

# An Efficient Process for Antibiotic Wastewater Treatment

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Pollution of the water environment in today's world has become a global issue and a shortage of water resources, more and more people pay attention to pollutants, antibiotics to treat industrial wastewater embarrassed wastewater discharged into the aquatic environment and human health caused by the ecological balance would hazards and antibiotic wastewater biodegradable poor governance more difficult. This article reported the formation of the by nano-copper oxide catalyst sheet to target antibiotic wastewater reactants, catalyst dose and studied the effect of the initial concentration of the antibiotic wastewater, hydrogen peroxide dose, pH, temperature and other factors that affect the performance degradation of antibiotic wastewater.

Keywords: CuO, Catalytic oxidation, Antibiotic wastewater, Reaction mechanism.

## INTRODUCTION

Water environment antibiotics can have serious harmful effects: first, there are some toxic effects of antibiotics to aquatic organisms, inhibit or kill pathogenic microorganisms, but also may inhibit the activity of beneficial microorganisms in the environment, ecosystem disturbance and even blocking the material circulation and energy flow. Secondly, the presence of trace amounts of antibiotics in the environment for long periods can stimulate pathogenic microorganisms resistant<sup>1-3</sup>. In recent years, often reported as "uperbugs", antibiotic residues in the environment is causing pathogens produce examples of strong resistance<sup>4-6</sup>. In addition, antibiotic residues in vivo, accumulation, along with the food chain into humans, a potential human health hazards. Antibiotic wastewater can cause serious environmental impact. Therefore, study on antibiotic wastewater treatment technology so that it can discharge standards is particularly important. Common sewage treatment methods can be divided into the following categories: biological treatment, physical treatment, chemical treatment<sup>7-11</sup>.

Nano-CuO is a novel nano-materials, because of its morphology and structure of the novel features which have some special physical and chemical properties of nano-CuO whether in the field of science and technology or in the civilian industry have a high the value in use, such as in medicine sensors and catalytic materials, which have played an excellent research value and application prospect, so far people have used different methods to prepare nano-CuO various morphologies and structures<sup>12-16</sup>.

In this study, antibiotic wastewater was treated by nanocopper oxide and hydrogen peroxide. We report here the experimental oxidation results of antibiotic wastewater treated by nano-copper oxide and hydrogen peroxide. The feasibility of the technology is examined as a pre-treatment to reduce toxic organic compounds. The effects of amount of catalyst, initial concentration of the antibiotic wastewater, amount of hydrogen peroxide, pH and temperature on the removal efficiency of COD were investigated.

## EXPERIMENTAL

Antibiotic wastewater used in this study was obtained from a local antibiotic industry producing amoxicillin. The antibiotic wastewater characteristics are summarized in Table-1.

TABLE-1 ANTIBIOTIC WASTEWATER CHARACTERISTICS			
Parameter	Value		
Amoxicillin (mg/L)	140		
COD (mg/L)	800		
$BOD_5(mg/L)$	80		
pH	6.9		
TP (mg/L)	8		
Cl⁻ (mg/L)	7		
NH <sub>3</sub> -N (mg/L)	12		
Turbidity (NTU)	50		
NO <sub>3</sub> <sup>-</sup> -N (mg/L)	6		

Hydrogen peroxide (30% w/w) were purchased from Marketing, USA

**Preparation of sheet copper oxide nanoparticles:** By the help of existing literature, 0.675 g of CuCl<sub>2</sub>·2H<sub>2</sub>O was dissolved in 7.5 mL of water and configured 5 mol/L NaOH solution. 10 mL solution of CuCl<sub>2</sub> was added dropwise to a magnetically stirred solution of NaOH and stirring was continued for 5 min. The mixed solution was injected into the autoclave 35 mL and placed in an oven at a reaction temperature 180 °C for 20 h, cooled to room temperature. The reaction product was collected by centrifugation and then the autoclave were washed with deionized water and washed several times with ethanol and then placed in a vacuum oven dried at 60 °C for 24 h, to give the final product is a black copper oxide nanosheet.

