



Ternary platinum alloy counter electrodes for high-efficiency dye-sensitized solar cells



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ABSTRACT

Theoretical understanding of the intrinsic mechanism for enhanced electrocatalytic activity and rational design for liquid-junction dye-sensitized solar cell (DSSC) are two persistent objectives in pursuing dual-functional counter electrode (CE) electrocatalysts with excellent electrochemical activity and stability. In the current work, we launch a strategy of synthesizing ternary platinum alloys (Pt-M-Ni, M = Co, Pd, Fe) CE and study on the synergistic effects of transition metals on catalytic activity. The preliminary results demonstrate that the ternary Pt-M-Ni alloy can markedly enhance the electrocatalytic behavior toward I_3^- , yielding maximum 8.71% efficiency in the optimized DSSC platforms in comparison with 7.10% for pure Pt based device.

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1. Introduction

Advanced electrocatalysts have been carefully studied for the development of renewable energy sources such as fuel cells and dye-sensitized solar cells (DSSCs) [1–3]. Generally, Pt species are still preferred in state-of-the-art electrocatalysts owing to their reasonable electrocatalytic activity and electron-conducting ability [4,5]. However, the high expense has been a burden for commercial application of advanced DSSCs. Therefore, reduction in Pt dosage without sacrificing electrocatalytic activity and stability is a persistent objective to create promising platinum alloy electrocatalysts.

Density functional theory calculations and experimental results reveal that bimetallic platinum alloys by alloying Pt with transition metals (such as Ni, Fe, Co, Cr, Pd, etc) are more active than pristine Pt [6,7]. When applied as CEs in DSSCs, the electrocatalytic activity and stability exposure to the iodide/triiodide (I^-/I_3^-) redox electrolyte are also significantly enhanced, arising from the unusual electronic structure and the competitive reaction between transition metal and I_2/I_3^- species in the near-surface region [8,9]. It is believed that the alloy nanostructure can shift down the *d*-band center of Pt and induce strain and ligand effect due to the

different lattice constants, creating gigantic active sites for I_3^- adsorption and therefore reduction reaction. However, the ternary Pt alloys have been paid sluggish attention for DSSC devices application. To our best knowledge, ternary Pt alloys can better optimize the electronic structures for providing electrons involving in I_3^- reduction and adjust work functions to match with redox potential of liquid electrolyte by metallic synergistic effects in comparison with bimetallic alloys [10,11]. In this fashion, the ternary alloys are analogically expected to be superior catalysts in I_3^- reduction reaction and endow the functional practicability of DSSCs. In our previous works [12–15], we have synthesized binary platinum alloys by either electrochemical deposition or mild solution technique, the alloying of transition metals such as Ni, Co, Fe, Mo, and Pd with Pt can markedly enhance the catalytic activity, arising from the adjustable work function, gigantic active sites, and redistributed electron structure. However, some platinum alloys such as PtCo and PtPd do not have matching work functions to redox potential of redox electrolyte, leading to unsatisfactory catalytic activity and charge-transfer ability. In this fashion, the development of ternary alloy seems to be crucial.

In the current work, we present here the experimental realization of advanced ternary Pt alloy (Pt-M-Ni, M = Co, Fe, Pd) CE electrocatalysts with an aim of studying the electrocatalysis of multimetallic systems. Due to the extraordinary aspect of ternary alloys in tunable surface strains and compositions, the electrocatalytic activity and persistent stability are markedly enhanced in comparison with pristine Pt or binary Pt alloys. The optimized

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DSSCs yield maximal power conversion efficiencies of 8.71%, 8.28%, and 7.89% for the solar cells with Pt-Co-Ni, Pt-Pd-Ni, and Pt-Fe-Ni alloy CEs, respectively.

2. Experimental

2.1. Preparation of Pt-M-Ni and Pt-M alloy CEs

The alloy CEs were synthesized by a cyclic voltammetry method on FTO glass substrates ($12 \Omega \text{square}^{-1}$). In details, prior to electrodeposition, the FTO substrates were thoroughly rinsed acetone, ethanol, and deionized water. Subsequently, the cleaned substrate was immersed into an aqueous solution containing 3 mM H_2PtCl_6 , 1 mM NiSO_4 and 1 mM metal salt precursors (FeSO_4 , CoSO_4 , PbCl_2) at room temperature. The electrochemical co-deposition was carried out in the potential range from -0.8 to 0.6 V at a scan rate of 10mV s^{-1} for 4 cycles. As references, Pt-M bimetallic alloys and pristine Pt were also prepared at the same conditions.

