Full paper

Three-dimensional porous g-C_3N_4 for highly efficient photocatalytic overall water splitting

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ARTICLE INFO

Keywords:
Overall water splitting
Graphitic carbon nitride
Three-dimensional structure
Ultrathin nanosheets
Photocatalyst

ABSTRACT

Herein, this work constructs a three-dimensional porous graphitic carbon nitride assembled by highly crystalline and ultrathin nanosheets (3D g-C_3N_4 NS). 3D g-C_3N_4 NS could directly split pure water into H_2 and O_2 with high evolution rate up to 101.4 and 49.1 μmol g^{-1} h^{-1} under visible light, respectively, approximately 11.8 and 5.1 times higher than bulk g-C_3N_4 and g-C_3N_4 NS. Besides, it achieves a notable apparent quantum yield of 1.4% at 420 nm, significantly superior to previously reported Pt/g-C_3N_4. The efficient activity of 3D g-C_3N_4 NS is mainly attributed to its 3D interconnected open-framework, assembled by highly crystalline ultrathin nanosheet unit, provides a pathway for faster charge carrier transport. Moreover, benefiting from its 3D structure for preventing agglomeration of nanosheets, 3D g-C_3N_4 NS is stable for more than 100 h of overall water splitting reaction.

1. Introduction

Photocatalytic overall water splitting into H_2 and O_2 without using sacrificial agents is considered as one of the most ideal ways to convert solar energy into renewable H_2 energy [1–6]. Although a number of inorganic semiconductor have been reported to be efficient photocatalysts for overall water splitting, most only operation under UV light irradiation owing their large band-gap energy [7–11]. To further utilize solar energy to split water, it is therefore essential to exploit a visible-light responsive photocatalyst. In the last years, numerous efforts have been made to develop such a material [12–14]. Especially, Domen's group have reported that certain oxynitrides, such as TaON, LaMg_xTa_{1-x}O_{1+3x}N_{2-3x} and (Ga_{1-x}Zn_x)(N_{1-x}O_x), are quite excellent photocatalysts for overall water splitting under visible light [15–18]. Recently, their group reported the SrTiO_3-La-Rh/Au/BIVO_3-Mo system with a solar-to-hydrogen (STH) energy conversion efficiency of 1.1% [19]. Besides, Mi's group have studied InGaN/GaN multiband nanowire heterostructures, which show highly efficient overall water splitting activity under visible light and have achieved a notable STH energy conversion efficiency of 3.3% [20]. However, the use of metal-free organic photocatalysts for overall water splitting in pure water has been much less reported.

Recently, conjugated graphitic carbon nitride (g-C_3N_4) polymers with earth abundant elements and appropriate band gap have been considered as effective photocatalysts for overall water splitting under visible light irradiation [21–23]. Compared with traditional inorganic semiconductors, g-C_3N_4 has more superiority, including being metal-free, nontoxic and low-cost, and the fact that its structure and composition can also be easily tuned via adjusting the building blocks of versatile organic protocols [24–30]. Indeed, significant achievements have been obtained on the g-C_3N_4 based photocatalysts for enhanced overall water splitting activity through carbon quantum dots deposition and so on [31–34]. Even so, direct and efficient photocatalytic water splitting over g-C_3N_4 always encounters great challenges of low specific surface area and sluggish photogenerated carrier transfer. Thus, to further improve the photocatalytic activity of g-C_3N_4, it is essentially necessary to employ a new structural design to provide large specific surface area and accelerate the intrinsic photogenerated carrier transfer kinetics. Although two-dimensional (2D) ultrathin nanosheets of g-C_3N_4 have been considered as a novel class of nano-materials to satisfy the demand, the fact of structure restacking and agglomeration owing to strong van der Waals force-induced adhesion among ultrathin nanosheets is inevitable, which evidently reduces the specific surface area [35,36]. Fortunately, three-dimensional (3D) photocatalysts assemblies...
of 2D ultrathin nanosheets can not only present great specific surface area, but also maximize the utilization of incident photons via the multireflection within the interconnected open-framework [37-42]. More importantly, 3D structure could be as supporters to prevent agglomeration of ultrathin nanosheets and provide a pathway for electron transport.

