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Truncated octahedral platinum–nickel–iridium ternary electrocatalyst for oxygen reduction reaction

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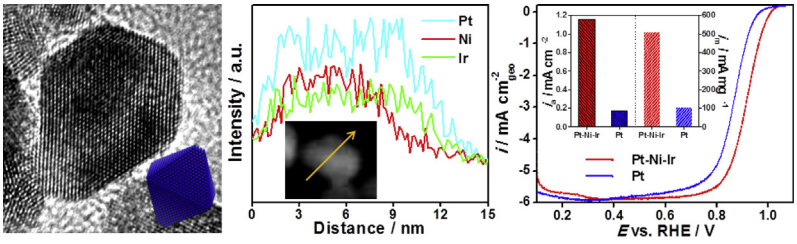
HIGHLIGHTS

- A Pt–Ni–Ir ternary nanocrystal was synthesized through a facile approach.
- The truncated octahedral ternary nanoparticle has a Ni-scarce surface.
- High catalysis and stability over oxygen reduction reaction were obtained.

GRAPHICAL ABSTRACT



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ABSTRACT

In present work, truncated octahedral Pt–Ni–Ir ternary nanoparticles with an average size of 10 nm and a Pt/Ni/Ir atomic ratio of 55/29/16 are synthesized through a facile method. Structural measurements show that the nanoparticles are surface Ni-scarce Pt–Ni–Ir ternary nanostructures. The mass activity and specific activity of the ternary catalyst are 511 mA mg^{-1} (Pt) and 1.03 mA cm^{-2} at 0.9 V (vs. reversible hydrogen electrode, RHE), respectively, which are nearly 4.8 and 6.0 times larger than that of the commercial Pt/C catalyst (106 mA mg^{-1} and 0.17 mA cm^{-2}). After duration test of 10,000 cycles at the potential range from 0.6 to 1.0 V, the ternary catalyst preserved 66% and 73% of its initial mass activity and specific activity, respectively, which are 9 and 7.6 times that of the commercial Pt/C catalyst (37.2 mA mg^{-1} and 0.0998 mA cm^{-2}, respectively) after the same durability examination. The dramatic enhancement of Pt–Ni–Ir ternary catalyst can be attributed to its morphology, composition and nanostructure. Our research suggests that Pt–Ni–Ir ternary catalyst could be an excellent candidate for ORR. Our investigation might also spur increasing attention to tailoring morphology and composition of ternary or multi-metallic nanostructure with high electro-catalytic performance.

1. Introduction

The exploration of nanomaterials with efficient catalytic activity over oxygen reduction reaction (ORR) and high stability in acid media is still a challenging task gbecause that ORR is widely involved in many electrochemical devices [1]. Pt-containing catalysts has been universally recognized as the most effective ORR catalysts due to their ever increasing catalytic activity and good stability in acid media [2]. However, the high cost and scarcity limit its prospective applications. In response to these issues, intensive efforts have been devoted to exploiting high performance electro-

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catalysts with less Pt content and higher utilization. One of the effective approaches is alloying with transitional metals, which can not only increase the intrinsic activity of Pt constituent but also reduce Pt content by diminishing the number of buried non-functional Pt atoms [3,4]. Accordingly, significant progress has been made by the rational tailor over composition of Pt binary nanocrystals [5–7]. Morphologies are as important as compositions in the optimization of chemical and physical properties, in turn the catalytic activities, because the exposed crystalline facets are compositions and morphologies dependent and the ORR is surface structure sensitive. Thus, much advance has been achieved by the control over morphologies and compositions of Pt alloys. More recently, octahedral and truncated octahedral Pt–Ni have been extensively studied following the prior report that a single-crystal alloy of Pt_3Ni enhanced the ORR activity by two orders of magnitude vs. Pt/C [8–16]. A nano-segregated Pt (111) skin structure on the catalysts surface was found to account vitally for the dramatic enhancement. Octahedrally shaped or truncated octahedral Pt alloys are stable in electrochemical condition due to the close-packed arrangement of surface atoms. Coincidentally, Pt based electrocatalysts with more lower index facets of {111} are normally recognized to display higher activity over ORR [6]. Up to now, very little work has been dedicated to the synthesis of Pt base ternary alloys ORR catalysts [17–22], due to the difficulty for either the formation of alloys at atomic level or the control over morphologies of multi-metals systems. Iridium is electrochemically stable in acid media (Ir^{3+}/Ir 1.16 V), and was successfully used to improve the stability of Pt-based catalysts [23–26]. The tight chemisorption of oxygenated species on Pt surface has been viewed as a remarkable block to ORR, which can be greatly weakened by alloying with Ir. Because that the intense chemisorption of OH and O on Ir will cause lateral repulsion, which can decrease the intensity of OH and O adsorption on the adjacent Pt atoms [27–29]. Therefore, the Pt–Ni–Ir ternary nanocrystals are expected to process high catalytic activity over ORR and good stability in acid media. The synthesis and morphologies control of Pt–Ni–Ir nanostructures are still cost and far from trivial.

