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Article

Generation and transformation of ROS on g-C₃N₄ for efficient photocatalytic NO removal: A combined *in situ* DRIFTS and DFT investigation

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ABSTRACT

Understanding the performance of reactive oxygen species (ROS) in photocatalysis is pivotal for advancing their application in environmental remediation. However, techniques for investigating the generation and transformation mechanism of ROS have been largely overlooked. In this study, considering g-C₃N₄ to be a model photocatalyst, we have focused on the ROS generation and transformation for efficient photocatalytic NO removal. It was found that the key to improving the photocatalysis performance was to enhance the ROS transformation from •O₂⁻ to •OH, elevating the production of •OH. The ROS directly participate in the photocatalytic NO removal and tailor the rate-determining step, which is required to overcome the high activation energy of the intermediate conversion. Using a closely combined experimental and theoretical method, this work provides a new protocol to investigate the ROS behavior on g-C₃N₄ for effective NO removal and clarifies the reaction mechanism at the atomic level, which enriches the understanding of ROS in photocatalytic environmental remediation.

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1. Introduction

Reactive radicals are of key importance in many fields of degradation of environmental pollutants, such as heterogeneous catalysis for the NO_x/VOCs removal [1,2], advanced oxidation for the organic pollutants treatment of wastewater [3,4]

and photocatalytic environmental remediation [5–7]. Thus, the generation and transformation of active radicals gives rise to a general concern in the environmental and catalysis fields. As in photocatalysis, the redox reactivity is brought about by photo-excited electrons (e⁻) and holes (h⁺) produced in the photocatalysts [8–10]. Several active radicals, which are the major

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source of oxidation capacity, are subsequently generated through the redox reaction with e^- and h^+ . As photocatalysis normally takes place involving oxygen and water as the reactants, the reactive radicals converted by oxygen and water are thus called reactive oxygen species (ROS) [11].

Since ROS are the primary oxidants in photocatalytic environmental remediation, the generation and transformation of ROS is important to understand the photocatalysis mechanism, which improves the photocatalytic efficiency and facilitates practical applications. Four major ROS are commonly recognized, which include superoxide radical ($\bullet O_2^-$), singlet oxygen (1O_2), hydrogen peroxide (H_2O_2) and hydroxyl radical ($\bullet OH$) [12–14]. In a typical photocatalysis reaction, since both reduction and oxidation occur simultaneously, ROS could be generated through the h^+ induced stepwise oxidation path of H_2O , along with the production of $\bullet OH$, H_2O_2 , $\bullet O_2^-$ and 1O_2 [15,16]. On the other hand, the stepwise reduction of O_2 generates $\bullet O_2^-$, H_2O_2 and $\bullet OH$ [17]. Among these, $\bullet OH$ and $\bullet O_2^-$ are recognized as the most stable radicals during the reaction, which can be effectively detected and are regarded as important reactants in the photocatalytic oxidation process.

In typical photocatalysts, their band structures, which possess wide bandgaps and suitable band edges to align the redox potential of $O_2/\bullet O_2^-$ and $OH^-/\bullet OH$ [18], normally provide a tolerated band range to produce both $\bullet O_2^-$ and $\bullet OH$ [19,20]. Hence, the generation mechanism of ROS can be directly summarized as: $O_2 + e^- \rightarrow \bullet O_2^-$ and $OH^- + h^+ \rightarrow \bullet OH$ [21]. However, in some new types of two-dimensional photocatalysts such as graphite carbon nitride ($g-C_3N_4$) [22–26] and MoS_2 [27–29], their valence band (VB) edge is not low enough to directly oxidize H_2O to generate $\bullet OH$ [30,31]. Thus, $\bullet OH$ are generated through the O_2 reduction at the conduction band (CB), along the paths: $O_2 + e^- \rightarrow \bullet O_2^-$, $\bullet O_2^- + 2H^+ + e^- \rightarrow H_2O_2$ and $H_2O_2 \rightarrow 2\bullet OH$ [32–34]. However, the rate-determining step in the reaction pathways has not been clarified yet. As a major ROS with high oxidation capacity, $\bullet OH$ manifests much more importance in photocatalytic remediation than $\bullet O_2^-$ [11,35]. The generation mechanism of $\bullet OH$ is thus required to be further investigated. Moreover, understanding the conversion of ROS driven pollutants is highly desirable, in order to direct the design of photocatalysts toward more efficient environmental and energy applications.