Herein, we therefore conceptually design a three-dimensional porous graphic carbon nitride assembled by highly crystalline and ultrathin nanosheets (3D g-C3N4 NS) via a facile bottom-up supramolecular self-assembly route. The prepared 3D g-C3N4 NS presents 3D interconnected open-framework with extremely great specific surface area, which could be as supporters to avoid restacking of nanosheets and provide a pathway for photogenerated electron transport. As a result, 3D g-C3N4 NS simultaneously realized steady and efficient photocatalytic overall water splitting activity with H2 and O2 evolution rate up to 102 and 51 μmol g⁻¹ h⁻¹ in pure water under visible light. Besides, it achieved a notable apparent quantum yield (AQY) of 1.4% at 420 nm.

2. Experimental section

2.1. Synthesis of 3D g-C3N4 NS

All the chemicals were purchased from Sigma-Aldrich and used without further purification. 3D g-C3N4 NS was synthesized by a facile bottom-up supramolecular self-assembly route. In a typical synthesis, 0.01 mol melamine and 0.01 mol cyanuric acid were mixed in 50 mL deionized water and magnetic stirred for 12 h at room temperature. Subsequently, the white cyanuric acid-melamine (CM) supramoleculars were centrifuged and dried by vacuum freeze drying. Then, the resulting CM supramolecular precursors were calcined in a muffle furnace at 550°C for 4 h with a heating rate of 5°C min⁻¹ to obtain light yellow 3D g-C3N4 NS. 2.3D g-C3N4 NS-1 also were prepared using 0.02 mol melamine and 0.01 mol cyanuric acid. As comparison, the bulk g-C3N4 and g-C3N4 nanosheets (g-C3N4 NS) were synthesized according to the literature [43,44].

2.2. Characterization

Field emission scanning electron microscope (FESEM) images were performed on a Hitachi SU-8000 at an accelerating voltage of 5 kV. Transmission electron microscope (TEM) images were operated on Hitachi HT7700 at an accelerating voltage of 100 kV. High-resolution transmission electron microscope (HRTEM) images were recorded on JEOL JEM-2100F at an accelerating voltage of 200 kV, and the images were analyzed by DigitalMicrograph software. Atomic force microscopy (AFM) study was carried out using Shimadzu SPM-960. BET surface area measurements were recorded by N2 adsorption at 77 K using a Micrometrics (ASAP 2010V5.02H) surface area analyzer. X-ray diffraction (XRD) patterns were obtained on a Rigaku D/max-2400 X-ray diffractometer using a Cu Kα1 (λ = 0.15418 nm) at 40 kV and 200 mA, with a scan step of 0.02°. The Fourier transform infrared (FT-IR) spectra were acquired on a Bruker V70 spectrometer. The solid-state 13C nuclear magnetic resonance (NMR) spectra were recorded on a JEOL JNM-ECZ600R spectrometer. UV–Vis diffuse reflectance spectroscopy (DRS) were spectra were obtained on a Hitachi U-3010 UV–Vis spectrophotometer with BaSO4 as reference. X-ray photoelectron spectroscopy (XPS) measurements were performed using an ESCALAB 250Xi instrument (Thermo Scientific) with Al Kα radiation. The electron paramagnetic resonance (EPR) measurements were carried out on a JEOL JES-FA200 spectrometer. Room temperature photoluminescence (PL) spectra were recorded on Edinburgh F900 (FLS) fluorescence spectrometer with an excitation wavelength of 340 nm. Time-resolved photoluminescence spectra were collected on Edinburgh FLSP920 fluorescence spectrometer.

2.3. Photoelectrochemical measurements

Photoelectrochemical measurements were performed on a three-electrode system using a CHI660E electrochemical workstation with the as-prepared samples covered ITO glass as the working electrodes, a Pt wires as the counter electrode, and a saturated calomel electrode (SCE) as reference. A 300 W xenon lamp with cutoff filter (λ ≥ 420 nm) was used as the light source and the electrolyte was 0.1 mol L⁻¹ Na2SO4 aqueous solution. For the working electrodes, a sample (1 mg) was dispersed in isopropanol (1 mL) to obtain a slurry. Then the slurry was coated onto the ITO glass and dried in an oven overnight. Subsequently, the working electrodes were exposed to UV light for 1 h to eliminate isopropanol and calcined at 120°C for 2 h. Electrochemical impedance spectroscopy (EIS) spectra were recorded under an AC perturbation signal of 10 mV over the frequency range from 100 kHz to 0.1 Hz. Mott–Schottky plots were collected with a scan rate of 5 mV s⁻¹ at different frequency (1000, 800 and 500 Hz).

