Simultaneous Exfoliation and Modification of Graphitic Carbon Nitride Nanosheets

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This study demonstrates the simultaneous exfoliation and modification of graphitic carbon nitride (g-C₃N₄) via reductive alkylation. Our results reveal that the method is feasible and highly efficient in producing 2D g-C₃N₄ nanosheets which greatly overcome the drawbacks of bulk g-C₃N₄ photocatalyst. The reduced band gap caused by the covalent functionalization of alkyl chains on g-C₃N₄ lattice improves the photoresponse and the visible-light harvesting ability, indicating more charge carriers will generate under visible-light irradiation. In addition, the significantly enlarged surface area and the nanosheet structural benefits are favorable for the separation of photogenerated charge carriers. The photocatalytic activity of AGCNNs under visible-light irradiation is highly enhanced because of the synergistic effects of these favorable factors. The RhB photodegradation rate and the photocatalytic H₂ evolution rate of AGCNNs are further improved to 9.1 and 3.3 times as fast as that of g-C₃N₄, respectively.

1. Introduction

Since the first report of photocatalytic activity of TiO₂ in splitting water into H₂ and O₂,[1] intense efforts have been made to search for TiO₂-based photocatalysts for environmental remediation and the production of renewable energy.[2] However, TiO₂ semiconductor exhibits photocatalytic activity only with ultraviolet light irradiation, limiting its practical applications in photocatalysis. Graphitic carbon nitride (g-C₃N₄), a metal-free polymeric semiconductor possessing a laminated structure, g-C₃N₄ has been considered as a “sustainable” photocatalyst.[3,4] Yet, the photocatalytic performance of this polymeric semiconductor is somewhat limited by its insufficient visible-light absorption, low surface area, and fast recombination rate of photogenerated charge carriers.[5] Therefore, many efforts have been devoted to designing and optimizing g-C₃N₄ photocatalysts with improved photocatalytic activity.[6]

Intrinsically, light absorbance, charge carrier mobility, and the consequently photocatalytic activity of semiconductor photocatalyst lies on its electronic structure. So far, the modification of electronic structure has been adopted as a conventional approach to enhance the photocatalytic activity of g-C₃N₄. Among various modification strategies, metal (e.g., Fe,[7] Zn,[8] Cu,[9] Pt, and Pd[10]) or nonmetal (e.g., S,[11] O,[12] N,[13] P,[14] I[15]) doping is very effective in broadening the light responsive range of g-C₃N₄. However, the formation of localized/delocalized states from dopants in band gap usually reduces the mobility of photogenerated charge carriers and also lowers the stability of the doped materials. The electronic structure of g-C₃N₄ can also be adjusted by organic protocols. For example, new g-C₃N₄ structures with light absorption red-shifting from 470 to 750 nm were synthesized by dissolving g-C₃N₄ precursor with organic monomers, followed by thermally induced copolymerization.[16] Based on the reactivity of −C≡N=− and the residual −NH₂ groups in CN heterocycle, postfunctionalization were also performed to modify the electronic structure of g-C₃N₄.[17] Since the g-C₃N₄ lattice is formed, postfunctionalization strategy has the extra advantages to introduce various organic functional groups into g-C₃N₄, which allows the designing of a much broader set of light-harvesting g-C₃N₄ photocatalysts.

Textural engineering is a promising methodology to modulate the properties of a semiconductor.[5] Recently, various kinds of nanostructured g-C₃N₄ with improved photocatalytic performance have been designed and developed, including g-C₃N₄ quantum dots,[18] g-C₃N₄ nanorods,[19] g-C₃N₄ nanowires,[20] g-C₃N₄ nanoribbons,[21] g-C₃N₄ nanosheets,[22] g-C₃N₄ nanofiber,[23] g-C₃N₄ nanospheres,[24] g-C₃N₄ nanowires,[25] and mesoporous g-C₃N₄.[26] Among the various nanoarchitectural g-C₃N₄, 2D g-C₃N₄ nanosheets show exceptional advantages for the...
deposition of nanoparticles and the coupling of semiconductors. Atomically thick g-C_{3}N_{4} nanosheets can be prepared via a top-down strategy such as oxidation “etching” or liquid phase exfoliation.[24–26] However, since g-C_{3}N_{4} is chemically stable and insoluble in common solvents, the production yield of g-C_{3}N_{4} nanosheets is very limited even in high temperature condition or harsh sonication treatment. Therefore, the preparation of g-C_{3}N_{4} nanosheets in high yield with a more feasible and reliable method is still a big challenge.

