One pot synthesis of Ti and O co-doped g-C₃N₄: a novel composite for efficient visible-light-driven photocatalytic inactivation of E. cloacae

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Abstract
Co-doping is one of the most facile and efficient strategies for boosting the photocatalytic performance of g-C₃N₄. Herein, a series of Ti and O co-doped g-C₃N₄ (Ti/OCN) composites with excellent photocatalytic inactivation activity of Enterobacter cloacae (E. cloacae) were successfully synthesized by simple calcination of urea/formic acid/TiCl₄ precursor. The obtained Ti/OCN-3 possessed the optimal photocatalytic capability with complete inactivation of 6.20 log CFU·ml⁻¹ E. cloacae under 60 min of visible light irradiation. The improved photocatalytic disinfection performance was probably attributed to the synergistic effect of Ti and O co-doping, which led to an extended visible light response and accelerated charge transport efficiency. Furthermore, scavenger trapping and electron spin resonance (ESR) experiments revealed that ·O₂⁻, h⁺ and ·OH were responsible for the enhanced inactivation activity. Overall, this research provided a promising strategy for fabricating g-C₃N₄-based catalysts to efficiently deal with the hazards of pathogenic microorganisms.

1. Introduction

Enterobacter cloacae (E. cloacae), like Escherichia coli (E. coli), an opportunistic Gram-negative pathogen, was implicated in various infections, could cause a variety of nosocomial diseases, such as urinary tract, lower respiratory tract, central nervous system, and so on [1]. Recently, the frequent emergence and spread of drug-resistant E. cloacae have become a huge threat to the health of human beings [2]. Therefore, it is urgent to exploit an efficient and eco-friendly technology to eradicate pathogen contamination. In recent years, photocatalytic technologies based on semiconductors have been universally recognized in the field of microbial contamination control due to the combined advantages of effective, reusable and low-energy [3, 4]. Thereby, numerous semiconductor photocatalysts have been explored to eliminate pathogens as green reagents [5]. Among the photocatalysts investigated, graphitic carbon nitride (g-C₃N₄) has undoubtedly aroused tremendous attention as the most promising photocatalyst owing to its unique features of facile synthesis, stubborn physicochemical stability, non-toxicity as well as a relatively suitable band gap (~2.7 eV) [6, 7]. Even so, the practical application of bare g-C₃N₄ was still suffered from its inherent restrictions of limited visible light response, rapid recombination of photoexcited charges as well as weak surface area [8, 9].

Aiming at surmounting the above drawbacks to enhance the photocatalytic property of g-C₃N₄, various endeavors have been carried out, mainly including doping with heteroatoms, nanostructures construction, heterojunctions fabrication and organic dyes sensitization [10, 11]. Among them, doping with heteroatoms was widely considered as the simplest strategy to improve the photocatalytic performance of g-C₃N₄. In detail, metal
doping could effectively regulate the electronic structure and accelerate the transport of photoexcited charges by providing new electron transport channels or enhancing electrical conductivity, thus observably boosted the photocatalytic properties. Similarly, doping with nonmetallic atoms could strengthen the electron delocalization, which resulted in an extended light response and efficient separation of photoexcited carriers, thereby improving the photocatalytic activity. More exciting, co-doping of metal and nonmetal atoms could further substantially improve the photocatalytic activities of g-C₃N₄ by combining the above-mentioned advantages. Hitherto, there had many achievements, including Fe-P, Mn-O and Cu-O co-doping, to heighten the photocatalytic capacity of g-C₃N₄ for pollutant degradation, H₂ evolution, and so on. However, to the best of our knowledge, there were less reports about the construction and application of Ti, O co-doping g-C₃N₄ composites.

Herein, for the first time, a series of Ti and O co-doped g-C₃N₄ composites were synthesized by a facile one-pot thermal calcination process. The subsequent co-doped g-C₃N₄ exhibited extended visible light absorption, accelerated charge transport and significantly reduced electrical resistance, which resulted in a superior photocatalytic activity for E. cloacae inactivation. Meanwhile, the photocatalytic inactivation mechanisms were proposed in detail based on the results of scavenger trapping and electron spin resonance (ESR) experiments.

2. Experimental section

2.1. Reagents

Urea (CO(NH₂)₂), formic acid (HCOOH), titanium tetrachloride (TiCl₄), 4-hydroxy-2, 2, 6, 6-tetramethylpiperidinyloxy (TEMPO), ammonium oxalate ((NH₄)₂C₂O₄, AO), isopropanol (IPA) and 5, 5-dimethyl-1-pyrroline N-oxide (DMPO) with analytical grade were obtained from Aladdin Biochemical Technology Co., Ltd (Shanghai, China) and utilized directly without additional treatment. Double-distilled water (>18.2 MΩ cm) was used throughout the experiments.