2.2. Electrochemical characterizations

The electrocatalytic activities of alloy CEs were investigated employing a three-electrode system on a conventional CHI660E electrochemical workstation. Cyclic voltammetry (CV) experiments were carried out in an electrolyte solution consisting of 50 mM LiI , 10 mM I_2 , and 500 mM LiClO_4 in acetonitrile at the scan rate of 50mV s^{-1} . Tafel polarization curves and the electrochemical impedance spectra (EIS) were recorded on symmetrical dummy cells with CE|electrolyte|CE architectures. The potential bias and the ac amplitude were set at 0 V and 10 mV for EIS characterization, while the frequency range was from 0.01 to 10^5Hz . Tafel polarization curves were scanned at a scan rate of 10mV s^{-1} .

2.3. Assemble of DSSCs

A TiO_2 film with a thickness of 10 μm and active area of 0.25cm^2 was prepared according to our previous publication [16]. Afterwards, the resultant TiO_2 anode was further sensitized by immersing the film into a 0.50 mM N719 ethanol solution for

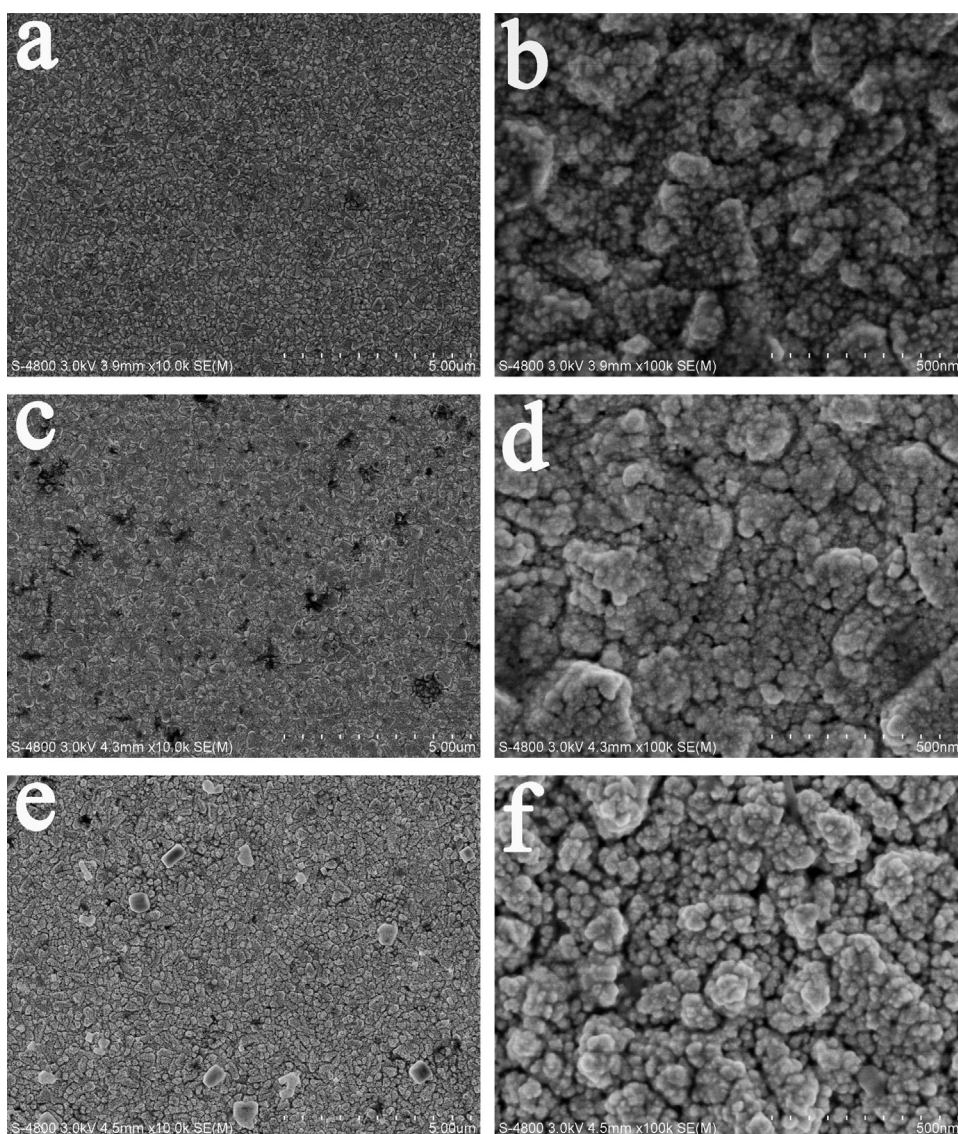


Fig. 1. The top-view SEM images of ternary Pt alloy CEs at low and high magnifications: (a & b) Pt-Co-Ni, (c & d) Pt-Pd-Ni, (e & f) Pt-Fe-Ni.

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