In this study, we have synthesized $g-C_3N_4$ with a facile method based on the property that it exhibits photocatalytic activity toward NO removal. By a combined *in situ* diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) and density functional theory (DFT) method, it is determined that the H_2O_2 dissociation is the rate-determining step of $\bullet OH$ generation in $g-C_3N_4$ and the reaction coordinates of ROS driven NO removal are presented. It is found that the oxidation capacity of $g-C_3N_4$ largely depends on the production and oxidation capacity of $\bullet OH$, rather than $\bullet O_2^-$. This article presents a research protocol to investigate the photocatalytic ROS generation and transformation, and clarifies the reaction mechanism of ROS driven NO removal. This approach could be extended to many environmental and catalytic applications.

2. Experimental

2.1. Photocatalyst fabrication

All chemicals used in this study were analytical grade and no further treatment was required. The CN product was fabricated with a reported method [36]. In a typical synthesis procedure, 10 g thiourea was added to a crucible with 20 mL deionized water. The mixed solution was dried at 80 °C for recrystallization. Then, the precursor was placed in a semi-closed alumina crucible with a cover and roasted at 550 °C for 2 h with a heating rate of 15 °C/min in static air. After the calcining treatment, the crucible was cooled to room temperature.

2.2. Characterization

X-ray diffraction (XRD) was used to investigate the crystal phases with Cu K_α radiation (model D/max RA, Rigaku Co. Japan). Scanning electron microscopy (SEM, model JSM-6490, JEOL, Japan) and transmission electron microscopy (TEM, JEM-2010, Japan) were used to investigate the morphology. Fourier transform infrared (FT-IR) spectra were recorded on a Nicolet Nexus spectrometer (IRPrestige-21, SHIMADAZU, Japan) using samples embedded in KBr pellets. A scanning UV-vis spectrophotometer (TU-1901, China) was used to characterize the UV-vis diffuse reflectance spectrometry. The photoluminescence (PL) spectra were investigated using a fluorescence spectrophotometer (Edinburgh Instruments, FLSP-920) equipped with a Xe lamp with an excitation wavelength of 420 nm. Electron spin resonance (ESR) spectra of chemical radicals were obtained on a JES FA200 spectrometer to determine the involvement of the ROS in methanol dispersion for DMPO- $\bullet O_2^-$ and aqueous dispersion for DMPO- $\bullet OH$, respectively.

2.3. Photocatalytic efficiency evaluation

The photocatalytic activity was investigated via the NO removal at 500 ppb in a self-designed continuous-flow reactor (Scheme S1 in the Supplementary Information). The rectangular reactor (30 cm × 15 cm × 10 cm) is made of polymeric glass and covered with Saint-Glass. The as-prepared sample (0.20 g) was dispersed onto two glass dishes for testing. A 150 W tungsten halogen lamp (0.16 W/cm²) was vertically placed above the reactor. After the adsorption-desorption equilibrium was achieved, the lamp was turned on. The NO gas was acquired from a compressed gas cylinder at a concentration of 100 ppm of NO (N_2 balance). The initial concentration of NO was diluted to about 550 ppb by a zero air generator and the relative humidity (RH) level of the NO flow was controlled at 50% by passing zero air stream through a humidification chamber. The flow rates of the air stream and NO were controlled at 2.4 L/min and 24 mL/min, respectively. The NO removal ratio (η) was calculated as

$$\eta(\%) = (1 - C/C_0) \times 100\% \quad (1)$$

where C and C_0 refer to the NO concentration in the outlet stream and feeding stream, respectively. The resulting final product (nitrate) was removed by washing the dilute NaOH

solution.

2.4. In situ DRIFTS investigation

In situ DRIFTS measurements were performed with a TENSOR II FT-IR spectrometer (Bruker) which was equipped with an *in situ* diffuse-reflectance cell (Harrick) and reaction chamber (HVC), as shown in Scheme S2. High-purity He, O₂, and 100 ppm of NO were mixed and fed into the reaction system. A tri-way ball valve was used to switch between the target gas (NO) and purge gas (He). The flow rate of total gas was set to 100 mL/min, and the concentration of NO was adjusted to 50 ppm by dilution with O₂. The chamber was enclosed in a dome having three windows, two for IR light entrance and detection, and one for illuminating the photocatalyst. The observation window was made of UV quartz and the IR windows were made of ZnSe. A Xe lamp (MVL-210, Japan) was used as the irradiating light source. Before the measurements, prepared samples were pretreated for 20 min at 300 °C to remove the adsorbed species on the catalyst surface. The figure of the reaction chamber under the working conditions is given in Fig. S1.