2.2. Synthesis of composites

The Ti and O co-doped g-C₃N₄ was synthesized through a facile one-pot thermal calcination the mixture of urea, formic acid and TiCl₄ (Scheme 1). Specifically, urea (20.0 g), formic acid (0.6 ml) and a certain amount of TiCl₄ (0.1, 0.2, 0.3, 0.4 ml) were completely dissolved in 5 ml of deionized water. After evaporating excess water from the above mixture, the residual solid was sent into covered alumina crucibles and directly calcined at 520 °C for 2 h under air atmosphere with a rate of 5 °C·min⁻¹. When cooling naturally to room temperature, the resultant products were collected and marked as Ti/OCN-1, Ti/OCN-2, Ti/OCN-3 and Ti/OCN-4, respectively.

For comparison, the bare g-C₃N₄ was obtained by direct calcination of urea at 520 °C for 2 h, and was marked as CN.

2.3. Characterizations

The phase structure and surface functional groups of as-synthesized composites were measured by x-ray diffraction (XRD, D8, Bruker, Germany) with Cu Kα radiation, and Fourier transform infrared spectrum (FT-IR, Nicolet Nexus 470, Thermo, USA), respectively. The morphology and surface element distribution were detected by scanning electron microscopy (SEM, SU8010, Hitachi, Japan) assembled with energy dispersive spectrometer (EDS) and energy dispersive x-ray spectroscopy (EDX). The x-ray photoelectron spectroscopy (XPS) data was obtained on an ESCALAB 250XI (Thermo, USA) spectrometer with Al Kα radiation. The optical
properties of as-synthesized composites were assayed by a UV-Visible spectrophotometer (UV-2550, Shimadzu, Japan) and the steady-state photoluminescence (PL) spectra were performed using a fluorescence spectrophotometer (FLS980, Edinburgh, UK) with excitation wavelength at 370 nm.

The electrochemical measurements of as-synthesized composites, including transient photocurrent responses and electrochemical impedance spectroscopy (EIS) were measured on CHI760E workstation (CH Instruments, China) with three-electrode system in 0.1 M Na2SO4 electrolyte solution.

2.4. Photocatalytic inactivation properties
The photocatalytic inactivation properties of as-synthesized composites were estimated by disinfection of E. cloacae (ATCC 700323) using a 300 W xenon lamp equipped with a cut off filter (>420 nm) as simulated visible light source. Typically, 40 mg of samples were uniformly suspended into 99 ml of sterile water by 10 min of ultrasonic treatment. Next, after introducing 1.0 ml pre-adjusted E. cloacae suspension with cell density of $\sim 1.0 \times 10^9$ CFU·ml$^{-1}$, the photocatalytic inactivation experiment was started under continuous magnetic stirring. At designed time intervals, 0.1 ml of the above mixture was collected and diluted immediately, and then spread evenly on LB nutrient agar plate. Finally, after being incubated at 37°C for 16 h, the number of colonies was counted for inactivation efficiency assessment.

Meanwhile, the visible light alone and the catalyst in dark were carried out as control experiments to assess their cytotoxicity against E. cloacae.

Moreover, the changes in bacterial cell membrane integrity during the inactivation process were examined by fluorescently staining of E. cloacae using Live/Dead Bacterial Viability Kits (Fisher Scientific, Thermo, USA) according to the previous literature reports [18, 19].

3. Results and discussion

3.1. Characterizations of as-synthesized composites
The structural information and optical properties of as-synthesized composites were systematically investigated by XRD, FT-IR, SEM, TEM, XPS and UV–vis DRS. The XRD patterns of bare CN and Ti/OCN composites were depicted in figure 1(a). All the samples possessed the same two typical graphite-like structure diffraction peaks at around 13.2° (100) and 27.3° (002), implying that addition of a small amount of formic acid and TiCl4 did not change the original crystal structure of CN. Here, the (100) peak represented the in-planar repeating structures of tri-s-triazine units and the (002) peak was derived from the interplanar stacking of aromatic rings, respectively [20]. Specifically, compared with CN, the (002) peak intensities of co-doped composites gradually weakened as TiCl4 dosage increased, which could be attributed to the poorer crystallinity and the generation of defects [21, 22]. Interestingly, no obvious diffraction peaks relevant to Ti species, such as TiO2, could be detected in any of the Ti/OCN composites, probably mainly due to the low additive content or monatomic form existence of Ti in CN framework [21].

