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5 Improved Visible Light Activity of Copper Oxide/Carbon Nitride Photocatalysts Prepared by Photodeposition for Phenol Degradation

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Abstract. A series of copper(II) oxide deposited on carbon nitride (CuO/CN) as visible light-active photocatalyst was successfully prepared via a photodeposition method. The CuO modification was carried out to improve the photocatalytic activity of CN having fast electron-hole recombination. The CuO loading was varied from 0.05 to 0.5 wt% and the photodeposition was carried out at room temperature under UV light illumination. The CuO/CN samples were confirmed to have similar optical properties and functional groups to those of the unmodified CN. Meanwhile, the emission intensity of the CN decreased with the increase of the copper species loading, which could correspond to the suppression of charge recombination on the CN. After a 24-h reaction under visible light irradiation, the best photocatalyst, *i.e.* the CuO(0.1)/CN, gave 41.7% phenol degradation, which was almost two times higher than the unmodified CN (21.3%). However, the photocatalytic activity decreased when the added copper species was more than 0.1 wt%. Therefore, the optimum amount of copper species deposited on the CN surface would have a prominent contribution to improve the photocatalytic activity. It was also confirmed that holes, hydroxyl radicals, and superoxide radicals were important for the photocatalytic degradation of phenol on the CuO(0.1)/CN.

1. Introduction

The role of photocatalytic reaction in the toxic organic pollutant degradation has gained much interest in the last decades, as this reaction could be performed under mild conditions by solar light irradiation [1]. It is proven that this safe process allows the organic pollutant to be decomposed into carbon dioxide, water, and benign minerals by involving reactive oxygen species (ROS) [2], thus simplify the further steps in water treatment. As this green technology process has the potential to be applied in practical using sunlight irradiation, the selection of suitable photocatalyst that can work effectively in the wider visible light region becomes a crucial approach. Appearing as the yellow colored-solid, carbon nitride (CN) photocatalyst is considered to be an active photocatalyst under visible light illumination having light absorption ability up to the blue light region (ca. 460 nm) [3,4]. Moreover, the CN also exhibits good resistance both in the various chemical attack conditions and in the heating thermal up to 600°C, implying its high chemical and thermal stability that are raised from its graphite-analog structure consisting of strong C-N bonds [5,6].



Unfortunately, the photocatalytic activity of CN under visible light irradiation is still limited by its rapid recombination rate of electron-hole pairs and narrow light absorption bandwidth [7]. Therefore, the modification of CN is importantly needed. For this reason, the development of a CN-based heterostructure has been widely adopted to repress these shortcomings [8,9]. Among the various CN composite heterostructures, the metal species/CN composites have been frequently reported to suppress the recombination of photogenerated charges, which subsequently improved the photocatalytic activity of the CN [10-12]. Since there is a different position of the conduction band (CB) and valence band (VB) between metal species and CN in the composite structure, the suitable band levels from that difference would produce a driving force for efficient transfer and separation process of electron-hole pairs [11]. In this case, the metal species would be considered as the electron trapper from the conduction band of CN [13], which could reduce the recombination of electron-hole pairs and thus would give a beneficial impact on the photocatalytic performance.

Particularly for photodegradation of organic pollutants under visible light irradiation, it has been reported that introducing the noble metal/CN [13] and other metal species in their oxide forms [14-16] can enhance the photocatalytic activity of CN in comparison to the unmodified CN. Apart from these reported metal species, copper is attractive as the potential metal species owing to its inexpensive, non-corrosive, abundant availability on the earth, and similar properties with gold and silver metals [17]. This makes copper species a promising material for various energy applications [18, 19], including in the photocatalysis field [20-29]. Moreover, the narrow bandgap of CuO (1.3–2.1 eV) indicates their ability to absorb visible light, supporting its use as a favorable semiconductor for the photocatalytic reaction [20]. Therefore, in terms of being composited with the CN, the CuO/CN has been reported as an effective photocatalyst for degradation of dyes [21], thermal decomposition of ammonium perchlorate [22], photoreduction carbon dioxide [23], and degradation of salicylic acid [24].

Common methods to prepare CuO/CN are using hydrothermal [21], facile precipitation [22], and impregnation [30], which all of them need heat treatment. The use of heat treatment would cause aggregation of the metal oxide and if the temperature of the heat treatment is too high, the decomposition of the CN could not be avoided. It has been reported previously that the photodeposition method could produce Fe₂O₃/TiO₂ nanocomposite with better catalyst properties and superior photocatalytic activity for the degradation of 2,4-dichlorophenoxyacetic acid as compared to the impregnation method which involved a heat treatment process [31]. The Ag/CN composite prepared by photodeposition method also showed an increased photocatalytic activity under visible light irradiation in comparison to the unmodified CN [13]. Hence, the photodeposition method without employing the heat treatment can be suggested to prepare the CuO/CN catalyst.