2.4. Photocatalytic water splitting test

The photocatalytic overall water splitting reaction was carried out in a Pyrex top-irradiation reaction vessel connected to a glass closed gas system (Labsolar-6A, PerfectLight) (Fig. S19c). 1 wt% Pt and 3 wt% IrO2 as co-catalysts were loaded on the photocatalysts by in situ photo-deposition method using H2PtCl6 and NaIrO2 as in Refs. [45,46] and 50 mg photocatalysts were dispersed in 100 mL distilled water without any sacrificial agent. The reaction solution was kept at 5°C by circulating cooling water system, then it was evacuated several times to remove air completely. And a 300 W Xe lamp with a cutoff filter (λ ≥ 420 nm, light intensity 438 mW cm⁻²) was used as the light source. The amounts of gases produced were measured by gas chromatography equipped with a thermal conductive detector (TCD) and a 5 Å molecular sieve column, using argon as the carrier gas.

The apparent quantum yield (AQY) of the overall water splitting is calculated from equation as follows:

$$\text{AQY} = \frac{2 \times \text{the number of evolved H molecules}}{\text{the number of incident photons}} \times 100\%$$

The solar-to-hydrogen (STH) conversion efficiency is given by:

$$\text{STH} = \frac{R(H_2) \times \Delta G_r \times 100\%}{P \times S}$$

Where R(H2), ΔG_r, P, and S represent the rate of hydrogen evolution, the Gibbs energy for the reaction (H2O(l) → H2(g) + 1/2O2(g)), the energy intensity of the AM1.5G solar irradiation (100 mW cm⁻²) and the irradiated sample area (1 cm²), respectively.

Several band-pass filters (FWHM = 15 nm) were employed to achieve a different incident light wavelength under a 300 W Xe lamp (FX300, PerfectLight) for measurement of the quantum yield. For full spectrum measurement, a 300 W Xe lamp (FX300, PerfectLight) with a standard AM1.5G filter, outputting the light density of 100 mW cm⁻², was used as irradiation light source. The average intensity was determined by an optical power meter (PM100D, THORLABS).

3. Results and discussion

3.1. 3D g-C3N4 assemblies of highly crystalline and ultrathin nanosheets

Different from the top-down approach to prepare 3D structured g-C3N4 by multi-step method reported before [35,47,48], the 3D g-C3N4 NS was fabricated via a facile bottom-up supramolecular self-assembly route, as shown in Fig. 1a. Firstly, the 3D network cyanuric acid-melamine (CM) supramoleculars (Fig. S1a) as precursor were assembled through the hydrogen bonding between melamine and cyanuric acid in aqueous solution, and followed by a freeze drying and high temperature polycondensation reaction. Finally, a light yellow 3D g-C3N4 NS sample
was obtained. Surprisingly, the prepared 3D g-C3N4 NS sample, just like an aerogel, exhibits the ultralight characteristics with a quite low volume fraction of about 18.3 mg cm$^{-3}$ (Fig. S1b), and can even stand on the green bristlegrass steadily. Moreover, the volume of 3D g-C3N4 NS with the same weight is much larger than that of bulk g-C3N4 or even g-C3N4 NS (Fig. S1c). Additionally, the lighter 3D g-C3N4 NS can also be obtained by increase the ratio of cyanuric acid and melamine.

The morphology and microstructure of 3D g-C3N4 NS were further investigated with the scanning electron microscope (SEM) and transmission electron microscope (TEM) as shown in Fig. 1b and c. Different from the usual bulk g-C3N4 consisting of solid agglomerates with a size of several micrometers, 3D g-C3N4 NS presents 3D interconnected open-framework with an average pore width of about 20 nm (Fig. S5) fabricated by ultrathin nanosheet units in Fig. 1b. The ultrathin g-C3N4 nanosheets appear as floppy agglomerates with a lateral scale of several micrometers, similar to the graphene nanosheets [49], and the ultrathin nanosheet unit trends to bend and its edges are ragged. The TEM image in Fig. 1c further confirms that the basic unit of 3D g-C3N4 NS is ultrathin nanosheet with a lateral scale of several micrometers. Meanwhile, lots of clear lattice fringes appear in HRTEM image (Fig. 1c inset) of the ultrathin nanosheet edges, indicating the high crystallinity of 3D g-C3N4 NS. The corresponding lattice fringes of 0.329 nm are ascribed to (002) π–π interlayer stacking motif, and only 4–5 lattice fringes can be observed, suggesting that 3D g-C3N4 NS consist of 4–5 g-C3N4 layer units. Furthermore, the atomic force microscopy (AFM) image (Fig. 1d) clearly shows the layer thickness of about 2.3 nm, exactly corresponding to about 5 g-C3N4 layer units, almost consistent with HRTEM result.

