



## Full Length Article

# Facile synthesis of jagged Au/Ir nanochains with superior electrocatalytic activity for oxygen evolution reaction



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## ABSTRACT

Au/Ir bimetallics are not easy to obtain because alloy phase diagrams are not available for these material combinations (Ronald et al., 2004). Herein, the Au/Ir nanochains (Au/Ir NCs) are synthesized *via* one-pot, hydrothermal method. By virtue of jagged surface with abundant atomic defects, the Au/Ir NCs are demonstrated much more active than commercial IrO<sub>2</sub> electrocatalysts for oxygen evolution reaction under alkaline media. The potential of the Au/Ir NCs sustains 1.53 V vs reversible hydrogen electrode (RHE) with no obvious decrease under current density at 10 milliamps per square centimeter, while that of IrO<sub>2</sub> increase to 1.65 V vs RHE after stability tests for ca. 32 h. The durability as well as the activity of Au/Ir NCs is attributed to the electronic interaction between Au and Ir, which is confirmed by X-ray photoelectron spectroscopy (XPS).

## 1. Introduction

China and other countries all over the world are facing increasingly serious energy crisis. Hydrogen energy is promising way to solve this crisis. To accelerate commercial process of hydrogen production, the activity, cost and other important factors of catalysts must be taken into consideration. Oxygen evolution reaction (OER), as one of half reactions of water splitting, is crucial reaction for hydrogen generation [1–14]. By far, iridium-based materials as highly efficient and robust OER catalyst in alkaline media have aroused increasing attention of researchers [15–17]. Exploring iridium-based catalysts for alkaline OER are widely recognized and continuously studied [18–27]. However, reducing the use of these state-of-the-art catalysts and enhancing their catalytic activity are still major challenges owing to its costs and reserves. Bimetallic catalysts not only enhance catalytic activity but also maximize the utilization of catalysts. For instance, Ir-Ni [28], Ir-Pi [29], and Ir-Co [30] have been proved improved activity for OER compared with single component. Nevertheless, there is still enormous room for improvement.

Recently, Au was found capable of enhancing the activity for OER in alkaline media because of the induced electronic effects, which can modify its local electronic environment, thereby accelerate the reaction of OER intermediates [1,31]. Up to date, plenty of Au-related catalysts for OER are tested in alkaline condition [1,31–35], this might because

Au is not stable in acid media, and dissolve during oxide formation and oxygen evolution [36,37]. In other reactions, Au also play an important role on stability of catalyst [38–40]. Adzic's group reported the Pt monolayers on Pd–Au electrocatalysts dramatically enhanced the activity and stability over 100,000 cycles [39]. Nevertheless, Au-induced Ir-based OER catalysts are not widespread concerned, research on Au/Ir systems is limited [41–43]. Sun et al. reported an Au–Ir bifunctional electrocatalysts supported with carbon black which have improved ORR and comparable OER activity [43]. However, the activity and durability are barely satisfactory, especially for OER under alkaline media. Therefore, enhancements of activity and durability by Au/Ir catalysts are a viable alternative.

It is generally agreed that catalytic activity is mainly depended on morphology and electronic structure of electrocatalysts. Recently, the jagged Pt nanowires with undercoordinated surface atoms was reported by Duan et al. [44]. The advantages of jagged features manifest the enlarged specific surface area and abundant surface unsaturated atoms [45]. On the other hand, the surface electronic structure of catalytic heavily relies on the unoccupied d-electron states [39], which affects corrosion resistance of electrocatalysts. By inducing a second metal (*e.g.* Au), the electronic structure of Ir-based catalysts can be significantly modified. However, Au/Ir bimetallics are hard to be obtained because alloy phase diagrams are not available for these material combinations [46]. As far as we know, typical jagged Au/Ir catalysts *via*

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electrochemical dealloying or wet chemical synthesis are still not developed yet. Therefore, it should be an interesting topic to investigate Au-Ir electrocatalysts for OER in alkaline water electrolysis.

Here, bimetallic Au/Ir nanochains (Au/Ir NCs) electrocatalysts with dramatically improved activity and durability are prepared by a facile hydrothermal method. Specifically, the OER overpotential is decreased to 0.3 V vs RHE at  $10 \text{ mA cm}^{-2}$ , which is more negative than that of commercial  $\text{IrO}_2$ . Furthermore, the Au/Ir NCs are also more stable than  $\text{IrO}_2$  under  $20 \text{ mA cm}^{-2}$ . The Au/Ir NCs show many advantages for remarkable catalytic activity: (i) The jagged surface expose large surface area and abundant low coordinated atoms, thus provides large number of active sites. (ii) The Au decrease the resistance of Au/Ir NCs. The resistance of Au/Ir NCs is lower than that of Ir particles and  $\text{IrO}_2$ ; and Au increase the electron transfers of Au/Ir NCs. (iii) The Au enhance activity by induced electronic effects. The electronic interactions between Au and Ir change the surface electronic structure, which also greatly improves the electrocatalytic activity.

