance spectroscopy (EIS) measurements were also carried out on the CHI660E Electrochemical Workstation in a frequency range of 0.01 Hz \sim 10^6 kHz and an ac amplitude of 5 mV at room temperature. The resultant impedance spectra were analyzed using the Z-view software. Tafel polarization curves were recorded on the same Workstation by assembling symmetric cell consisting of Pt–Ni alloy CE|redox electrolyte|Ni–Pt alloy CE.

2.4. Photovoltaic measurements

The photocurrent–voltage (J – V) curves of the assembled DSSCs were recorded on an Electrochemical Workstation (CHI660E) under irradiation of a simulated solar light from a 100 W xenon–mercury arc lamp (CHF–XM–500 W, Beijing Trusttech Co., Ltd) in an ambient atmosphere. The incident light intensity was calibrated using a FZ–A type radiometer from Beijing Normal University Photoelectric Instrument Factory to control it at 100 mW cm^{-2} (AM1.5 calibrated by a standard silicon solar cell). Each DSSC device was measured at least five times to eliminate experimental error and a compromise J – V curve was employed.

2.5. Other characterizations

The morphologies of the Pt–Ni alloy CE were observed with a scanning electron microscope (SEM, S4800). The XRD data were

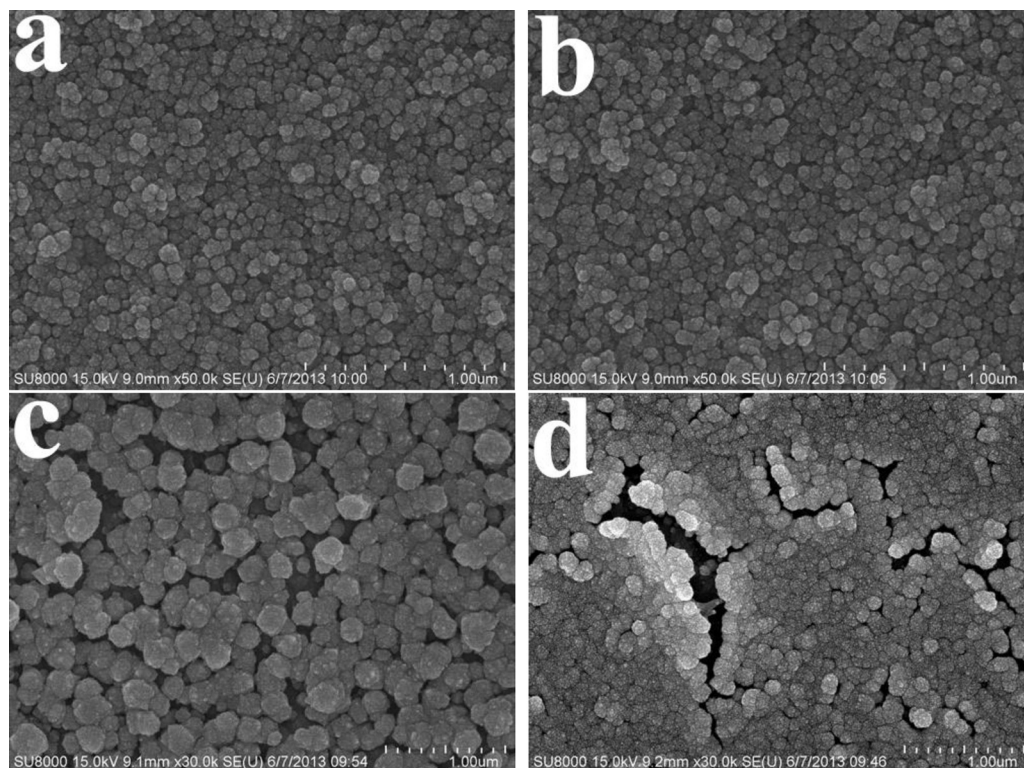


Fig. 1. Top–view SEM photographs of (a) PtNi_{0.25}, (b) PtNi_{0.50}, (c) PtNi_{0.75}, and (d) PtNi alloy CEs.

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