When using the photodeposition method, the formed copper species on the titanium dioxide (TiO₂) was clarified to be highly dispersed of Cu(II) species [25]. In this study, we studied the effect of depositing CuO to the CN surface by the photodeposition method on the properties and photocatalytic activity of the CN. The photocatalytic activity was evaluated under visible light irradiation for the degradation of phenol, which is known as a toxic compound for human health and the environment [32]. We observed that the optimum amount of CuO deposited on the CN surface resulted in high photocatalytic activity, and thus, the prominent contribution of CuO optimum amount was discussed. Besides, the scavenging experiment was also studied to understand the important role of active species contributing to the photocatalytic activity of the CuO/CN photocatalyst and propose the charge transfer mechanism involved in the reaction.

2. Materials and Methods

2.1. Materials

All chemicals used in this study were available commercially and did not receive any purifications before the usage. Urea (CO(NH₂)₂, 98%, Sigma-Aldrich) was used as the precursor for the synthesis of the CN, while copper(II) nitrate trihydrate (Cu(NO₃)₂·3H₂O, 99–104%, Sigma-Aldrich) was used as the copper precursor for the preparation of CuO/CN samples. Phenol (C₆H₅OH, 99.5%, Merck) was used

as the pollutant model in the photocatalytic activity test. For the scavenging experiment, ammonium oxalate ((NH₄)₂C₂O₄·H₂O, 99.5–101%, Merck) was used as a hole species scavenger, *tert*-butanol (C₄H₉OH, 99.5%, Merck) was used as a hydroxyl radical scavenger, and ascorbic acid (C₆H₈O₆, 99.5%, Chameleon Reagent, Kishida Chemicals) was used as a superoxide radical scavenger.

2.2. Preparation of CN and CuO (x)/CN photocatalysts

CN was synthesized by heating urea (25 g) at 550 °C under a limited air atmosphere for 4 h with a heating rate of 2.2 °C min⁻¹ to reach the final temperature of 550 °C. The yellow solid was obtained after the calcination and then was ground into fine powder to be used as the photocatalyst. The CuO/CN composites were prepared by the photodeposition method. The CN photocatalyst (0.1 g) was dispersed in 20 ml of water/methanol mixture (1:1 v/v). The required amount of copper species from the copper(II) nitrate trihydrate solution was then added to the mixture. For making a homogeneous suspension, the mixture was sonicated for 1 min, followed by stirring under irradiation of ultraviolet (UV) lamp (UVLS-28 EL Series, 8 W, at 365 nm) from the top at room temperature for 5 h. Afterward, the separated solid of CuO/CN composite was washed a few times by using methanol and distilled water. The solid was finally dried in an oven at 80 °C overnight. The prepared samples were labeled as CuO(x)/CN, where x represents the ratio of added CuO to CN *i.e.*, 0.05, 0.1, 0.2, and 0.5 wt%.

2.3. Characterization of Photocatalysts

Fourier transform infrared (FTIR) spectra of all CN samples were measured by JASCO FT/IR-6800 spectrometer using attenuated total reflection (ATR) technique in the scan range of 400–4000 cm⁻¹, where the air was recorded as the baseline. The diffuse reflectance ultraviolet-visible (DR UV-vis) spectra were obtained by JASCO V-760 UV-vis spectrophotometer in the scan range of 225–600 nm with scan speed 100 nm min⁻¹, at room temperature. For the fluorescence properties, both excitation and emission spectra were measured by JASCO FP-8500 spectrofluorometer with very low sensitivity mode. The excitation spectra were monitored at the emission wavelength of 442 nm, while the emission spectra were monitored at three excitation wavelengths of 277, 331 nm, and 369 nm.

2.4. Photocatalytic Activity Test

Photocatalytic degradation of phenol under ambient conditions was used to evaluate the photocatalytic activity of the CN and the CuO/CN composites. In the 100 ml of a round bottom flask, the photocatalyst (50 mg) was dispersed in 50 ml of aqueous phenol (70 mg L⁻¹). To reach the adsorption equilibrium between the photocatalyst and phenol before visible-light illumination, the mixture was stirred under the dark condition for 2 h. While the mixture was stirred constantly, the mixture was then exposed to a visible-light lamp (Dolan-Jenner Fiber Lite MI-157, 150 W halogen lamp) for 24 h. Upon the completion of visible-light lamp exposure, the solution was separated from the photocatalyst by using a filter syringe equipped with a 0.2 μm nylon membrane filter. The remaining concentration of phenol after the reaction was determined using a high-performance liquid chromatography instrument (HPLC Shimadzu LC-20AT, with a photodiode array (PDA) detector and column C-18). The eluent used in this HPLC analysis was acetonitrile 100% and the concentration of phenol was monitored at 272 nm. The percentage of phenol degradation was determined by using the following equation (1), where C₀ is the initial concentration of phenol and C is the concentration phenol after reaction under visible light irradiation.