## 2. Experimental

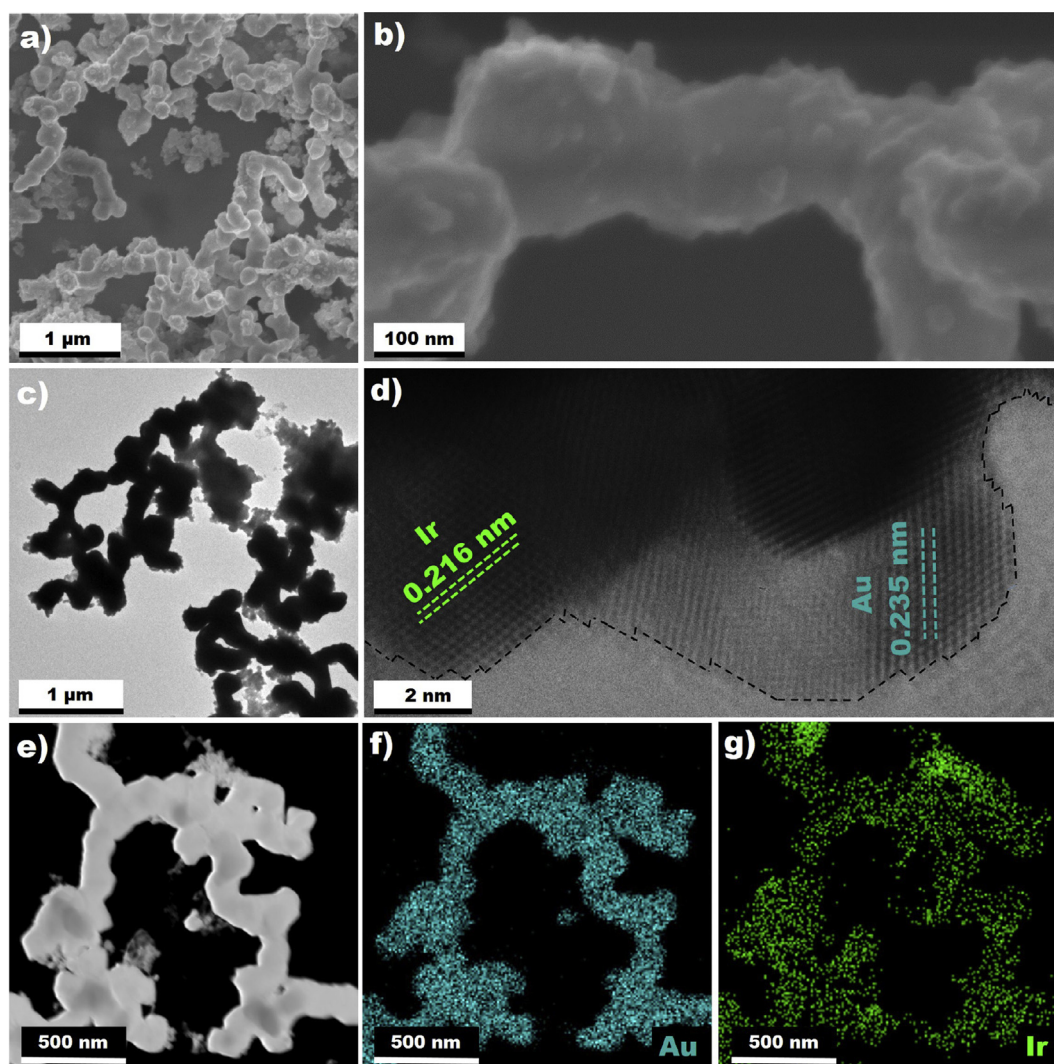
### 2.1. Synthesis

The phosphor samples were synthesized by one pot hydrothermal reaction with 2 mL 0.5 M gold(III) chloride trihydrate ( $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$ ,

Aladdin) and 2 mL 0.5 M iridium chloride hydrate ( $\text{IrCl}_3 \cdot 3\text{H}_2\text{O}$ , Aladdin) as precursors, 1 mL 0.5 M potassium bromide (KBr, Sinopharm) as capping agent, and 1 mL formaldehyde ( $\text{HCHO}$ , 37–40 wt%, Sinopharm) as reducing agent. These reagents were mixed homogeneously in a sealed Teflon-lined autoclave, subsequently moved to high temperature oven heated at  $180^\circ\text{C}$  for 10 h. The Au particles were systemized by same reaction condition with  $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$  as procedure. Similarly, the Ir particles were systemized by same reaction condition with  $\text{IrCl}_3 \cdot 3\text{H}_2\text{O}$  as procedure. The products were collected by centrifugation at 10,000 rpm for 3 min and washed several times with water. Then the collected products were dried at  $40^\circ\text{C}$  for 10 h. Next, XRD, SEM, TEM, XPS, and electrochemistry measurements were carried out. The Au/Ir composites were carry out under same condition without KBr.

### 2.2. Characterization

X-ray diffraction (XRD) patterns of the samples were investigated by a Bruker AXS D8 Advance X-ray diffractometer (40 kV, 40 mA,  $\text{Cu-K}\alpha 1$ ,  $\lambda = 1.5405 \text{ \AA}$ ,  $1.2^\circ \text{ min}^{-1}$ ) in the  $2\theta$  range from  $30^\circ$  to  $95^\circ$ . The surface morphologies, particle size and composition of the sample were analyzed by scanning electron microscopy (SEM, Quanta 250, FEI), transmission electron microscopy (TEM, Tecnai G2 F20 S-TWIN, FEI) with an energy-dispersive X-ray (EDS, TEAM Apollo XLT, EDAX) accessory.



**Fig. 1.** The (a) SEM image and (b) enlarged area SEM image of Au/Ir NCs. The (c) TEM image and (d) high resolution TEM image of Au/Ir NCs. The (e) HADDF-STEM image, and corresponding (f) Au element and (g) Ir element maps.

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