In this study, we demonstrate a simultaneous exfoliation and surface modification strategy to prepare alkylated g-C_{3}N_{4} nanosheets (AGCNNs). The as-prepared AGCNNs were systematically characterized by various analytic techniques. Results reveal that the method is feasible and highly efficient in producing 2D g-C_{3}N_{4} nanosheets which significantly overcome the drawbacks of g-C_{3}N_{4} photocatalyst. The reduced band gap caused by the covalent functionalization of dodecyl functional groups on g-C_{3}N_{4} lattice improves the photoresponse and the visible-light harvesting ability, suggesting more charge carriers will generate under visible-light irradiation. Meanwhile, the significantly enlarged surface area and the nanosheet structural benefits are favorable for the separation of photogenerated charge carriers. The as-prepared AGCNNs exhibit much improved photocatalytic activity in pollutant degradation and photocatalytic H_{2} evolution under visible-light irradiation because of the synergistic effects of these favorable factors. The RhB photodegradation rate and the photocatalytic H_{2} evolution rate of AGCNNs are 3.9 and 1.9 times as fast as that of g-C_{3}N_{4}, respectively. Benefiting from the dispersible 2D nanosheet structure of AGCNNs, graphene sheets were highly dispersed and employed as electron acceptors in AGCNNs/graphene oxide (GO) composite photocatalysts, further promoting the separation of photogenerated charge carriers. The RhB photodegradation rate and the photocatalytic H_{2} evolution rate of AGCNNs were further improved to 9.1 and 3.3 times as fast as that of g-C_{3}N_{4}, respectively.

2. Results and Discussion

The simultaneous exfoliation and surface alkylation of g-C_{3}N_{4} is illustrated in Scheme 1. In anhydrous tetrahydrofuran (THF) solution, g-C_{3}N_{4} was reduced by lithium in the presence of naphthalene. The strong Coulombic repulsions between reducive g-C_{3}N_{4} layers overcome the van der Waals interactions. Meanwhile, the spontaneously intercalated Li^{+}(THF)_{x} complex ions further increased the lattice spacing and weakened π...π interaction between adjacent layers, resulting in the swelling and final exfoliation of g-C_{3}N_{4} sheets. The proposed reduction and exfoliation mechanism of g-C_{3}N_{4} is generally similar to the spontaneous dispersion of carbon nanotubes in a THF solution containing solvated electrons.[32] Upon reductive activation, the exfoliated g-C_{3}N_{4} layers were covalently functionalized by alkyl radicals that were in situ generated by the addition of 1-bromododecane. We have previously reported the surface alkylation of carbon nanotubes and graphene sheets.[33,34] Though the reactivity of g-C_{3}N_{4} (–C≡N−) differs from that of carbon nanotube and graphene (–C=C–), the reductive alkylation can proceed efficiently and yield alkylated g-C_{3}N_{4}, where the alkyl chains are covalently bonded with C, forming a new C−C bond. In fact, the –C≡N− has emerged as a radical acceptor and several intermolecular alkyl radical addition reactions have been investigated for the forming of new C−C bond.[35] The attached alkyl chains prevent the re-stacking of g-C_{3}N_{4} sheets and also improve their dispersibility in common solvents.