Benefiting from 3D porous interconnected open-framework assemblies of ultrathin nanosheet, 3D g-C3N4 NS exhibit quite large specific surface area, up to about 130.00 m$^2$ g$^{-1}$, and is approximately 13 and 1.4 times larger than bulk g-C3N4 and g-C3N4 NS (Fig. 2a), which could provide more active sites and improve the solar light harvesting. In X-ray diffraction (XRD) patterns (Fig. 2b), two characteristic diffraction peaks located at 13.0° and 27.4° corresponding to (100) in-plane structural packing motif and (002) interlayer stacking of aromatic segments appear in all g-C3N4 samples. With respect to bulk g-C3N4, the (002) diffraction peak in 3D g-C3N4 NS-2 is shifted from 27.38° to 27.77°, implying a shortened gallery distance between the basic layer unit, and the significantly reduced full width at half maximum further demonstrates the higher crystallinity of 3D g-C3N4 NS. In Fourier transform infrared (FTIR) spectroscopy (Fig. 2c), the sharp peak at 811 cm$^{-1}$ is assigned to a signature of the formation of tri-s-triazine. The strong absorption bands in the region of 1100–1700 cm$^{-1}$ are ascribed to the typical stretching vibration modes of C-N heterocycles, these absorption bands become sharper probably owing to the more ordered packing of tri-s-triazine units [50]. The wide peaks between 3000 and 3500 cm$^{-1}$ are assigned to O-H and N-H stretching vibration modes. Moreover, in the solid-state $^{13}$C CP-MAS NMR spectra (Fig. S6) of 3D g-C3N4 NS, two peaks at 155.6 (C1) and 164.3 ppm (C2) are ascribed to the C(i) atoms of methyl (CN$_3$) and C(e) atoms of [CN$_2$(NH$_x$)] $^{[44]}$, respectively, further confirming the existence of tri-s-triazine in 3D U-C$_3$N$_4$ NS. Additionally, the element composition of C, N and O in 3D g-C3N4 NS was also investigated by XPS analysis (Fig. S7). In C 1s spectra (Fig. S7c), the peak centered at 288.5 eV is assigned to sp$^2$-hybridized carbon in the N-containing aromatic ring (N=C-N), and the decrease in the peak intensity at 285.0 eV of 3D g-C3N4 NS suggests that the fewer impurities of graphitic carbon species (C(sp$^2$)-C(sp$^2$) bonds) originating from defective polymerization, further demonstrating the more ordered packing of tri-s-triazine units relative to bulk g-C3N4. The fewer graphitic carbon species is helpful for the optimization of the aromatic π-conjugated system for charge separation, since too many graphitic carbon centers doped in the g-C$_3$N$_4$ conjugated system will act as defect centers for charge recombination [51].

Then, the redox potential of 3D g-C3N4 NS was calculated according to the UV–Vis diffuse reflectance spectra and Mott-Schottky plots (Fig. S8 and Fig. S9) [52,53], as shown in Fig. 2d. The conduction band potential 3D g-C$_3$N$_4$ NS is more negative than the reduction of H$^+$/H$_2$, and the valence band potential is more positive than the oxidation potential of O$_2$/H$_2$O, which completely meets the thermodynamic energy criterion of
direct water splitting. Additionally, With respect to bulk g-C$_3$N$_4$, the conduction band potential moves up obviously, whereas the valence band potential of 3D g-C$_3$N$_4$ NS has little change, meaning a stronger capability of 3D g-C$_3$N$_4$ NS for overall water splitting.