$$\text{Percentage of phenol degradation (\%)} = \frac{C_0 - C}{C_0} \times 100 \quad (1)$$

The investigation of the responsible active species for phenol photocatalytic degradation was carried out using a scavenging experiment on the CuO(0.1)/CN composite, which showed the best photocatalytic activity in this work. Scavenger agents used in this experiment were ammonium oxalate, *tert*-butanol, and ascorbic acid for scavenging holes, hydroxyl radicals, and superoxide radicals, respectively. The photocatalytic activity test in the presence of scavenger agents was conducted under irradiation of the visible light lamp (Dolan-Jenner Fiber Lite MI-150, 150 W halogen lamp) for 24 h. It was noted here that the light intensity used for this scavenging experiment was lower than that used for

photocatalytic evaluation. However, it was confirmed that the trend of the activity remained the same and the light intensity difference would not affect the content of the discussion. To ensure that the scavenger agents would react to the active species formed during the photocatalytic reaction, an excess amount of scavenger agent was added into the phenol solution, which was ten times from that of the phenol amount in mol ratio.

3. Results and Discussions

3.1. Photocatalysts Properties

Structural properties of CN and CuO/CN samples in this study were investigated by FTIR spectroscopy. From the FTIR spectra shown in Figure 1, the functional groups of the CN and its composites were identified by the appearance of some vibration bands. As the CN structure consists of a triazine ring with single and double bonds between carbon and nitrogen, the bending mode of heterocyclic 1,3,5-triazine ring was observed at 810 cm^{-1} , while the C-N heterocycles stretching mode was observed at $1200\text{--}1640\text{ cm}^{-1}$ [4,33], where the C=N stretching mode appeared at 1632 cm^{-1} . Furthermore, the stretching mode of the uncondensed C=O group from the urea precursor was also detected at 1680 cm^{-1} . The observed vibration bands around $3000\text{--}3300\text{ cm}^{-1}$ were attributed to the presence of an uncondensed amino group (N-H bond) and the hydroxyl group of adsorbed water (O-H bond) on the CN structure [4,33,34]. Therefore, the FTIR spectra confirmed the successful formation of the CN structure on the CN and CuO/CN samples. While CuO nanoparticle was reported to give a vibration band at 532 cm^{-1} [35], such vibration band could not be observed on the CuO/CN samples. This could be due to the low amount of added copper species on the surface of the CN. The addition of copper species did not give a significant effect on the functional groups of the CN structure, as have been reported in other literature [22,24,36].

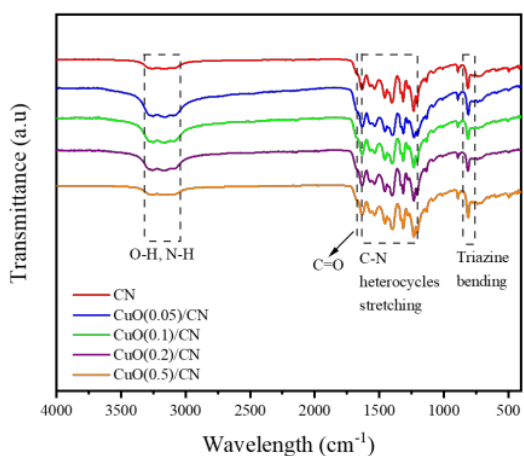


Figure 1. FTIR spectra of the unmodified CN and all CuO/CN composites.

The optical absorption properties of unmodified CN and CuO/CN samples were investigated by DR UV-vis spectroscopy and the results are shown in Figure 2(a). All the samples showed light absorption in both UV and visible regions up to the wavelength of 430 nm , which was associated with the blue light absorption [37]. Two strong absorption peaks of the CN sample appeared at 287 and 370 nm , which could be corresponded to the $\pi \rightarrow \pi^*$ charge transfer at the C=N groups in the aromatic ring of triazine and $n \rightarrow \pi^*$ charge transition at C-N terminal of triazine unit, respectively. On the other hand, a weak

absorption peak was observed at 330 nm, which could be related to the presence of charge transfer ($\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$) at the uncondensed C=O group in the triazine unit [33,36,37].