Figure 1a shows the photos of bulk g-C_{3}N_{4} (right) and exfoliated AGCNNs (left) with same quantity. The bulk g-C_{3}N_{4} produced by thermal polymerization of urea shows pale yellow
color, while the color of AGCNNs became darker with notable increased volume. In comparison to g-C$_3$N$_4$, the exfoliated AGCNNs exhibit much improved dispersibility in common solvents such as CHCl$_2$, CHCl$_3$, and N,N-dimethylformamide (DMF) (Figure 1b). Benefiting from the attached alkyl chains, the dispersion of AGCNNs is very stable, without aggregation upon standing for two months. The morphology of bulk g-C$_3$N$_4$ before and after exfoliation was investigated by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) (Figure 1c,e). The synthesized g-C$_3$N$_4$ exhibits compact structure with lateral size up to 10 µm. However, as shown in Figure 1d, after the simultaneous exfoliation and modification process, the compact graphitic-like structures were exfoliated into thin layers, exhibiting curled and fluffy morphology. The detailed structure of the exfoliated AGCNNs is further studied by TEM. As shown in Figure 1f, the very thin and free standing AGCNNs with a diameter of ~400 nm is stable under electron beam. The thickness (~1.5 nm) of AGCNNs measured by tapping mode atomic force microscopy (AFM) indicates that the exfoliated nanosheets are comprised of several CN layers (Figure S1, Supporting Information). The specific surface area of g-C$_3$N$_4$ before and after exfoliation was examined by nitrogen sorption isotherm analysis (Figure 2a). The specific surface area of AGCNNs is calculated as 50 m$^2$ g$^{-1}$. As a comparison, the specific surface area of g-C$_3$N$_4$ reference is calculated as 10 m$^2$ g$^{-1}$. The significant nanostructure-induced surface enlargement is expected to provide more active sites in photocatalysis, promoting mass transfer and charge carriers separation on the interface of photocatalysts.

X-ray diffraction (XRD), Fourier Transform Infrared (FTIR), and thermogravimetric analysis (TGA) were used to investigate the structure and chemical composition of g-C$_3$N$_4$ before and after exfoliation. Figure 2b shows the XRD patterns of bulk g-C$_3$N$_4$ as well as AGCNNs. The strong characteristic (002) peak centered at 27.5° is the interlayer stacking reflection of conjugated aromatic structures, corresponding to the

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**Figure 1.** a) Photos of bulk g-C$_3$N$_4$ (right) and AGCNNs (left) powder with same quantity (100 mg), b) CHCl$_2$, CHCl$_3$, and DMF dispersion of bulk g-C$_3$N$_4$ (right) and AGCNNs (left), c,e) SEM and TEM images of bulk g-C$_3$N$_4$, d,f) SEM and TEM images of AGCNNs.

**Figure 2.** a) Nitrogen adsorption–desorption isotherms, b) XRD patterns, c) FTIR spectra, and d) TGA analysis of bulk g-C$_3$N$_4$ and AGCNNs.
interplanar distance of 0.324 nm. The weak (100) peak centered at 13.0° is related to the in-plane separation of the tri-s-triazine pores. For the XRD pattern of AGCNNs, the (002) peak was reserved. This result suggests that the main structure of carbon nitride did not change in the reductive alkylation, which is consistent with literature reports.\textsuperscript{[24,25]} However, the (002) peak is broadened with significantly decreased intensity. It can be explained by the size-dependent properties of nanomaterials,\textsuperscript{[30]} because the thickness and size of g-C\textsubscript{3}N\textsubscript{4} were significantly reduced through the exfoliation and modification process as shown in SEM and TEM images.

Figure 2c shows the FTIR spectra of bulk g-C\textsubscript{3}N\textsubscript{4} and AGCNNs. It is found that g-C\textsubscript{3}N\textsubscript{4} and AGCNNs exhibit very similar FTIR spectra, indicating that the CN backbone in g-C\textsubscript{3}N\textsubscript{4} layer did not change in such a reductive alkylation. The peak at 808 cm\textsuperscript{-1} is a characteristic vibration peak of s-triazine ring system in g-C\textsubscript{3}N\textsubscript{4}.\textsuperscript{[36]} The absorbance peaks in the range of 1200–1650 cm\textsuperscript{-1} are the characteristic stretching modes of C–N in aromatic CN structures.\textsuperscript{[36]} The broad band at 2900–3600 cm\textsuperscript{-1} is attributed to the N–H stretching of residual –NH\textsubscript{2} in g-C\textsubscript{3}N\textsubscript{4}.\textsuperscript{[24,37]} It is noted that the FTIR spectrum of AGCNNs also exhibit peaks at 2919, 2847 cm\textsuperscript{-1}, which can be assigned to the alkyl C–H stretching of dodecyl groups attached on g-C\textsubscript{3}N\textsubscript{4} layers.\textsuperscript{[33]} The alkyl C–H stretching modes are less prominent probably because they are overlapped by the N–H stretching modes (2900–3600 cm\textsuperscript{-1}) of –NH\textsubscript{2} in g-C\textsubscript{3}N\textsubscript{4}.

