

## Full Length Article

Size effect of Pt co-catalyst on photocatalytic efficiency of g-C<sub>3</sub>N<sub>4</sub> for hydrogen evolutionYunqing Zhu<sup>a,b,\*</sup>, Tian Wang<sup>a</sup>, Tao Xu<sup>c,\*</sup>, Yingxuan Li<sup>a</sup>, Chuanyi Wang<sup>a,\*</sup><sup>a</sup> School of Environmental Science and Engineering, Shaanxi University of Science & Technology, Xi'an 710021, PR China<sup>b</sup> Zhejiang Wenzhou Research Institute of Light Industry, Wenzhou 325804, PR China<sup>c</sup> Department of Chemistry and Biochemistry, Northern Illinois University, 1425 W. Lincoln Hwy. DeKalb, IL 60115-2828, USA

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## ABSTRACT

Graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) with Pt co-catalyst was synthesized by in-situ photoreduction and used as a visible light photocatalyst. The size of Pt co-catalyst can be controlled from single atoms to nano-clusters by the induced amount of Pt precursor. The results indicated that compared with nano-clusters, the Pt<sub>0.1</sub>-CN (with 0.1 wt% Pt loading amount) which is characterized as single-atom Pt exhibits a pronounced photocatalytic hydrogen evolution capability. For Pt<sub>0.1</sub>-CN with single-atom Pt as co-catalyst, the H<sub>2</sub> generation is up to 473.82 μmol mg<sub>pt</sub><sup>-1</sup> under visible light irradiation (λ > 420 nm). The enhanced photocatalytic performance is mainly attributed to the synergistic effect of high light adsorption efficiency, effective charge separation, and high dispersed active sites of Pt atoms. The results of this work highlighted that loading g-C<sub>3</sub>N<sub>4</sub> with Pt single atoms will achieve a maximum utilization efficiency of Pt atoms and an improvement photocatalytic performance.

## 1. Introduction

Photocatalytic water splitting to produce hydrogen fuel by harnessing sunlight holds particular promise as the process is economic and environmentally friendly [1,2]. TiO<sub>2</sub> is the most widely investigated photocatalyst [3], but the wide-band gap and limited optical absorption restricted its practical application [4,5]. To develop the optimal photocatalysts being adequately efficient, earth abundant, stable, non-toxic and capable of harvesting visible light is still a major issue for photocatalytic water splitting [6].

g-C<sub>3</sub>N<sub>4</sub> consisted by tri-s-triazine units has been considered as a metal-free photocatalyst [7–9], which can be synthesized via a simple poly-condensation method with different cheap N-rich precursors [10]. The g-C<sub>3</sub>N<sub>4</sub> exhibits a medium band gap (E<sub>g</sub> ≈ 2.7 eV), high stability and “earth-rich” nature. All these excellent properties make g-C<sub>3</sub>N<sub>4</sub> an outstanding photocatalyst under visible light irradiation. The g-C<sub>3</sub>N<sub>4</sub> has been successfully used in photocatalytic water splitting since its band energy is high enough for photocatalytic H<sub>2</sub> evolution [11,12]. Nevertheless, pristine g-C<sub>3</sub>N<sub>4</sub> used as a main catalyst cannot achieve a high efficiency for water splitting [13,14], because the fast recombination speed of photogenerated charges, and low electronic conductivity restrict its photocatalytic efficiency for hydrogen production [15]. Hence, various approaches have been proposed to overcome

these glitches [16–19], including design of heterojunctions, metallic and non-metallic doping, construction of C or N defects, and morphology control. Among the abovementioned approaches, the choice of noble metals as co-catalyst is a key remedy [20,21]. Pt is the most efficient co-catalyst to enhance the charge separation in the photocatalytic process [22–24]. However, the present morphology of Pt co-catalyst is the most important issues that affect the photocatalytic efficiency [25]. It was known that the size reduction of Pt cluster will cause (1) an increase of low-coordination environment of metal centers [26], (2) quantum size effects [27], and (3) strong metal-support interactions [28], especially when the Pt size is reduced to single atom [29].

Inspired by the abovementioned excellent works, we prepared Pt modified g-C<sub>3</sub>N<sub>4</sub> materials with different Pt contents and the effect of Pt size (from single atom to clusters) on the photocatalytic H<sub>2</sub> evolution was studied systematically. The Pt<sub>0.1</sub>-CN sample (loading with 0.1 wt% Pt), in which the Pt species was characterized as Pt single atoms, displayed the highest photocatalytic H<sub>2</sub> evolution performance (473.82 μmol mg<sup>-1</sup>pt) under visible light irradiation among the Pt modified g-C<sub>3</sub>N<sub>4</sub> materials with different Pt contents. We demonstrated that the remarkably increased photocatalytic performance of Pt<sub>0.1</sub>-CN was mainly ascribed to the synergistic effect of high light adsorption efficiency, effective charge separation, and high dispersed active sites of