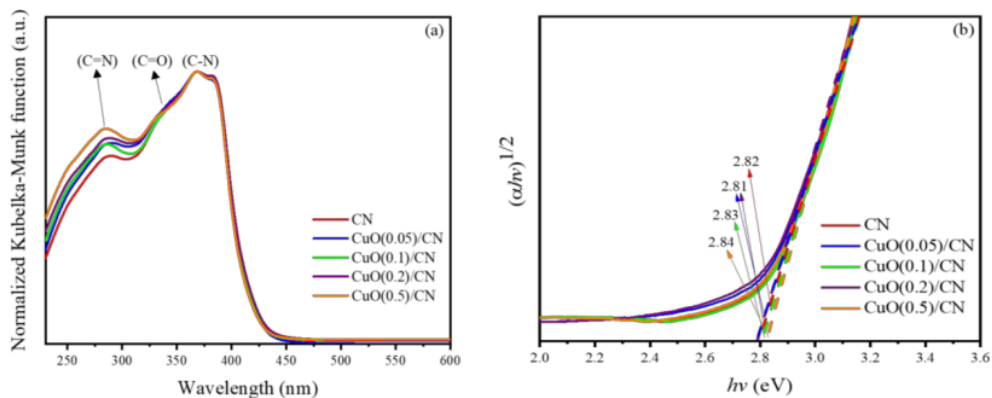


Figure 2. (a) DR UV-vis spectra and (b) Tauc plots of the unmodified CN and all CuO/CN composites.

The CuO/CN samples also showed three similar absorption peaks to those of the unmodified CN. It could be observed that the presence of copper species did not extend the absorption edge of the CN to a larger wavelength as the photodeposition method would only deposit a small amount of the copper species on the surface of the photocatalyst to support the photocatalyst activity [38]. However, the absorption peaks below 350 nm increased gradually along with the increase in the copper species loading amount. These increased absorption intensities suggested the presence of copper species on the surface of CN since the absorption peaks below 350 nm could be associated with the charge transfer transition between Cu^{2+} and O^{2-} species [36,39].

As shown in Figure 2(b), the bandgap energies (E_g) of CN and CuO/CN samples were evaluated by Tauc plot considering the samples have the indirect bandgap. The linear extrapolation from the x -intercept in the plot gave the E_g values. The CN sample gave an E_g value of 2.82 eV, while the E_g values of CuO/CN samples were found to be similar to the CN sample, which was in the range of 2.81–2.84 eV. This result again indicated that the copper species added by the photodeposition method were deposited on the surface of the CN and thus, would not alter the bandgap energy of the CN, similar to the previously reported result on TiO_2 [25].

The role of copper species as the electron trapper in suppressing the charge recombination on the CN photocatalyst was studied by fluorescence spectroscopy. As shown in Figure 3(a), the CN and CuO/CN samples exhibited three excitation peaks at 277, 331, and 369 nm, which were associated with the charge transfer at C=N, C=O, and C-N, respectively, in good agreement with their absorption peak from DR UV-vis spectra [33,36]. Meanwhile, the emission peaks from these three excitation peaks were observed only at 442 nm, which were illustrated in Figure 3(b)–(d) for each excitation wavelength of 277, 331, and 369 nm. The intensities of both the excitation and emission spectra of all CuO/CN samples were found to decrease progressively as the loading amount of copper species increased. The lower intensities of CuO/CN samples as compared to the unmodified CN denoted that there were obvious interactions between the copper species and the CN [36]. This phenomenon is well-agreed with the results from the previous literature, which reported that the copper species would act as the electron trap in the photocatalytic reaction system [17,25,40]. The copper species could give a pathway to trap the photogenerated electrons from the conduction band of CN, preventing the recombination with holes and then would lead to the improved phenol photocatalytic degradation.