2.5. Theoretical calculations

All the spin-polarized DFT-D2 calculations were performed with the “Vienna *ab initio* simulation package” (VASP 5.4.1 code) [37,38], using a generalized gradient correlation functional with the exchange and correlation functional of the Perdew-Burke-Ernzerhof [39]. A plane-wave basis set with the cut-off energy at 450 eV within the framework of the projector-augmented wave method was used [40]. The Gaussian smearing width was set to the value of 0.2 eV. K points in the Brillouin zone were sampled with a $5 \times 5 \times 1$ mesh. The van der Waals correction in the D2 calculations was set at the default parameters by the Grimme’s method [41]. All atoms were al-

lowed to relax at the convergence below 0.01 eV Å⁻¹. A hybrid functional based on the Heyd-Scuseria-Ernzerhof (HSE06) method was applied to estimate the exact band structures [42]. The reaction coordinates were calculated with the climbing image nudged elastic band (CI-NEB) code, which located the minimum energy pathways (MEPs) from the initial state (IS) to its final state (FS) [43]. The transition state (TS) was verified with a single imaginary frequency [44]. A $2 \times 2 \times 3$ supercell of g-C₃N₄ that includes 168 atoms was designed and relaxed. The lattice parameters were set to $14 \times 16 \times 20$ Å with a vacuum region of 10 Å. The adsorption energy (E_{ads}) is defined as

$$E_{\text{ads}} = E_{\text{tot}} - (E_{\text{mol}} + E_{\text{CN/OCN-K-CN}}) \quad (2)$$

where E_{tot} , E_{mol} and $E_{\text{CN/OCN-K-CN}}$ depict the total energy of the adsorption complex, the isolated molecule and the CN/OCN-K-CN, respectively.

3. Results and discussion

3.1. Crystal and morphological structure

XRD was used to investigate the crystal structure of the as-prepared g-C₃N₄. As shown in Fig. 1(a), two characteristic peaks at 13.1° and 27.4° were prominent, which were assigned to the (100) and (002) facets of g-C₃N₄, respectively. This result reflects the generation of the in-plane repeating aromatic units and interlayer reflection in g-C₃N₄, which agrees with the published works [24]. The FT-IR spectrum was further studied to verify the basic substructures of g-C₃N₄ polymers. As shown in Fig. 1(b), the absorption bands in the 1200–1700 cm⁻¹ range and the sharp peak at 810 cm⁻¹ are both attributed to the stretching vibration of the C₆N₇ units [45], demonstrating the existence of basic atomic structure. Moreover, the absorption bands (3100–3500 and 890 cm⁻¹) are maintained, which correspond to the N–H components stemming from uncondensed amino groups [46]. In addition, there is evidence of the

Fig. 1. XRD pattern (a), FT-IR spectrum (b), SEM image (c) and TEM image of g-C₃N₄ (d).

stretching vibration of N=C=N (2212 cm⁻¹). These results illustrate that the g-C₃N₄ photocatalyst has been successfully synthesized. The morphology and microstructure of g-C₃N₄ were examined by SEM (Fig. 1(c)) and TEM (Fig. 1(d)). These analyses showed that the typical layered nanosheets were distinctly perceived.

3.2. Photoelectric properties and band structure

The PL spectrum was then studied to investigate the light response and charge dynamics (Fig. 2(a)). It was found that g-C₃N₄ possessed light absorption and charge separation properties in the visible light region ($\lambda \geq 420$ nm), which manifests great potential for ROS generation and NO removal under visible-light irradiation. The light absorption properties were measured by using a combined experimental and theoretical method. As depicted by the UV-Vis DRS spectrum (Fig. 2(b)), the light harvesting capacity of g-C₃N₄ in the visible light region is authenticated, confirming the PL result. The calculated absorption spectrum also supports the experimental result. The band energy was measured from the intercept of tangents to the plot of $(\alpha h\nu)^{1/2}$ vs. photoenergy (Fig. 2(b), inset). It was estimated that the energy gap (E_g) of g-C₃N₄ was ca. 2.39 eV. We then calculated the density of states (DOS) using a hybrid DFT method, measuring the E_g to be 2.33 eV, which is in good accordance with the experimental value. These theoretical results reveal that the constructed DFT model of g-C₃N₄ (Fig. S2) agrees with the experimental prepared sample.