3.2. Highly efficient and stable water splitting performance

The photocatalytic overall water splitting experiment of 3D g-C$_3$N$_4$ NS was performed in pure water under visible light irradiation, and Pt and IrO$_2$ as co-catalysts were photoloaded in situ on samples to boost H$_2$ and O$_2$ generation in advance [44,46], respectively. The typical time course of gas evolution over 3D g-C$_3$N$_4$ NS-2 is shown in Fig. 3a. Notably, both H$_2$ and O$_2$ were generated simultaneously under light irradiation, and the evolved H$_2$ to O$_2$ ratio was very close to the expected 2:1 stoichiometry for overall water splitting. The average H$_2$ and O$_2$ evolution rates are 101.4 and 49.1 μmol g$^{-1}$ h$^{-1}$ over 3D g-C$_3$N$_4$ NS-2, respectively, approximately 11.8 and 5.1 times higher than that of bulk g-C$_3$N$_4$ and g-C$_3$N$_4$ NS (Fig. 3b), significantly superior to a great many previously reported g-C$_3$N$_4$-based photocatalysts (Table S5). To further confirm that the photocatalytic overall water splitting reaction was driven by the absorption of an incident photon, the wavelength-dependent overall water splitting experiments were also carried out. As shown in Fig. 3c, the AQY values for overall water splitting over 3D g-C$_3$N$_4$ NS-2 is well coincident with its UV–Vis DRS, and the AQY at 420 nm was calculated to be 1.4%, significantly higher than that of Pt/ g-C$_3$N$_4$ (0.3% at 405 nm) by Wang’s group [21]. Then, the solar-to-hydrogen (SHT) energy-conversion efficiency of 3D g-C$_3$N$_4$ NS was also evaluated under AM 1.5G simulated sunlight (100 mW cm$^{-2}$, 1 cm$^2$). After 6 h illumination, a total of 5.73 μmol H$_2$ was produced, and the SHT value of 3D g-C$_3$N$_4$ NS was determined to be 0.06% (Fig. S12).

In addition, no notable deactivation emerges over 3D g-C$_3$N$_4$ NS-2 during a continuous more than 100 h measurement (Fig. 3d), indicating its robust resistance to water and light corrosion at the soft interface. However, the overall water splitting performance of g-C$_3$N$_4$ NS dropped sharply during the repeated cycles (Fig. S13), and its gas evolution rate was significantly reduced by 44% after 102 h reaction. As shown in Fig. S14, serious agglomeration of nanosheets occurred over g-C$_3$N$_4$ NS after the continuous cycles, which should be the main reason for the decrease of its water splitting activity. In contrast, 3D g-C$_3$N$_4$ NS-2 still presents ultrathin nanosheet structure, further demonstrates that 3D structure could be as supporters to prevent agglomeration of ultrathin nanosheets.

3.3. Faster charge carriers separation over 3D g-C$_3$N$_4$ NS

Although the specific surface area of 3D g-C$_3$N$_4$ NS-2 is slightly higher than that of g-C$_3$N$_4$ NS, even that of 3D g-C$_3$N$_4$ NS-1 is lower, 3D g-C$_3$N$_4$ NS still shows significantly superior overall water splitting performance, indicating that the specific surface area doesn’t mainly account for the highly efficient activity of 3D g-C$_3$N$_4$ NS. It’s well known that the photocatalyst activity is closely related to its photogenerated charge carrier separation efficiency. To further understand why the 3D g-C$_3$N$_4$ NS exhibited superior photocatalytic overall water splitting performance, the charge recombination behavior of all samples was investigated. In EPR spectra (Fig. 4a), all samples show one single Lorentzian line with a g value of 2.0036 that is induced by unpaired electrons on sp$^2$-carbon atoms of the heptazine rings [54]. Compared to bulk g-C$_3$N$_4$ and g-C$_3$N$_4$ NS, the stronger EPR spin intensity of 3D g-C$_3$N$_4$ NS-2 implies the obviously higher concentration of unpaired electrons and greater ability to delocalize electrons, which is favorable to the photogenerated electron-hole pair separation. The excitonic process of 3D g-C$_3$N$_4$ NS-2 was also monitored by photoluminescence (PL) measurements. In Figs. 4b and 3D g-C$_3$N$_4$ NS-2 exhibit an obviously decreased PL emission intensity with respect to bulk g-C$_3$N$_4$ and g-C$_3$N$_4$ NS, which further demonstrates its suppressed electron-hole pair recombination rate. Meanwhile, time-resolved PL spectra
monitored at the corresponding emission peaks provide the average radiative lifetime ($\tau$) of the charge carriers. The $\tau$ of bulk g-C$_3$N$_4$, g-C$_3$N$_4$ NS, and 3D g-C$_3$N$_4$ NS-2 are 4.24 ns, 5.38 ns, and 6.69 ns, respectively. As this increased exciton lifetime of 3D g-C$_3$N$_4$ NS-2 significantly implies enhanced exciton dissociation, and the longer diffusion length and lifetime of charge carriers are further confirmed.