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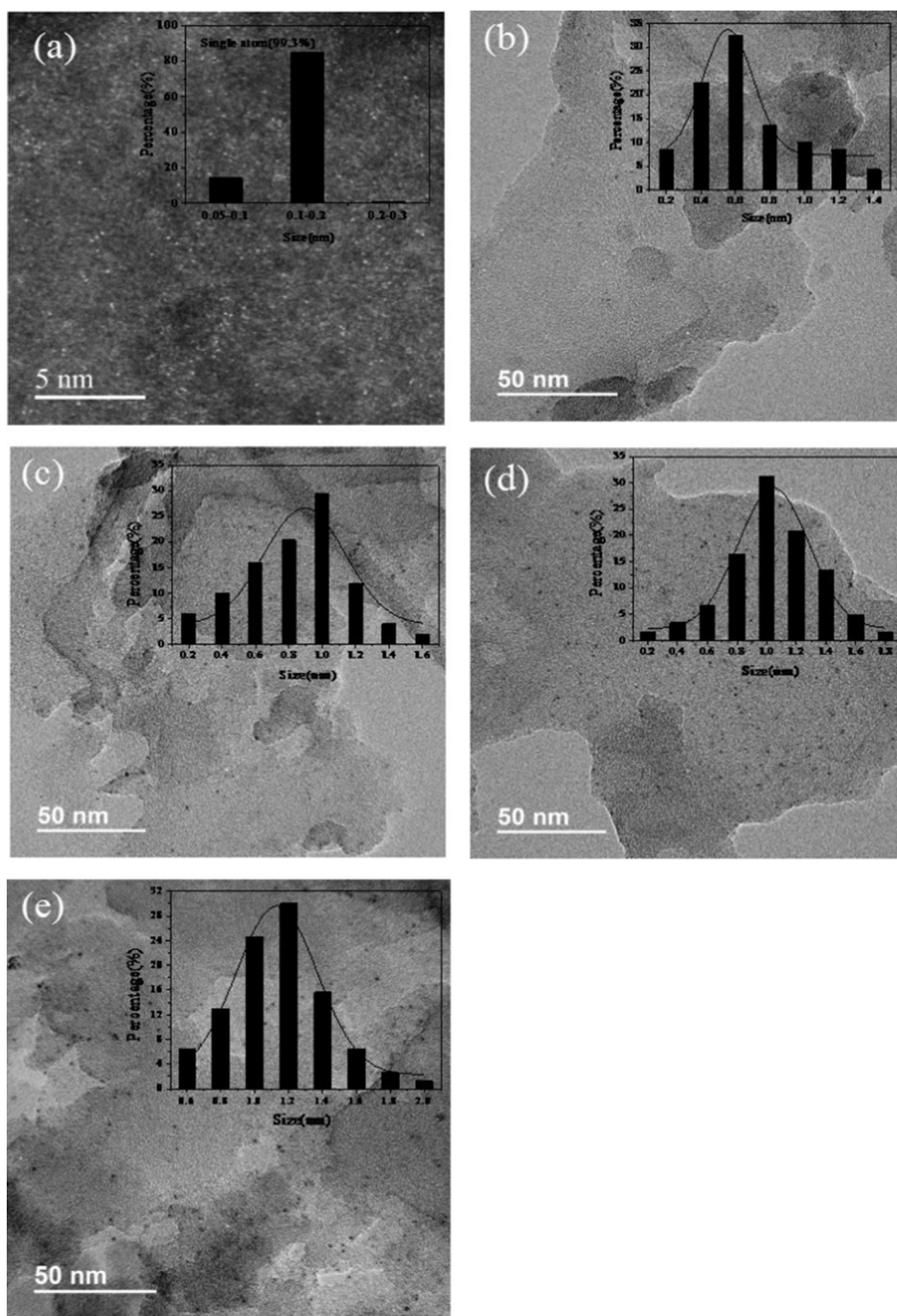


Fig. 1. Aberration-corrected-STEM images of Pt<sub>0.1</sub>-CN (a) and HRTEM of Pt<sub>0.3</sub>-CN (b), Pt<sub>1</sub>-CN (c), Pt<sub>2</sub>-CN (d), Pt<sub>3</sub>-CN (e).

Pt atoms.

## 2. Experimental

### 2.1. Materials synthesis

First, the g-C<sub>3</sub>N<sub>4</sub> powder was fabricated by poly-condensation method with urea as precursor which is consistent with previous report [30]. Typically, urea (10 g) was added in to a covered crucible and sintered at 550 °C with a heating rate of 3 °C/min for 4 h, then the muffle was cooled to 250 °C, and again heated to 550 °C and hold for 2 h.

Second, g-C<sub>3</sub>N<sub>4</sub> powder (0.50 g) was dispersed in H<sub>2</sub>O (400 mL) and treated by a 100 W ultrasonic crusher for 60 min to obtain a g-C<sub>3</sub>N<sub>4</sub>/

H<sub>2</sub>O suspension. Pt/g-C<sub>3</sub>N<sub>4</sub> catalysts were fabricated by in situ photo-reduction of H<sub>2</sub>PtCl<sub>6</sub>·6H<sub>2</sub>O. In detail, H<sub>2</sub>PtCl<sub>6</sub>·6H<sub>2</sub>O aqueous solution (1 × 10<sup>4</sup> mg/L) was added drop-wise into the above g-C<sub>3</sub>N<sub>4</sub>/H<sub>2</sub>O suspension, and irradiated under visible light (λ > 420 nm) for 4 h. After washing and drying, the final products were labeled as Pt<sub>x</sub>-CN, where x represents the actual mass percentage of Pt in the products.

### 2.2. Characterization

Aberration-corrected-STEM (ac-STEM) was performed on a JEM-ARM300F at an accelerating voltage of 60 kV. High resolution transmission electron microscopy (HRTEM) images were recorded on a JEOL-JEM 2010 electron microscope. X-ray diffraction (XRD) characterization was performed using a D/max2200PC diffractometer via

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