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# Surface plasmon enhanced ethylene glycol electrooxidation based on hollow platinum-silver nanodendrites structures

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## ABSTRACT

The surface plasmon resonance (SPR) effect on noble metals to convert solar energy into chemical has attracted a lot of interest. However, the lack of highly efficient photocatalysts is still the forbidden obstacle as well as their large-scale development. Therefore, we focus on plasmon resonance enhanced electrocatalytic oxidation of liquid fuel employing photocatalysts to develop unique hollow platinum-silver (Pt-Ag) nanocrystals. The hollow Pt-Ag is formed of nanodendrites (Pt<sub>1</sub>-Ag<sub>1</sub>) which display a great enhancement in catalytic activity towards ethylene glycol oxidation with the mass and specific activity found to be: 7045.2 mA/mg and 14.1 mA/cm<sup>2</sup>, respectively. This is due to: the SPR effect, efficient electronic distribution and synergistic properties, together with the unique hollow dendritic nanostructures. Impressively, the SPR effect also induces the optimum Pt-Ag nanocatalyst under visible light irradiation conditions to display 1.7-fold enhancements in catalytic activity compared to that under dark conditions. In addition, 6.2 and 7.0-fold enhancements were obtained when the optimized Pt-Ag was employed as photoelectrocatalyst compared to the commercial Pt/C. Therefore, we present a unique catalyst which produces a high catalytic activity and long-term stability compared to those previously reported. More importantly, we also introduce a promising approach towards the designing of a plasmonic metal nanocatalyst with ideal nanostructures for liquid fuel oxidations.

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## 1. Introduction

The production and conversion of solar energy have received much attention in recent years [1]. The use of different photocatalysts to directly convert solar energy into chemical has attracted a great interest. Although most of the photocatalysts are semiconductors, it has also been well demonstrated that plasmonic nanostructures of noble metals such as gold and silver can also serve as efficient photocatalysts [2,3]. Plasmonic metallic nanostructures are featured with their strong interactions with resonant photons via exciting the SPR [4,5]. In addition, the SPR can be characterized by the collective oscillation of valence electrons induced by resonance photons [6,7]. Therefore, the investigation of SPR induced photoelectrocatalytic effect based on plasmonic metallic nanostructures

is of vital significance for exploring highly efficient photoelectrocatalysts for electrocatalytic oxidation reactions.

It has been demonstrated that controlling the shape and size of the plasmonic metallic nanocrystals can regulate their properties to meet the requirements of different reactions, for which the SPR is largely dependent on the size and shape of nanocrystals [8]. For instance, the localized surface plasmon resonance (LSPR) wavelengths of Au can be efficiently tuned from the visible light to near-infrared region by controlling the size and morphology of Au nanoparticles [9–11]. The SPR of Ag can also be controlled by adjusting the shape and size of the Ag nanoparticles [12]. Accordingly, by manipulating the compositions, sizes and morphologies of the plasmonic metals, it is possible to engineer the nanocatalysts with enhanced photoelectrocatalytic activity and durability towards various chemical reactions [13–15].

The distinctive capability of plasmonic metallic nanostructures to concentrate scatter electromagnetic fields, biosensing, or convert the energy of photons into chemical energy make them suitable for various applications [16–18]. The SPR effects have been widely reported to enhance the catalytic activities of a series of reactions

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such as: water splitting [10], reduction of CO<sub>2</sub> [13] and degradation of organic molecules [8] among others. During these processes, the SPR effect of plasmonic metallic nanostructures plays a significant role in the substantial enhancement of catalytic activities [18–20]. However, the applications of the photoelectrocatalytic processes in liquid fuel oxidation reactions induced by the SPR effects have remained unexplored.

Therefore, we herein report a facile Pt-Ag hollow nanodendrites synthesis. And the as-prepared hollow Pt-Ag nanodendrites displayed a significant enhancement in the electrocatalytic performances compared with the commercial Pt/C catalyst. These outstanding properties are attributed to the unique hollow dendritic structure as well as synergistic and electronic effects between Pt and Ag. More importantly, we took advantage of the plasmonic Ag combination with the SPR to enhance the catalytic activity towards the oxidation of ethylene glycol (EG) by Pt-Ag hollow nanodendrites under visible light illumination. The resulting Pt<sub>1</sub>Ag<sub>1</sub> nanocatalysts achieved 1.7-fold enhancements in mass and specific activities, showing a class of ideal photoelectrocatalysts towards liquid fuel oxidation.