Fig. 3. (a) Time-dependent overall water splitting over 3D g-C$_3$N$_4$ NS-2, (b) overall water splitting rate over bulk g-C$_3$N$_4$, g-C$_3$N$_4$ NS, and 3D g-C$_3$N$_4$ NS, (c) the wavelength-dependent AQY of overall water splitting over 3D g-C$_3$N$_4$ NS-2, and the repeated cycles of overall water splitting over 3D g-C$_3$N$_4$ NS-2. Reaction condition: 1 wt% Pt and 3 wt% IrO$_2$ as co-catalysts; distilled water (pH 7.0, 100 mL); light source, 300 W Xe lamp equipped with a visible light filter ($\lambda \geq 420$ nm) or various band-pass filters; All reaction were carried out at 278 K and 3 kPa.

Fig. 4. (a) EPR spectra, (b) PL spectra (time-resolved PL spectra collected under an excitation of 330 nm inset), (c) electrochemical impedance spectra (The equivalent circuit impedance model inset), and (d) transient photocurrent density of bulk g-C$_3$N$_4$, g-C$_3$N$_4$ NS and 3D g-C$_3$N$_4$ NS-2.
Furthermore, as shown in Fig. 4c, the charge transfer resistance ($R_{ct}$) of 3D g-C$_3$N$_4$ NS-2 estimated via fitting the hemicycle radius on electrochemical impedance spectroscopy (EIS) was only 4.134$\Omega$, much lower than that of bulk g-C$_3$N$_4$ and g-C$_3$N$_4$ NS (Table S7), demonstrating a faster electron transfer kinetics in 3D g-C$_3$N$_4$ NS-2. Additionally, the enhanced photocurrent response (Fig. 4d) further reveals the significantly improved mobility of charge carriers in 3D g-C$_3$N$_4$ NS-2. Except the 3D structure of 3D g-C$_3$N$_4$ NS-2, g-C$_3$N$_4$ NS and 3D g-C$_3$N$_4$ NS-2 both presents ultrathin nanosheet structure, and their layer thickness and specific surface area are very close. However, 3D g-C$_3$N$_4$ NS-2 exhibits longer lifetime of charge carriers, much lower charge transfer resistance and significantly enhanced photocurrent response than that of g-C$_3$N$_4$ NS, which fully demonstrates that the 3D structure of 3D g-C$_3$N$_4$ NS-2 provides a pathway for electron transports.

Therefore, benefitting from the 3D porous interconnected open-framework to prevent agglomeration of nanosheets and provide a pathway for electron transport, the suppressed generated electron-hole pair recombination rate and a faster charge carrier transfer kinetics emerge in 3D g-C$_3$N$_4$ NS with respect to bulk g-C$_3$N$_4$ and g-C$_3$N$_4$ NS, resulting to its superior photocatalytic overall water splitting activity.

4. Conclusion

In summary, 3D g-C$_3$N$_4$ assembled by highly crystalline and ultrathin nanosheets were successfully fabricated through a facile bottom-up supramolecular self-assembly route. The unique 3D porous interconnected open-framework makes 3D g-C$_3$N$_4$ NS exhibit quite large specific surface area, being as supporters to prevent agglomeration of nanosheets and provides a pathway for electron transports. As a consequence, 3D g-C$_3$N$_4$ NS realized highly efficient and stable overall water splitting under visible light, and achieved considerable AQY as high as 1.4% at 420 nm. Briefly, this work throws light on designing polymer photocatalysts with 3D porous structure arrangements of low-dimensional nanomaterials for improved solar energy capture and conversion.

Conflicts of interest

The authors declare no conflict of interest.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (grant numbers 21437003, 21673126, 21761142017, and 21621003) and the Collaborative Innovation Center for Regional Environmental Quality.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.nanoen.2019.03.010.

References