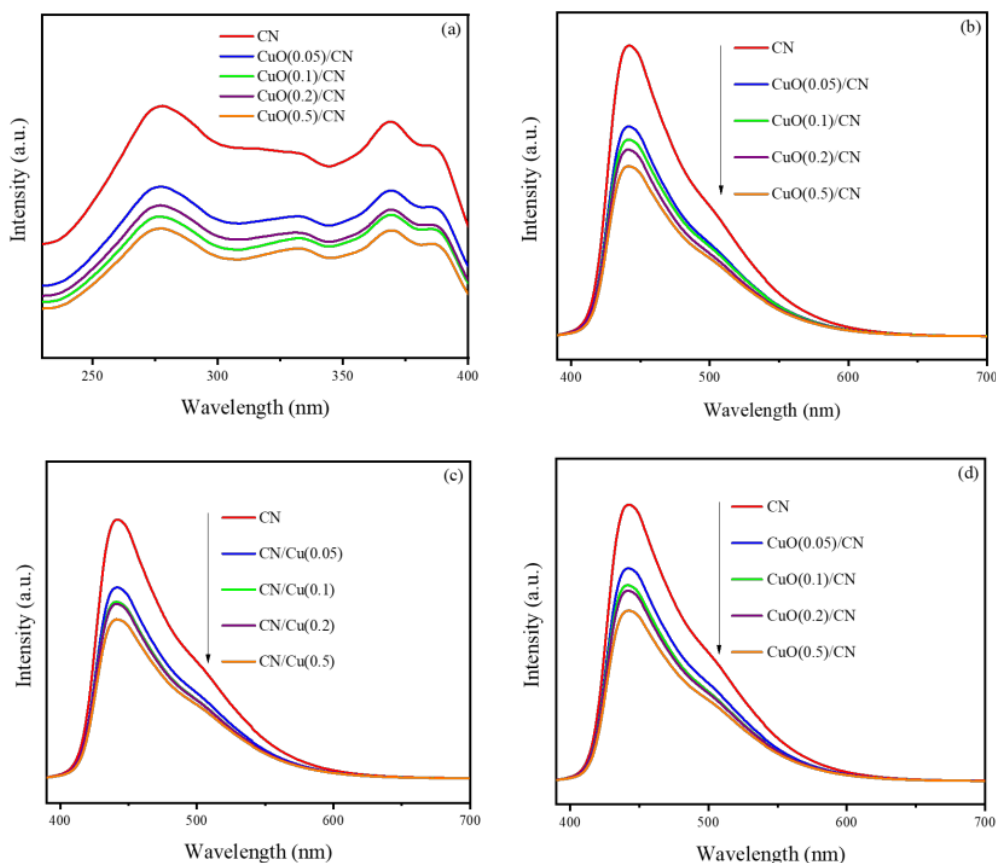


Figure 3. (a) Excitation spectra monitored at the emission wavelength of 442 nm and emission spectra monitored at the excitation wavelength of (b) 277 nm, (c) 331 nm, and (d) 369 nm for the unmodified CN and all CuO/CN composites.

Since all the emission sites of the CN were quenched after the addition of CuO, the favorable interaction site between the copper species and the CN structure was further investigated. The relative emission intensity (I_0/I) of CN and its composite was evaluated from the ratio of CN emission intensity (I_0) to the CuO/CN emission intensity (I) from three excitation wavelengths of 277, 331, and 369 nm for C=N, C=O, and C-N sites. As displayed in Figure 4, the highest I_0/I ratio was observed on CuO(0.5)/CN for all emission sites, while for each CuO/CN sample, the highest I_0/I ratio was observed at the C=N site. This result indicated that the most favorable interactions between copper species and CN occurred at the C=N site, which was consistent with the reported result from other work [36].

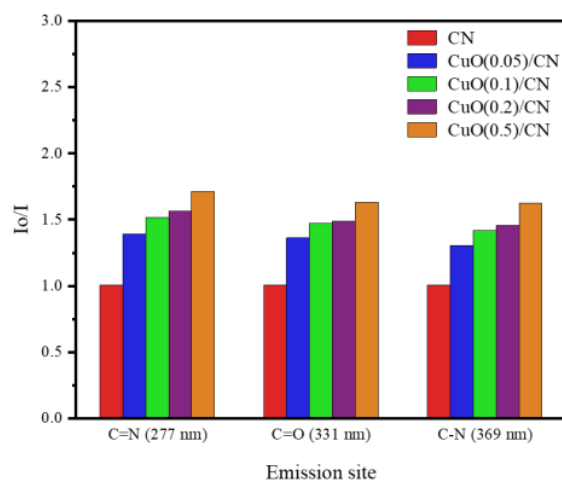


Figure 4. Relative emission intensity (I_0/I) of the unmodified CN and all CuO/CN composites.

3.2. Photocatalytic Activity of Photocatalysts

The photocatalytic activities of the CN and the CuO/CN samples were evaluated for degradation of phenol under visible light irradiation. The remaining concentration of phenol after the reaction was determined by HPLC and the results are displayed in Figure 5(a) as the percentages of phenol degradation. These results confirmed that all the CN and CuO/CN samples were active for degradation of phenol as they could utilize the visible light of more than 400 nm as the light source, which is in agreement with the medium bandgap energy of CN. The unmodified CN photocatalyst was observed to give 21.30% phenol degradation after a 24-h reaction. A similar phenol degradation (20.9%) was achieved upon the addition of 0.05 wt% copper species on the CN, suggesting that the addition of copper species in a very small amount would not give a significant effect in enhancing the photocatalytic activity of the unmodified CN. On the other hand, the addition of 0.1 wt% copper species on the CN, denoted as CuO(0.1)/CN, was found to give a remarkable increase of photocatalytic activity under visible light (41.70%), which was almost two times higher than the activity of the unmodified CN. This enhancement in photocatalytic activity would be closely related to the role of the copper species in decreasing the electrons-holes recombination as also supported by their fluorescence properties.

Further increases in the CuO loading amount did not further increase the photocatalytic activities of the CN. The CuO(0.2)/CN and the CuO(0.5)/CN gave phenol degradation percentages of 24.1% and 3%, respectively. This could be caused by the formation of CuO clusters along with the increase in the copper species amount added to the CN [17]. The CuO cluster would cover the active sites of CN and prevent the visible light to reach the active sites. This phenomenon is known as the masking effect and has been reported in previous works, either for copper species [24,25,40] and for other metal species [13]. Since the visible light has weaker penetration depth into the active site of photocatalyst as compared to the UV light source, the masking effect in visible light became more prominent which resulted in the decreased activity after the addition of copper species was more than 0.1 wt% [25]. This result confirmed that the copper species deposited to CN by the photodeposition method could enhance the photocatalytic activity of the CN when the optimum amount of the CuO was achieved, which was 0.1 wt% in this work.