The surface modification by alkyl groups was further confirmed by TGA (Figure 2d). Under N\textsubscript{2} atmosphere, bulk g-C\textsubscript{3}N\textsubscript{4} shows negligible weight loss up to 600 °C.\textsuperscript{[5]} At elevated temperature, the sample was completely decomposed with a decomposition peak at 700 °C. In comparison to bulk g-C\textsubscript{3}N\textsubscript{4}, AGCNNs show apparent weight loss when temperature was elevated to 200 °C. The total weight loss in the temperature range of 200–600 °C is about 17.1%. It is mainly caused by the thermal defunctionalization of alkyl chains on g-C\textsubscript{3}N\textsubscript{4} layers. Correspondingly, 11.2% of the s-triazines moieties were covalently modified by alkyl chains. The FTIR spectrum of defunctionalized AGCNNs which obtained at 550 °C in another experiment is shown in Figure S2 in the Supporting Information. The very similar features as that of g-C\textsubscript{3}N\textsubscript{4} indicate that the alkylation chemistry is reversible. At elevated temperature, AGCNNs was completely decomposed as that of bulk g-C\textsubscript{3}N\textsubscript{4}. However, the decomposition peak was found down-shifted to 690 °C, indicating less thermal stability than that of bulk g-C\textsubscript{3}N\textsubscript{4}.

The electronic structure and optical properties of AGCNNs were studied by UV–vis diffuse reflectance spectroscopy in conjunction with photoluminescence (PL) spectroscopy. As shown in Figure 4a, the absorption edge of AGCNNs was found red-shifted from 465 to 520 nm with respect to the bulk counterpart. The band gap was decreased from 2.67 to 2.38 eV. The decreased band gap confirmed the modification of electronic structure induced by surface functionalization, in which the alkyl chains were covalently attached rather than adsorbed on the lattice of g-C\textsubscript{3}N\textsubscript{4}. Also, the absorption of AGCNNs in the electromagnetic spectrum was extended to the whole visible-light range. The intrinsic absorption features of AGCNNs indicate that more charge carriers will generate under visible-light irradiation with respect to g-C\textsubscript{3}N\textsubscript{4}. Since PL emission originates from the recombination of charge carriers, PL analysis can be used to demonstrate the fate of photogenerated change carriers.
As presented in Figure 4b, the PL emission spectra of AGCNNs and g-C_3N_4 with an excitation wavelength of 330 nm are very similar. However, the PL signal of AGCNNs is much weaker, suggesting that the recombination of photogenerated charge carriers is greatly inhibited. As aforementioned characterization, the as-prepared AGCNNs exhibit 2D nanosheet structures. The peculiar nanoshaped structure and the greatly enlarged surface area not only offer abundant active sites in photocatalysis, but also shorten the diffusion length of charge migration, promoting the separation of charge carriers.

The photocatalytic activity of AGCNNs as well as bulk g-C_3N_4 were studied by the degradation of RhB (10 mg L\(^{-1}\)) under visible-light (\(\lambda > 400\) nm) irradiation (Figure 5a). For bulk g-C_3N_4 photocatalyst, only 63% of RhB was degraded within 60 min irradiation. When using AGCNNs as photocatalyst, 98% of RhB was removed within the same irradiation time. In Figure 5b, a pseudo-first-order linear relationship was shown by the plots of \(-\ln(c/c_0)\) versus irradiation time (t), in which \(c_0\) is the initial concentration of RhB and \(c\) is the concentration of RhB at irradiation time \(t\). The determined reaction-rate constant, \(k\), are 0.0162 and 0.0628 h\(^{-1}\) for g-C_3N_4 and AGCNNs, respectively. The photodegradation rate of RhB over AGCNNs was 3.9 times as fast as that of g-C_3N_4. These results indicate that the as-prepared AGCNNs possess superior photocatalytic activity in comparison to bulk g-C_3N_4.

The application of AGCNNs in photocatalysis was further confirmed by the photocatalytic H\(_2\) evolution from water/triethanolamine solution under visible-light (400–780 nm) irradiation. Through an in situ photodeposition method, 3 wt% Pt cocatalyst was deposited on the surface of AGCNNs. As shown in Figure 6a, AGCNNs show remarkably improved activity in photocatalytic H\(_2\) evolution over g-C_3N_4 reference. The average hydrogen evolution rate (HER) of AGCNNs is 1034.6 \(\mu\)mol h\(^{-1}\), which is 1.9 times the rate of g-C_3N_4 (544.3 \(\mu\)mol h\(^{-1}\)) (Figure 6b). In addition, the amount of H\(_2\) produced during the operation time of 12 h nearly increased linearly with the increasing of irradiation time, indicating the sufficient stability of AGCNNs as photocatalyst.