Herein, we demonstrate a facile synthesis for Pt–Ni–Ir trimetallic nanocrystals, which involves the simultaneous reduction of $Pt(acac)_2$, $Ni(acac)_2$, and $Ir(acac)_3$ in a sealed autoclave at 210 °C. Transmission electron microscopy (TEM) measurements show that the trimetallic nanoparticles are truncated octahedral in shape with an average size of 10 nm. Energy dispersive X-ray spectroscopy (EDS) and inductively coupled plasma mass spectrometry (ICP-MS) tests demonstrated that the material is composed of Pt, Ni and Ir with an atomic ratio of 55/29/16. Scanning transmission electron microscopy (STEM) and X-ray photoelectron spectroscopy (XPS) analysis revealed that the ternary nanoparticles have a Ni-scarce surface. Electrochemical measurements demonstrated that the carbon supporting ternary Pt–Ni–Ir catalysts reported herein are promising ORR catalysts with high catalytic activity and excellent stability in acid media.

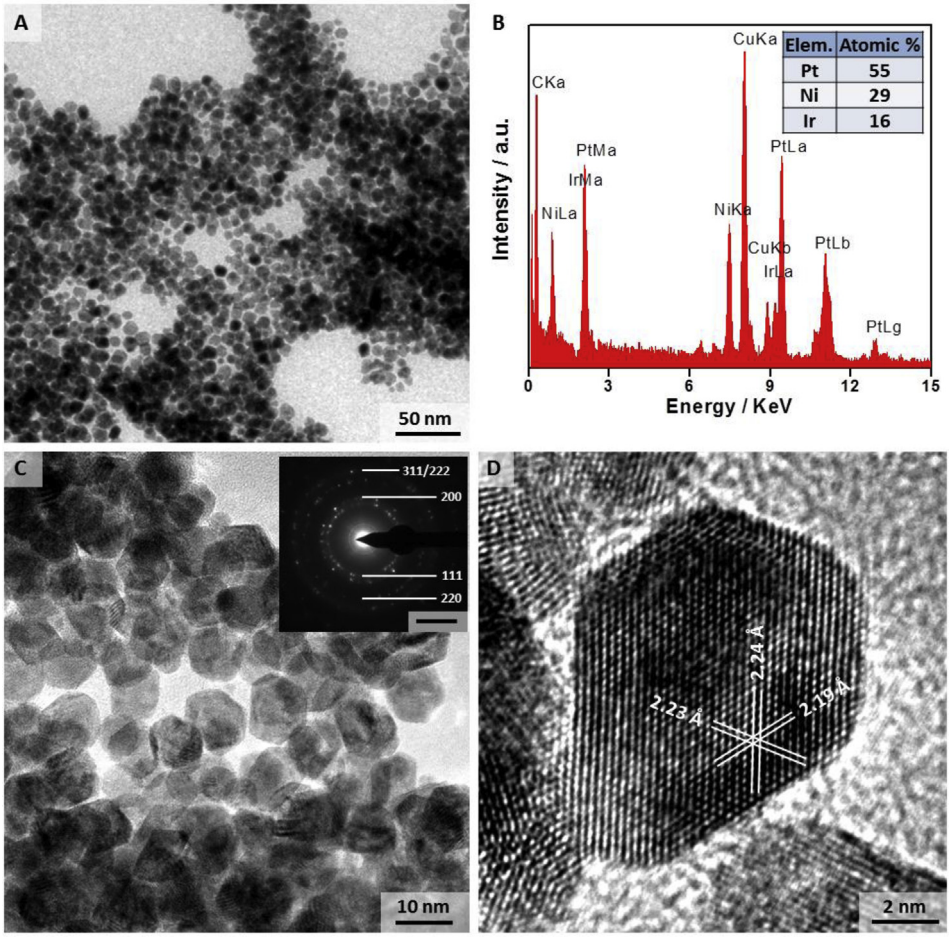

Fig. 1. (A) TEM image and (B) the corresponding EDS spectra of the as made particles. (C) TEM image and (inset) the corresponding SAED pattern of the product. (D) HRTEM image of a single particle. The scale bar in inset of (C) is 5 × 1/nm.