To understand the active species that were responsible for the phenol photodegradation reaction, the free radicals and holes scavenging experiments were studied on the best composite *i.e.*, CuO(0.1)/CN by using ammonium oxalate, *tert*-butanol, and ascorbic acid as the hole, hydroxyl radical, and superoxide radical scavenger agents, respectively [13,30,41]. The ascorbic acid has been studied as the

superoxide radical scavenger, analogous with *p*-benzoquinone [42]. The results from the scavenging experiments are shown in Figure 5(b). After the addition of ammonium oxalate to the reaction system, the photocatalytic activity of CuO(0.1)/CN was drastically inhibited as there was no phenol degradation occurred. This result strongly suggested that the photogenerated holes were important active species for phenol degradation. Meanwhile, when *tert*-butanol was added to the reaction system, the phenol degradation was reduced from 12.22 to 1.60%, indicating that hydroxyl radicals also played a role as the important active species in the phenol degradation. A dramatic decrease in phenol degradation percentage was also observed after the addition of ascorbic acid to the photocatalytic reaction system, demonstrating that the superoxide radicals have an important role as the active species in the phenol degradation system. From these scavenging experiments, it was revealed that the photogenerated holes, hydroxyl radicals, and superoxide radicals were all the active species in the phenol photocatalytic degradation reaction, with the hydroxyl radicals gave the least inhibition effect to the phenol degradation.

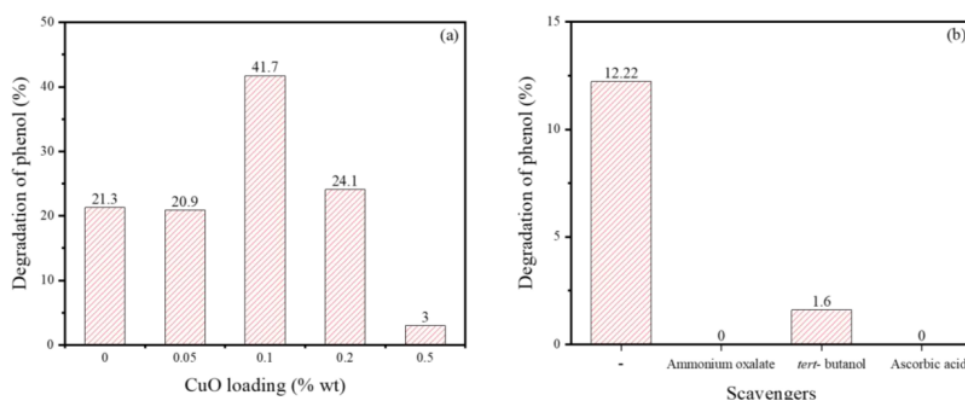


Figure 5. (a) Photocatalytic activity of the unmodified CN and all CuO/CN composites under visible light irradiation for 24 hours and (b) Photocatalytic activity of CuO (0.1)/CN in the absence and the presence of various scavengers under visible light irradiation for 24 hours.

Based on the photocatalyst characterizations and scavenging experiments, the schematic mechanism of photocatalytic phenol degradation on the CuO/CN composites is proposed and shown in Figure 6. Under visible light irradiation having larger energy than the bandgap energy of the CN, the electrons would be excited from the VB of the CN to the CB of the CN and/or trapped by the CuO. The photogenerated electrons would further reduce the dissolved oxygen to produce superoxide radicals, which would oxidize phenol to final products of carbon dioxide (CO₂) and water (H₂O). On the other hand, the photogenerated holes at the VB of the CN would also oxidize phenol. From the scavenging experiment, it could be seen clearly that hydroxyl radicals affected phenol degradation. However, the CN would not be able to directly generate hydroxyl radicals from the water due to the energy restriction and thus, the hydroxyl radicals shall be produced through another pathway, such as multiple-electron reductions of oxygen to produce hydroperoxy radicals, hydrogen peroxide, and finally the hydroxyl radicals [43]. The last process would require a good electron supplier, which could be possibly provided by the CuO.

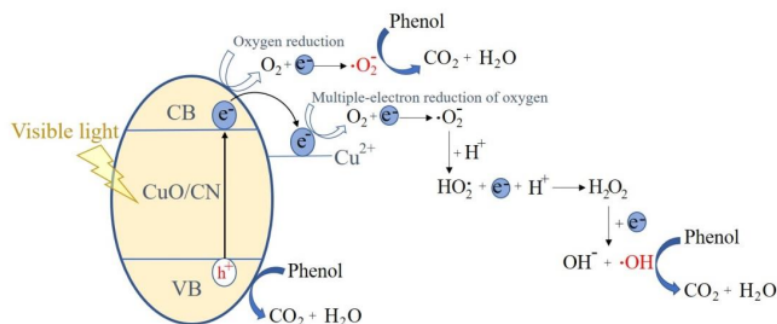


Figure 6. The proposed mechanism of photocatalytic phenol degradation on the CuO/CN composite.