Subsequently, FT-IR spectra of as-synthesized composites were exhibited in figure 1(b). The FT–IR spectra of Ti/OCN composites were essentially consistent with the bare CN except for some slight changes, indicating that the doping process did not destroy the fundamental structure of CN. In detail, the broad peak centered at around 3200 cm$^{-1}$ was originated from the stretching vibration of terminal C—NH$_2$, C—OH groups and adsorbed H$_2$O [23]. The absorption bands in the range of 1200—1650 cm$^{-1}$ were attributed to the typical...
stretching vibration models of aromatic tri-s-triazine rings [24]. While the sharp peak at 812 cm$^{-1}$ was assigned to the respiration pattern of tri-s-triazine units [23]. Interestingly, it was also noted that two new peaks at 2165 cm$^{-1}$ and 1080 cm$^{-1}$ emerged in the FT-IR spectra of all Ti/OCN composites, which were due to the formation of $\equiv$C–N and C–O–C groups after doping treatment, indicating the existence of nitrogen defects resulting in Ti and O co-doping [25, 26].

The microstructure information of Ti/OCN-3 was analyzed by SEM and the corresponding results were displayed in figure 2. As seen in figures 2(a) and (b), Ti/OCN-3 shown a representatively continuous curled layered structure, which could be attributed to the gas templating effect by decomposing of urea and formic acid [27]. In addition, from the results of EDS elemental mapping in figure 2(c) and EDX analysis in figure 2(d), the C, N, O and Ti elements were evenly distributed throughout the whole Ti/OCN-3, indicating that Ti and O atoms have been successfully introduced into the framework of CN.

The detailed microstructure of Ti/OCN-3 was investigated by TEM. As depicted in figures 3(a) and (b), the TEM images of Ti/OCN-3 further confirmed its layered structure, and meanwhile, no black dots representing

![Figure 2. SEM images (a), (b), the corresponding EDS elemental mapping (c) and EDX image (d) of Ti/OCN-3.](image-url)
TiO₂ nanoparticles could be observed, which could be treated as a direct evidence of atomic doping of Ti in CN lattice.

To prove the existence of Ti, O atoms and the chemical structure of Ti/OCN-3, XPS measurement was further carried out. As shown in figure 4(a), the wide-scan XPS spectra of Ti/OCN-3 confirmed the coexistence of C, N, O and Ti elements. In high-resolution C 1s spectra shown in figure 4(b), two typical peaks at 284.69 eV and 287.98 eV were correspondingly stemmed from the adventitious C–C/C=O carbon and N=C–N tertiary carbon in the basic CN aromatic rings [28]. Expectantly, two new peaks at 285.64 eV and 289.09 eV corresponding to C–O–C/H–C≡N and C–O bonds appeared in Ti/OCN-3, suggesting that the O atoms were doped into the matrix of CN heterocycles by bonding with C atoms and cyano groups formed through one-pot thermal calcination [29, 30]. Figure 4(c) presented the N 1s spectra of Ti/OCN-3. The four peaks at 398.41 eV, 399.65 eV, 401.00 eV and 404.10 eV were separately derived from C–N=C(sp²-hybridized nitrogen), N–(C)₃(sp³-hybridized nitrogen), C–N–H (terminal amino functional groups) and the π excitation of CN aromatic structure [31]. Moreover, a weak and wide peak centered at 398.19 eV should be ascribed to the Ti N bonds appeared in Ti/OCN composites, which were probably derived from the synergistic effect of p doped Ti bonded with N atoms in the skeleton of CN [32].

Finally, the light-harvesting behavior of as-synthesized composites was examined by UV–vis DRS. As indicated in figure 5(a), the bare g-C₃N₄ had an intrinsic absorption borderline at 438 nm with negligible light response above 500 nm. In contrast, the absorption edge of Ti/OCN composites gradually redshifted to 516 nm as well as intensive absorption intensities. Moreover, a new absorption band in the region of 450–750 nm emerged in all Ti/OCN composites, which were probably derived from the synergistic effect of p →π* electron transitions arising from the lone pairs of terminal O atoms [33]. Correspondingly, as shown in figure 5(b), the bandgap energy of CN was calculated to be 2.70 eV according to Kubelka-Munk function [34], while the intrinsic bandgap of Ti/OCN composites were gradually narrowed to 2.46, 2.28, 2.04 and 1.92 eV. The considerably reduced bandgap could boost the visible light harvesting efficiency and then promote the generation of electron–hole pairs, which would result in an enhanced photocatalytic activity [35].