## 2. Experimental section

### 2.1. Chemicals

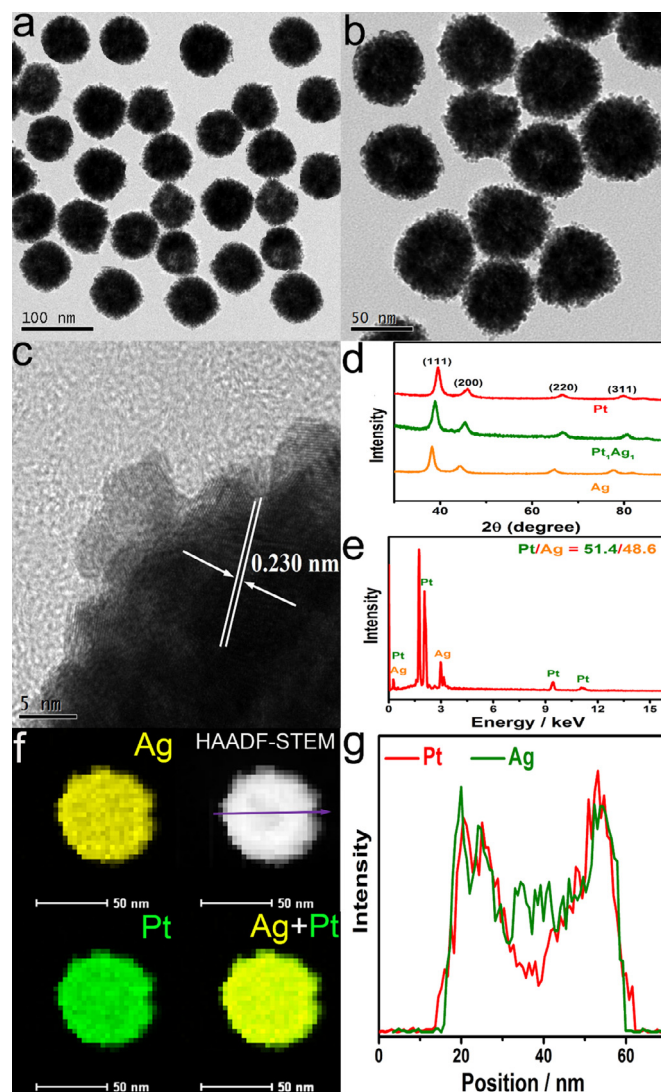
Silver nitrate (AgNO<sub>3</sub>, 99%), chloroplatinic acid (H<sub>2</sub>PtCl<sub>6</sub>, 99.9%), hexadecyltrimethylammonium chloride (CTAC, 99%), L-ascorbic acid (AA, C<sub>6</sub>H<sub>8</sub>O<sub>6</sub>, 99%), potassium hydroxide (KOH), dehydrate trisodium citrate (Na<sub>3</sub>C<sub>6</sub>H<sub>5</sub>O<sub>7</sub>·2H<sub>2</sub>O), acetone (C<sub>3</sub>H<sub>6</sub>O, 99%), ethanol (C<sub>2</sub>H<sub>5</sub>OH, A.R. grade, > 99.5%) and EG ((CH<sub>2</sub>OH)<sub>2</sub>, A.R. grade, > 99.5%) were purchased from Sinopharm Chemicals Reagent Co., Ltd, China. Doubly distilled water was used throughout the experiments.

### 2.2. Preparation of Pt-Ag hollow nanodendrites

The Ag seeds were firstly prepared according to the method described by our previously reported work [21]. In the standard synthesis of Pt-Ag hollow nanodendrites, 1.4 mL H<sub>2</sub>PtCl<sub>6</sub> (7.7 mM) was firstly injected into 10 mL aqueous solution in a glass vial, which contained 10 mg CTAC. After vigorous stirring for 10 min, 4 mL of AA (10 mg) was added dropwise to the above solution to serve as the reducing agent. After continuous reaction for 3 min, 5 mL of freshly-prepared Ag seeds were dropped into this aqueous solution. Then, the aqueous solution was violently shaken and capped, and then sonicated at ambient temperature for another 2 h. For comparison, the Pt<sub>1</sub>Ag<sub>0.5</sub> and Pt<sub>1</sub>Ag<sub>1.5</sub> hollow nanodendrites were also prepared by tuning the amounts of Ag seeds to 2.5 and 7.5 mL, respectively, while keeping the other reaction parameters unchanged.

### 2.3. Characterizations

In this work, we firstly used the Tecnai G220 (FEI America) to investigate the morphology and structure of the samples. After that, the FEI Tecnai F20 transmission electron microscope operated at an accelerating voltage of 200 kV was employed to record the scanning transmission electron microscopy (STEM), high-magnification transmission electron microscopy (HR-TEM) and EDX elemental mapping images. An X' Pert-Pro MPD diffractometer (Netherlands PANalytical) with a Cu K $\alpha$  X-ray source ( $\lambda = 1.540598 \text{ \AA}$ ) was used to obtain the powder X-ray diffraction (PXRD) patterns of the samples. The compositions and elemental valences were characterized by X-ray photoelectron spectroscopy (XPS), which was performed on a VG Scientific ESCALab 220XL electron spectrometer using 300 W Al K $\alpha$  radiation.



**Fig. 1.** (a, b) Representative TEM images of Pt<sub>1</sub>Ag<sub>1</sub> hollow nanodendrites with different magnifications. (c) HRTEM image of an individual Pt<sub>1</sub>Ag<sub>1</sub> hollow nanodendrite. (d) XRD patterns of Pt<sub>1</sub>Ag<sub>1</sub> hollow nanodendrites, Pt and Ag. (e) The TEM-EDX elemental mapping, HAADF-STEM and corresponding (f) line-scan of individual Pt<sub>1</sub>Ag<sub>1</sub> hollow nanodendrites for element distribution analyses.

### 2.4. Electrochemical measurements

Cyclic voltammetry (CV) was employed to investigate the electrocatalytic performances carried out in a standard three-electrode system, which included a Pt wire, glassy carbon electrode (GCE), a saturated calomel electrode (SCE) and as counter, working and reference electrodes, respectively. In the standard preparation of catalysts-coated electrode, the catalysts should firstly be re-dispersed in a mixture solvent containing Nafion and isopropanol and to form a homogeneous catalyst ink by sonicating for 30 min. Next, five microliters of the dispersion was transferred onto the GC electrode and dried naturally. The CV was operated using in 1 M KOH + 1 M EG at the potential ranging from −0.9 to 0.3 V with the sweeping rate of 50 mV/s. Every time before examination, the GCE was polished with alumina powder and then rinsed with doubly deionized water and ethanol for several times. Besides, the measurements of successive CVs of 500 cycles were also conducted for investigating their long-term durability. To achieve precise results, all the electrochemical measurements were repeated five times. The photocurrent responses of the modified electrode

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