The energies of the valence band (E_{VB}) and conduction band (E_{CB}) are subsequently calculated, to investigate the exact band structure of g-C₃N₄ (Fig. 2(d)), by the following equations:

$$E_{VB} = X - E^e + 0.5E_g \quad (3)$$

$$E_{CB} = E_{VB} - E_g \quad (4)$$

where X and E^e represent the electronegativity and energy of free electrons in hydrogen atoms (about 4.5 eV), respectively. E_g is the gap energy calculated from the UV-Vis DRS spectrum.

Based on the band structure of g-C₃N₄, it is apparent that $\bullet\text{O}_2^-$ can be directly produced via O_2 reduction at the VB, while the CB position is not low enough to oxidize H_2O for $\bullet\text{OH}$ generation. As the photo-oxidation capacity mainly originates from ROS, it is required that the generation and transformation mechanism of ROS in g-C₃N₄ should be further investigated.

3.3. ROS generation and transformation

The DMPO spin-trapping ESR experiment is utilized in methanol dispersion for DMPO- $\bullet\text{O}_2^-$ (Fig. 3(a)) and aqueous dispersion for DMPO- $\bullet\text{OH}$ (Fig. 3(b)), respectively. As expected, the signals of $\bullet\text{O}_2^-$ and $\bullet\text{OH}$ are detected and increase as the irradiation time progresses, which indicates that both $\bullet\text{O}_2^-$ and $\bullet\text{OH}$ can be produced in g-C₃N₄. Based on the band structure and ESR results, the generation and transformation pathways of ROS can be summarized as follows:

As stated before, $\bullet\text{OH}$ possesses higher oxidation capacity than $\bullet\text{O}_2^-$ and cannot be directly generated in g-C₃N₄. In order to investigate the transformation mechanism of ROS, we subsequently applied DFT calculations to estimate the reaction coordinates at the atomic level. As Fig. 3(c) shows (Refer Fig. S3 for the detailed minimum reaction pathways), energy and charge transfer took place once the isolated O_2 molecule approached the g-C₃N₄, which leads to a significant energy release. The energy-favorable adsorption and activation of O_2 contributes to the formation of $\bullet\text{O}_2^-$ (Path 1), which agrees with

Fig. 2. PL spectrum (a), experimental UV-vis spectrum (above) and calculated absorption spectrum (below) (b), calculated density of states (DOS) (c), the Fermi level is set to 0 eV and proposed band structure (d).

Fig. 3. DMPO-ESR spectra in methanol dispersion for $\bullet\text{O}_2^-$ (a) and aqueous dispersion for $\bullet\text{OH}$ (b), (c) calculated reaction coordinates of ROS generation and transformation in g- C_3N_4 . Red and pink spheres depict O and H atoms, respectively.

the ESR result. Moreover, $\bullet\text{O}_2^-$ is further reduced to generate $\bullet\text{O}_2^{2-}$, combining with two hydrogen ions (H^+) to generate H_2O_2 (Path 2) and dissociation of the O–O bond in H_2O_2 generates $\bullet\text{OH}$ (Path 3). Notably, the dissociation required 0.48 eV to materialize (TS3), making it the rate-determining step in $\bullet\text{OH}$ generation in g- C_3N_4 . Hence, tailoring the rate-determining step and enhancing the production of $\bullet\text{OH}$ are of key importance in the design of g- C_3N_4 based photocatalysts for environmental remediation.

3.4. Photocatalytic NO removal and reaction mechanism

The photocatalytic performance of the as-prepared g- C_3N_4 was evaluated for NO removal under visible-light irradiation ($\lambda > 420$ nm). As shown in Fig. 4(a), the maximum NO removal ratio (28%) was reached in approximately 5 min. We further carried out a five-time cycling test and after the first two runs,

some decrease in activity was observed. But the removal ratio remained constant for the five cycles (19%), which indicates that NO can be removed by photocatalysis of g- C_3N_4 .