The potential of VB and CB for bare CN and Ti/OCN-3 were estimated based on the following formula [36]:

\[ E_{VB} = X - E_o + 1/2E_g \]
\[ E_{CB} = E_{VB} - E_g \]

Therefore, the \(E_{VB}\) edge potentials of CN and Ti/OCN-3 were estimated to be 1.58 eV and 1.25 eV, meanwhile, the \(E_{CB}\) potentials were −1.12 eV and −0.79 eV, respectively.

### 3.2. Photocatalytic inactivation performance

The photocatalytic inactivation efficiency of as-synthesized composites towards *E. cloacae* was evaluated and the results were illustrated in figure 6 and 7. As shown in figure 6(a), the blank control and all the composites in darkness displayed negligible inactivation effects, despite about 0.41 log CFU ml⁻¹ of cell density reduction appeared in Ti/OCN-4 within 60 min. This result indicated that the intrinsic cytotoxicity of these composites on *E. cloacae* was neglectable. Similarly, as shown in figure 6(b), the cell density remained almost constant in visible light control treatment, confirming that no negative effect on *E. cloacae* viability was induced by visible light.

Figure 6(b) illustrated the inactivation results of *E. cloacae* in the presence of composites. Bare CN exhibited a very low inactivation efficiency, and only approximately 1.33 log CFU·ml⁻¹ of cell density reduction was
observed within 60 min of visible light irradiation. Comparatively, after introduction of Ti and O atoms, the inactivation activities of Ti/OCN composites dramatically enhanced, and about 4.04, 5.67 and 5.88 log CFU·ml⁻¹ of inactivated cell density were observed for Ti/OCN-1, Ti/OCN-2 and Ti/OCN-4 composites, respectively. Surprisingly, Ti/OCN-3 exhibited the highest inactivation behavior, achieving 6.20 log CFU·ml⁻¹ of E. cloacae cells totally inactivated, which also demonstrated a highly competitive photocatalytic inactivation activity in comparison with the results reported by literature [18, 37, 38]. The reduced photocatalytic activity of Ti/OCN-4 was mainly due to the excessive doped Ti served as recombination centers, resulting in the reduced utilization efficiency of photoexcited charges [39].

Furthermore, to reveal the details of inactivation process, figure 7(a) visually reflected the number of E. cloacae colonies occurred in the presence of Ti/OCN-3 at different treatment time. The digital photos illustrated that Ti/OCN-3 could inhibit the growth of E. cloacae in a time-dependent manner, causing a gradual decrease in the number of living cells when increasing irradiation time. Meanwhile, the variation in cell membrane integrity of E. cloacae was detected by using fluorescent-based cell Live/Dead staining, in which, two fluorescent DNA
Figure 5. UV–vis DRS (a), the estimated band gaps (b) of pristine CN and Ti/OCN composites.

Figure 6. Time-dependent inactivation of *E. cloacae* with as-prepared composites in the dark (a) and (b) under visible light irradiation.

Figure 7. The digital photos (a) and the fluorescence-based Live/Dead staining (b) of *E. cloacae* treated with Ti/OCN-3 at different irradiation time.
dyes named SYTO9 and PI were employed to stain bacterial cells. As depicted in figure 7(b), before irradiation, *E. cloacae* mixed with Ti/OCN-3 presented intense green fluorescence, indicating that the cells were alive with intact cell membrane [18]. After being treated for 30 min, some cells turned to red fluorescence, suggesting that these cells lost the membrane integrity. Impressively, with prolonged treatment time to 60 min, almost all *E. cloacae* cells changed to red fluorescence, demonstrating that most cells were disrupted, eventually leading to cell death [19].

### 3.3. Improvement photocatalytic inactivation mechanism

Generally, multifarious reactive species, such as holes (h$^+$), electrons (e$^-$), superoxide radicals (·O$_2^-$), and hydroxyl radicals (·OH) participated in the photocatalytic inactivation process [40]. To elucidate the contribution of photoexcited reactive species involved in *E. cloacae* inactivation over Ti/OCN-3, a scavenger-trapping experiment was performed, in which TEMPOL, AO and IPA were selected as the scavengers of ·O$_2^-$, h$^+$ and ·OH, respectively [41]. It could be observed in figures 8(a) and (b), with incorporating of AO and IPA into the photocatalytic system, the inactivation performance was obviously inhibited and the residual living cells reached up to 4.23 and 3.05 log CFU·ml$^{-1}$, respectively, which suggested that h$^+$ and ·OH partially involved in the photocatalytic inactivation process. Moreover, following introduction of TEMPOL, the inactivation efficiency decreased the most and achieved the highest living cells of 5.56 log CFU·ml$^{-1}$, respectively, which suggested that ·O$_2^-$ performed a critical role in photocatalytic inactivation of *E. cloacae* via Ti/OCN-3. Therefore, these results confirmed that ·O$_2^-$, h$^+$ and ·OH worked together in the disinfection process.