The photocatalytic activity of photocatalyst is greatly related to its surface area, light harvesting ability, and the separation efficient of photogenerated charge carriers. The drawbacks of g-C_3N_4 photocatalyst including insufficient visible light absorption, low surface area, and fast recombination rate of photogenerated charge carriers are successfully overcome via the simultaneous exfoliation and surface modification strategy. First, the promoted photoresponse and visible-light harvesting ability of AGCNNs indicate that more charge carriers will generate under visible-light irradiation. Second, the significant nanostructure-induced surface area enlargement and the nanosheet structural benefits of AGCNNs not only offer more reactive sites in photocatalysis, but more importantly shorten the migration length of photogenerated charge carriers for reducing the recombination probability. The greatly improved visible-light photocatalytic activity of AGCNNs was undoubtedly related to...
the synergistic effects of these favorable factors. In addition, by change the type of alkyl halide in the simultaneous alkylation, the wavelength dependence of alkylated g-C₃N₄ is expected to extend and tune the light absorption for H₂ generation.

Although the as-prepared AGCNNs show considerably improved photocatalytic activity, there is still much room for activity improvement. For example, the 2D nature of AGCNNs offers ample sites for the loading of other semiconductors, which will be a promising method to boost the photocatalytic performance. In this study, a series of composite photocatalysts, AGCNNs/x GO (where x standds for the wt% of GO), built between AGCNNs and GO were produced by sonochemical method. Upon solvothermal treatment at 180 °C for 6 h, GOs in AGCNNs/x GO composites were reduced to rGO.

The morphology and structure of AGCNNs/x GO composite photocatalysts were studied by SEM and TEM. As shown in Figure 7a–c, AGCNNs remain fluffy morphology and nanosheet structures after combination with GO. The insets in Figure 7a–c show that the color of AGCNNs/0.02GO, AGCNNs/0.05GO, and AGCNNs/0.10GO changes from dark yellow to gray with increasing content of GO. In the TEM image of AGCNNs/0.02GO (Figure 7d), the thin layered structure of AGCNNs is distinct, while graphene sheets cannot be seen clearly probably because of the very low content. In the TEM images of AGCNNs/0.05GO and AGCNNs/0.10GO (Figure 7e,f), however, a clear distinction between AGCNNs and graphene sheets was obtained, where the transparent and thin graphene sheets overlaid on the surface of AGCNNs.

The structure of AGCNNs/x GO composites was further studied by XRD (Figure S3, Supporting Information). All AGCNNs/x GO composites show similar XRD pattern as that of pure AGCNNs, featuring a main diffraction peak at 27.4°. Notably, no apparent peak of rGO was observed even in AGCNNs/0.10GO composite. This can be explained by the high dispersion of rGO in composite, in which the regular stacking of graphene sheets was totally destroyed. However, the combination of graphene sheets in the as-prepared composites is clearly evidenced by their FTIR spectra (Figure S4, Supporting Information). Apart from the intrinsic features of AGCNNs, new peaks emerged at 3000–3700, 1216, and 1083 cm⁻¹ are assigned to the stretching vibrations of —OH, —O—C, and C—OH that remained in rGO.[40,41] Moreover,

**Figure 7.** SEM and TEM images of AGCNNs/GO composite photocatalysts. a,d) AGCNNs/0.02GO, b,e) AGCNNs/0.05GO, and c,f) AGCNNs/0.10GO.
In summary, highly efficient alkylated g-C₃N₄ nanosheet photocatalysts were successfully synthesized via a simultaneous exfoliation and surface modification method. Due to the synergistic effects of increased visible-light harvesting ability and promoted separation efficiency of charge carrier, the as-prepared AGCNNs exhibit much improved photocatalytic activity under visible-light irradiation. The photocatalytic activity of AGCNNs was further improved when combined with a small amount of graphene sheets, in which graphene sheets were employed as electron acceptor promoting the separation of photogenerated charge carriers. The method reported in this work is feasible and highly efficient in producing 2D g-C₃N₄ photocatalysts for the production of renewable energy and the environmental remediation. A much broader set of light-harvesting g-C₃N₄ photocatalysts for various applications are able to prepare by changing the type of alkyl halide in the reductive alkylation.