In order to understand the reaction process and clarify the mechanism in detail, *in situ* DRIFTS was performed to measure the dynamic reaction intermediates and final products in a time sequence. The baseline was measured before the NO injection into the reaction chamber. In dark conditions, the absorption bands of NO appear once the gas flow contacts g- C_3N_4 (Fig. 5(a)). The absorption bands of NO (2198 cm^{-1}), N_2O (2285 cm^{-1}) and NO_2 (2067 cm^{-1}) were prominently detected, which is assigned to the chemical adsorption of NO in g- C_3N_4 [47,48]. As the adsorption time increases, the intensities of these peaks gradually increased, indicating the accumulation of reactants and reaction intermediates on the catalyst surface. It is notable that some new absorption bands were developed, namely NO_2^- (1153, 1133, 1088, 890 and 877 cm^{-1}) and NO_3^- (1011 cm^{-1})

Fig. 4. Photocatalytic activity evaluation for NO removal (a) and cycling test (b).

Fig. 5. *In situ* DRIFT spectra of NO adsorption in dark conditions (a) and the reaction with light on (b).

[49,50]. The accumulation of these reaction products is presumed to arise from the two-coordinated pyridine-like N atoms on g-C₃N₄, which possess an extra lone-pair of electrons to facilitate the NO oxidation in dark conditions [51].

After the adsorption equilibrium (Fig. 5(b)) is reached, the *in situ* DRIFT was subsequently recorded under visible-light irradiation. As shown in Fig. 5(b), once the light was turned on, the absorption bands of the reactants, NO and N₂O, are no longer detected, which illustrates that NO could be effectively oxidized in g-C₃N₄. Moreover, the reaction intermediates (NO₂, 2070 and 933 cm⁻¹) are also detected, which restrain the over-all NO photocatalytic removal and hamper the generation of the final products. Over time, the absorption bands of the reaction products, NO₂⁻ (1100, 1000, 883 and 866 cm⁻¹) and

NO₃⁻ (1044 and 984 cm⁻¹ are gradually intensified) [52]. All the observed DRIFTS bands of the adsorbed species are listed in Tables S1 and S2. As the ROS are a major source of the oxidation capacity, the primary reaction pathways are proposed as follows:

Table 1

Calculated imaginary frequency (f/i) of ROS transformation and NO oxidation on CN.

Reaction path	1	2	3	4	5	6	7
$f/i, \text{cm}^{-1}$	—	—	510	452	612	257	424

Fig. 6. Calculated reaction coordinates and minimum reaction pathways (MEPs) for •O₂⁻ (a–c) and •OH (d–f) driven NO photocatalytic oxidation. All energies are given in eV. Blue, brown, red and pink spheres depict N, C, O and H atoms, respectively.

3.5. Calculation of reaction coordinates

In order to elucidate the primary reaction mechanism that is proposed by the *in situ* DRIFTS, the ROS driven reaction coordinates were thus calculated using the CI-NEB method with a single imaginary frequency (f/i , Table 1). As shown in Fig. 6, $\bullet\text{O}_2^-$ driven NO oxidation was composed of two elementary reactions (Paths 4 and 5, Fig. 6(a)–6(c)). It was found that path 4 required an activation energy (E_a) of 0.47 eV in order to transfer NO to NO_2^- . However, the oxidation of NO_2 (Path 5) is more difficult as the activation energy for climbing over the TS5 is as high as 0.66 eV, leading to a lower reaction rate than that in Path 4. These results confirm our experimental results that NO_2 oxidation is the rate-determining step in photocatalytic NO removal.

$\bullet\text{OH}$ driven reactions were further conducted (Fig. 6(d)–6(f)), which manifest similar patterns with $\bullet\text{O}_2^-$. The E_a for NO_2 oxidation (Path 7, 0.46 eV) is higher than that for NO oxidation (Path 6, 0.27 eV). Specifically, as the rate-determining step, NO_2 is more likely to be oxidized by $\bullet\text{OH}$ (0.46 eV) than $\bullet\text{O}_2^-$ (0.66 eV). Based on these results, it can be concluded that the key to enhancing the photocatalytic efficiency of NO removal in $\text{g-C}_3\text{N}_4$ is to facilitate the ROS transformation from $\bullet\text{O}_2^-$ to $\bullet\text{OH}$.