To further confirm the generation of ·O$_2^-$ and ·OH during photocatalytic inactivation process, the ESR test on Bruker A300 spectrometer was employed using DMPO as radical capture agents. As exhibited in figures 8(c) and (d), no clear ESR signals appeared in the dark, but the characteristic signals of DMPO··O$_2^-$ and DMPO··OH were captured in both CN and Ti/OCN-3 systems under visible light irradiation. Compared with CN, Ti/OCN-3 system obtained stronger DMPO··O$_2^-$ and DMPO··OH intensities, signifying that abundant of ·O$_2^-$ and ·OH were produced [42]. Meanwhile, it was worth noting that a significantly enhanced intensity of ·O$_2^-$ was achieved compared to the ·OH, which further corroborated the crucial role of ·O$_2^-$ in the photocatalytic inactivation process of Ti/OCN-3.
The efficient separation and migration of photoexcited charge are extraordinarily important for the improvement of photocatalytic performance. Hence, the PL, photocurrent response and EIS properties were monitored to elaborate the photocatalytic inactivation superiority of Ti/OCN-3. The PL spectra of bare CN and Ti/OCN composites was exhibited in figure 9(a). Compared with CN, Ti/OCN composites possessed remarkable decreasing PL intensity, and Ti/OCN-3 displayed the weakest PL intensity, representing that the recombination of photoexcited charge was substantially suppressed, which was beneficial to the improvement of photocatalytic activity [43]. Subsequently, the photocurrent response and EIS measurements were further executed to evaluate the separation efficiency of the photoexcited charges. As shown in figure 9(b), Ti/OCN-3 exhibited over 2.5 folds higher photocurrent density than that of CN, indicating the more efficient separation and transfer of photoexcited charges [44]. Similarly, it could be seen in figure 9(c) that Ti/OCN-3 possessed smaller semicircle diameter of Nyquist plot in comparison with CN, implying the lower charge transfer resistance, which could facilitate the charge separation during photocatalysis [45].

Thus, according to the above-mentioned results and analysis, the possible mechanisms of photocatalytic E. cloacae inactivation over Ti/CON-3 was illustrated in figure 10. Exposed to visible light, the photoexcited electrons were excited efficiently and instantly migrated to the CB of Ti/CON-3, which mainly benefitted from the significant reduction of charge transfer resistance caused by Ti and O co-doping. Meanwhile, the Ti, O atoms and cyano groups on the surface of composites could serve as electron traps to avoid the recombination of charge carriers. Therefore, these photoexcited charges were invested with more possibilities to reduce dissolved O2 to produce more ·O2−, which was mainly due to the more negative CB potential of Ti/CON-3 (−0.79 eV, versus NHE) than that of O2/2− (−0.33 eV, versus NHE) [46]. Subsequently, the produced ·O2− could be further converted to ·OH by combining with H2O [47]. Furthermore, the remained holes located on the VB of Ti/CON-3 with strong oxidation capacity could also attack E. cloacae directly. Therefore, the participation of more ·O2−, ·OH and h+ species ensured Ti/CON-3 to possess a high efficiency on photocatalytic inactivation of E. cloacae.
4. Conclusions

In conclusion, an innovative Ti and O co-doped g-C$_3$N$_4$ was successfully synthesized through a facile one-pot thermal calcination route for the first time. The resulting Ti/OCN composites possessed extended visible light response, efficient suppresses of charges recombination and expediting photoexcited charges separation and transfer, which were benefited from the synergistic effect of Ti and O co-doped in the g-C$_3$N$_4$ framework. Among them, the optimal Ti/OCN-3 exhibited the maximum photocatalytic performance with 6.20 log of live *E. cloacae* totally inactivated within 60 min visible light irradiation. According to radical scavenger and ESR experiments, ·O$_2$•, h$^+$ and ·OH played the main role in photocatalytic *E. cloacae* disinfection process. It is expected that the effective co-doping strategy presented here can be explored to construct g-C$_3$N$_4$-based photocatalyst for water disinfection.

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Declaration of competing interest

The authors formally announced that they have no known correlative financial interests or personal relationships that could influence this research work.

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