The presence of CuO was indeed important to enhance the photocatalytic of the CN. Since most reports used different reaction conditions and CuO amounts, the impact of CuO shall be analyzed from the activity enhancement factor before and after its addition on the photocatalyst. As compared to the reported CuO-based photocatalysts listed in Table 1, the activity enhancement observed on the CuO/CN (1.96 times) was comparable to those of the CuO/TiO₂ (2.17 times) [26] and CuO/ZnO (2.05 times) [27], and even better than those of CuO/Ag₃AsO₄/GO (1.37 times) [28] and CuO/BiPO₄/Fe₂O₃ (1.29 times) [29]. This data again supported that the photodeposition is a good method to prepare the CuO/CN photocatalysts.

Table 1. Comparison of photocatalytic activity over CuO-based photocatalysts for phenol photodegradation.

Composite photocatalyst	Preparation method	Reaction conditions	Activity (enhancement factor)	Reference
CuO(0.1)/CN	Photo-deposition	Photocatalyst (50 mg), phenol (70 ppm), visible light (halogen, 150 W), reaction time (24 h)	41.7% (1.96)	This work
CuO(2 wt%)/TiO ₂	Sol-gel	Photocatalyst (0.5 g/L), phenol (200 ppm), UV lamp, reaction time (2 h), H ₂ O ₂ (12.5 mL)	43.21% (2.17)	[26]
CuO(7 mol%)/ZnO	Deposition by wet chemistry method	Photocatalyst (50 mg), phenol (10 ppm), UV lamp (Xe, 300 W), reaction time (3 h)	78% (2.05)	[27]
CuO(11 wt%)/Ag ₃ AsO ₄ /GO	Self-assembly	Photocatalyst (6 mg), phenol (20 ppm), visible light (300 W-halogen lamp), reaction time (20 min)	85% (1.37)	[28]
CuO(0.25)/BiPO ₄ /Fe ₂ O ₃	Impregnation	Photocatalyst (0.1 g), phenol (10 ppm), UV lamp (Xe, 150 W), reaction time (1 h)	35% (1.29)	[29]

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4. Conclusion

The CuO/CN composites were successfully prepared by depositing copper species onto the CN surface via photodeposition method using copper(II) nitrate trihydrate as the copper precursor. This method gave no significant alteration in the CN structure and bandgap energy after the addition of copper species, which were confirmed respectively from the FTIR and DR UV-vis spectra of the CuO/CN composites. The interactions between copper species and CN were revealed by the fluorescence spectra, which favorably occurred at the C=N sites. The CuO/CN composite showed the best photocatalytic activity for phenol degradation under visible light exposure when the copper species loading amount was 0.1 wt%, which gave almost two times higher activity than that of the unmodified CN photocatalyst. The improved photocatalytic activity on the CuO/CN composite was due to the presence of copper species that could suppress the electron-hole recombination of the CN. This study also demonstrated prominent contribution from the optimum amount of copper species loading, considering the importance of low masking effect to achieve high photocatalytic activity of CN under visible light illumination. The active species contributing to the phenol photodegradation reaction were photogenerated holes, hydroxyl radicals, and superoxide radicals as presented by scavenger studies.