4. Conclusions

In summary, we have proposed a combined experimental and theoretical approach to investigate the generation and transformation of ROS in $\text{g-C}_3\text{N}_4$ for effective photocatalytic NO removal. This work provided direct evidence that the generation of $\bullet\text{OH}$ in $\text{g-C}_3\text{N}_4$ arises from the O_2 reduction at CB, and the H_2O_2 dissociation is the rate-determining step for $\bullet\text{OH}$ formation. Moreover, the reaction mechanism of NO oxidation is shown by *in situ* DRIFTS and DFT calculations. It certified that NO_2 oxidation was the rate-determining step, which suppresses the over-all NO removal and leads to the accumulation of toxic intermediates. In order to overcome the rather high activation energy, the production of $\bullet\text{OH}$ requires to be enhanced. This work reveals that understanding and tailoring the generation and transformation of ROS is crucial for elevating the performance of photocatalysts in environmental and energy-related applications.

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Graphical Abstract

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Generation and transformation of ROS on g-C₃N₄ for efficient photocatalytic NO removal: A combined *in situ* DRIFTS and DFT investigation

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Using a closely combined theoretical and experimental approach, the reactive oxygen species behavior on g-C₃N₄ for effective NO removal and the reaction mechanism at the atomic level were clarified, which enriches the understanding of ROS in photocatalytic environmental remediation.

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活性氧自由基生成及传输对 g-C₃N₄ 上 NO 光催化去除的影响： 原位红外光谱与计算模拟结合法

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摘要: 石墨相氮化碳 (g-C₃N₄) 具有独特的二维层状结构和合适的能带结构, 因而在可见光催化领域广受关注. 尤其是在可见光去除环境污染物领域, 得到了较为充分的研究与应用. 然而 g-C₃N₄ 去除环境污机理的反应机理尚不明确. 因此, 本文采用理论计算与实验高度结合的研究方法, 以光催化 NO 去除为例, 深入阐述了光照下 g-C₃N₄ 表面活性氧物种 (ROS) 的生成及转化过程, 及其介导下的 NO 光催化氧化机理.

X 射线衍射结果表明, g-C₃N₄ 是三嗪环层内聚合后层层堆叠而成, 并由红外光谱确定了其表面的官能团类型. 该结构经扫描电镜和透射电镜得到了进一步的验证. 采用光致激发谱和紫外可见漫反射光谱等实验表征与密度泛函理论计算结合的光电性质分析, 我们发现, g-C₃N₄ 在可见光下具有一定的响应, 这为其在光催化去除 NO 中奠定了基础. 同时, 其价带位置过高, 无法自行产生氧化性较强的羟基自由基 (·OH).

电子自旋共振技术结果表明 g-C₃N₄ 在光照下能捕获到·O₂⁻和·OH 两种活性自由基. 采用反应路径计算发现, ·OH 是由

$\cdot\text{O}_2^-$ 在导带上逐步得到电子被还原而生成, 其中的速率控制步骤是 H_2O_2 的解离. 因此, 促进 O_2 分子的吸附和活化和克服 H_2O_2 解离的反应活化能是产生 $\cdot\text{OH}$ 和提升 $\text{g-C}_3\text{N}_4$ 光催化氧化活性的关键.

采用原位红外光谱技术对 $\text{g-C}_3\text{N}_4$ 上 NO 的氧化去除过程进行了表征, 发现其主要中间产物为 NO_2 , 主要终产物为 NO_2^- 和 NO_3^- . 采用反应路径计算对该反应过程进行了理论模拟, 发现在 $\cdot\text{O}_2^-$ 介导下, 最高反应活化能为 0.66 eV, 而在 $\cdot\text{OH}$ 介导下, 该活化能降低至 0.46 eV, 表明 $\cdot\text{OH}$ 的氧化性要明显强于 $\cdot\text{O}_2^-$. 总之, 本文采用一种可行的、高度结合的实验与计算手段研究了 $\text{g-C}_3\text{N}_4$ 上 ROS 的生成及转化过程及其对 NO 去除的反应历程, 在原子尺度揭示了该反应的机理, 加深了对 ROS 在光催化环境污染降解过程中作用的理解.

关键词: 光催化; 活性氧物种; 石墨相氮化碳; NO 去除; 反应机理

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