4. Experimental Section

Polymerization Synthesis of g-C₃N₄: Bulk g-C₃N₄ was prepared by the thermal treatment of dicyandiamide as reported in previous paper.[44] Typically, dicyandiamide was put in a crucible with a cover, followed by thermal treatment in a muffle furnace at 550 ºC for 4 h. After that, the products were cooled down to room temperature at a cooling rate of 5 ºC min⁻¹. The resultant products were milled into powder for further investigation.

Preparation of AGCNNs: The preparation of AGCNNs was achieved by reductive alkylation as reported in the previous work.[45] In this work, 368 mg of g-C₃N₄ powder was dispersed in the THF solution of lithium and naphthalene. After 1 h reaction, 1-bromododecane (C₁₂H₂₅Br) was slowly injected until the quenching of dark green color. The final products were collected by vacuum filtration and dried in a vacuum oven at 60 ºC for 24 h.

Preparation of AGCNNs/xGO Composites: GO was prepared via a modified Hummers method.[44] To prepare AGCNNs/xGO composites (where x stands for the wt% of GO), 100 mg AGCNNs were dispersed in 35 mL deionized water, followed by the addition of 5.0, 12.5, and 25.0 µL of GO aqueous dispersion (4.02 mg mL⁻¹), respectively. After a further sonication for 30 min, the mixture was transferred into a 50 mL Teflon autoclave for thermal treatment. The thermal treatment on the conduction band of AGCNNs tend to transfer to rGO, further promoting the separation of charge carriers and inhibit their recombination. Figure S7 in the Supporting Information shows that all AGCNNs/xGO composites exhibit weaker PL signal than that of sole AGCNNs, confirming the promoted separation of charge carriers. In the photocatalytic H₂ evolution, the transferred electrons were injected to Pt co-catalyst and reduced H₂O (or H⁺) to H₂, while holes in the valance band of AGCNNs react with sacrificial reagent. In the photo-degradation of RhB, the highly active holes oxidize water to produce OH·, which would decompose RhB molecules into CO₂ and H₂O. It is worth to note that the optimal photodegradation efficiency of RhB and evolution rate of H₂ was achieved on AGCNNs/0.05GO rather than on AGCNNs/0.10GO. It means that the photocatalytic activity of AGCNNs decreased with over loading of GO. We think this is related to the opacity and light scattering caused by the over loading of GO in composite.[42]
was held at 180 °C for 10 h before cooling down to room temperature. The final AGCNNs/0.10GO nanocomposites, named AGCNNs/0.02GO, AGCNNs/0.05GO, and AGCNNs/0.10GO, were collected by vacuum filtration and dried in a vacuum oven at 60 °C for 24 h.

Photocatalytic Performance Studies: The photocatalytic activity of all products was examined by the photodegradation of pollutant under visible-light irradiation. Typically, photocatalyst (30 mg) was dispersed in 45 mL aqueous solution of RhB (10 mg L−1) in a quartz reactor. Prior to visible-light irradiation, the suspension was magnetically stirred in dark for 1 h to ensure the adsorption–desorption equilibrium between photocatalyst and model molecules. A 300 W Xe lamp (CEL-Xe300, Beijing Au-light, China) with a 400 nm cutoff filter was served as the light source. During irradiation, 2 mL of the dispersion was taken out at different time intervals (5 or 10 min) and then centrifuged at 10 000 rpm to remove the photocatalysts. The concentration of RhB was examined by measuring the absorption intensity at 554 nm using Varian Cary 5000 UV–vis spectrometer.

The photocatalytic hydrogen evolution from water over various photocatalysts was performed on a CEL-SPH2N photocatalytic activity evaluation system (Beijing Au-light, China). Photocatalyst (25 mg) was dispersed in 45 mL aqueous solution containing 10 vol% triethanolamine used as sacrificial reagent. The loading of 3 wt% Pt co-catalyst was achieved by dissolving H2PtCl6 in the above 45 mL triethanolamine used as sacrificial reagent. The loading of 3 wt% Pt was examined by measuring the absorption intensity at 554 nm using Varian Cary 5000 UV–vis spectrometer.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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