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References

- [1] Chen C, Ma W and Zhao J 2010 *Chem. Soc. Rev.* **39** 4206
- [2] Ma H Y, Zhao L, Guo L H, Zhang H, Chen F J and Yu W C 2019 *J. Hazard. Mater.* **369** 719
- [3] Markushyna Y, Smith C A and Savateev A 2019 *European J. Org. Chem.* **2020** 1294
- [4] Zhang W, Zhang Q, Dong F and Zhao Z 2013 *Int. J. Photoenergy* **2013** 1
- [5] Yang Z, Zhang Y and Schnepf Z 2015 *J. Mater. Chem. A* **3** 14081
- [6] Yang G, Chen T, Feng B, Weng J, Duan K, Wang J and Lu X, 2019 *J. Alloys Compd.* **770** 823
- [7] Xu B, Ahmed M B, Zhou J L, Altaee A, Xu G and Wu M 2018 *Sci. Total Environ.* **633** 546
- [8] Zhao Z, Sun Y and Dong F 2015 *Nanoscale* **7** 15
- [9] Raza W and Ahmad K 2020 *Handbook of Nanomaterials and Nanocomposites for Energy Environmental Applications*, ed O Kharissova et al (Cham: Springer International Publishing) p 7
- [10] Masih D, Ma Y and Rohani S 2017 *Appl. Catal. B Environ.* **206** 556
- [11] Wang L, Wang C, Hu X, Xue H and Pang H 2016 *Chem. - An Asian J.* **11** 3305
- [12] Koltsov I, Wojnarowicz J, Nyga P, Koziorowska J S, Stelmakh S, Babyszko A, Morawski A W and Lojkowski W 2019 *Molecules* **24** 1
- [13] Yang Y, Guo Y, Liu F, Yuan X, Guo Y, Zhang S, Guo W and Huo M 2013 *Appl. Catal. B Environ.* **142–143** 828
- [14] Sridharan K, Jang E and Park T J 2013 *Appl. Catal. B Environ.* **142–143** 718
- [15] Sun J X, Yuan Y P, Qiu L G, Jiang X, Xie A J, Shen Y H and Zhu J F 2012 *Dalt. Trans.* **41** 6756
- [16] Katsumata K, Motoyoshi R, Matsushita N and Okada K 2013 *J. Hazard. Mater.* **260** 475
- [17] Janczarek M and Kowalska E 2017 *Catalysts* **7** 1
- [18] Li J, Jin X, Li R, Zhao Y, Wang X, Liu X and Jiao H 2019 *Appl. Catal. B Environ.* **240** 1
- [19] Fujimoto K, Oku T, Akiyama T and Suzuki A 2013 *J. Phys. Conf. Ser.* **433** 1
- [20] Deng X, Wang C, Shao M, Xu X and Huang J 2017 *RSC Adv.* **7** 4329
- [21] Muthukumar M, Prasath P V, Kulandaivelu R, Sagadevan S, Mohammad F and Oh W C 2020 *J. Mater. Sci. Mater. Electron.* **31** 2257
- [22] Xu J, Li S, Tan L and Kou B 2017 *Mater. Res. Bull.* **93** 352
- [23] Jiang X X, Hu X D, Tarek M, Saravanan P, Alqadhi R, Chin S Y and Khan M M R 2020 *J. CO₂*

Util. **40** 101222

- [24] Duan Y 2018 *Mater. Res. Bull.* **105** 68
- [25] Siah W R, Lintang H O, Shamsuddin M, Yoshida H and Yuliati L 2016 *Catal. Sci. Technol.* **6** 5079
- [26] Sohrabi S and Akhlaghian F 2016 *Process Saf. Environ.* **99** 120
- [27] Zhu L, Li H, Liu Z, Xia P, Xie Y and Xiong D 2018 *J. Phys. Chem. C* **122** 9531
- [28] Rakibuddin M, Mandal S and Ananthakrishnan R 2017 *New J. Chem.* **41** 1380
- [29] Chen S, Mu Z, Yan R, Zhang X, Khan I, Li Z, Sun N, Zhang Q and Jing L 2019 *J. Alloy Comp.* **800** 231
- [30] Sanni S O and Idemudia O G 2015 *J. Nanomater.* **2015** 1
- [31] Lee S C, Lintang H O and Yuliati L 2017 *Beilstein J. Nanotechnol.* **8** 915
- [32] Chowdhury P, Nag S and Ray A K 2017 *Phenolic Compounds-Natural Sources, Importance and Applications*, ed M S Hernández *et al* (London: IntechOpen) chapter 16 p 396
- [33] Lee S C, Lintang H O and Yuliati L 2014 *J. Exp. Nanosci.* **9** 78
- [34] Xia P, Antonietti M, Zhu B, Heil T, Yu J and Cao S 2019 *Adv. Funct. Mater.* **29** 1
- [35] El-Trass A, Elshamy H, El-Mehasseb I and El-Kemary M 2012 *Appl. Surf. Sci.* **258** 2997
- [36] Jasman S M, Lintang H O and Yuliati L 2017 *Macromol. Symp.* **371** 84
- [37] Cui Y, Zhang J, Zhang G, Huang J, Liu P, Antonietti M and Wang X 2011 *J. Mater. Chem.* **21** 13032
- [38] Zhang Y, Lighthart D A J M, Quek X Y, Gao L and Hensen E J M 2014 *Int. J. Hydrogen Energy.* **39** 11537
- [39] Smith E A, Ambrogi E K, Barnes E and Brame J A 2020 *Colloids Surfaces A Physicochem. Eng. Asp.* **603** 125179
- [40] Chiang K, Amal R and Tran T 2002 *Adv. Environ. Res.* **6** 471
- [41] Nandi A and Chatterjee I B 1987 *J. Biosci.* **11** 435
- [42] Wenzel U, Nickel A, Kuntz S and Daniel H 2004 *Carcinogenesis* **25** 703
- [43] Cui Y, Huang J, Fu X and Wang X 2012 *Catal. Sci. Technol.* **2** 1396

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