### **RESULTS AND DISCUSSION**

Effect of catalyst dose on antibiotic wastewater degradation: Copper oxide nano-catalyst system is a complex system composed of gas, solid, liquid-phase composition. In present experiments the range 0-300 mg/L catalyst performance degradation effects on antibiotic wastewater was investigated. The results are presented in Fig. 1. As it is expected, rising catalyst dose increased the COD removal. At a concentration of 4000 mg/L of antibiotic wastewater, at room temperature, pH of the reaction solution is 6.9, 1.5 mg/L of hydrogen peroxide concentration conditions, the ability of different amounts of catalyst and hydrogen peroxide alone for degradation of antibiotic wastewater are increasing with the increase of time. In addition to increased significantly hydrogen peroxide than antibiotics alone wastewater COD removal by adding catalyst. The catalyst inputs to 200 mg/L COD removal until 25 min after reaching a high of 86.5 % significantly while hydrogen peroxide in a separate removal of 30.8 %, which is due to increased amount of the catalyst increase the surface area of the catalyst, can be made through the first hydrogen peroxide in contact with the catalyst increases. The second hydrogen peroxide can easily dissolved in water, increasing its solubility improved oxidation efficiency.



Fig. 1. Effect of catalyst dose on antibiotic wastewater degradation

COD removal reached about 95.6 % after 25 min at 25 °C. Therefore, catalyst dose had a significant impact on the oxidation of antibiotic wastewater.

Effect of hydrogen peroxide dose on antibiotic wastewater degradation: Experimental study of the effect of concentration with the range of hydrogen peroxide 0.5-2 mg/L studied for degradation of antibiotic wastewater. As can be seen in Fig. 2, COD removal increased when hydrogen peroxide dose increased. 95.6 % of COD was removed when hydrogen peroxide dose was 2 mg/L.



Fig. 2. Effect of hydrogen peroxide dose on antibiotic wastewater degradation

**Effect of pH on antibiotic wastewater degradation:** Experiments investigated the effects of solution pH value within the range of 5-11 on antibiotic wastewater degradation and the experimental result is shown in Fig. 3.



At a concentration of 4000 mg/L antibiotic wastewater, at room temperature, the catalyst amount is 300 mg/L and 2 mg/L hydrogen peroxide concentration, it can be seen from Fig. 4, when the initial pH value of from 5 to 11, antibiotic wastewater COD removal efficiency is greatly improved, from 19.26 to 46.5 %.



Fig. 4 shows that when the pH changes from 5 to 11, the COD removal rate of antibiotic wastewater increased from 41.62 to 95.6 %, when the pH is less than 7, the solution is acidic, then the part of the copper oxide in acidic solution dissolve into  $Cu^{2+}$  homogeneous catalytic oxidation of hydrogen peroxide, the degradation rate of antibiotic wastewater dropped significantly, when the solution pH greater than 7 is alkaline solution, CuO not only will not dissolve, because by the oxidation catalytic activity copper surface to improve the rate of decomposition of hydrogen peroxide to generate more free radicals making antibiotic wastewater COD removal rate.

**Effect of temperature on antibiotic wastewater degradation:** At a concentration of 4000 mg/L antibiotic wastewater pH of the reaction solution is 6.9, the amount of catalyst 300 mg/L and hydrogen peroxide concentration 2 mg/L, when the temperature of the solution increases, the antibiotic wastewater COD removal rate increased significantly, especially when the temperature increased from 15 to 45 °C, separate hydrogen and oxygen peroxide copper catalyzed hydrogen peroxide systems antibiotic wastewater COD removal efficiency increased from 57.26, 61.02, 67.5 and 95.6 %, COD removal efficiency increased by 10.24, 34.58 %.

Effect of temperature on the degradation of the antibiotic wastewater mainly for two points, the first temperature rises

will reduce the activation energy of the reaction, so that the reaction can not occur previously occurred in the reaction temperature is raised or the original reaction can occur now more efficient and the second is that, as the temperature increases, the solubility of the gas decreases, the affect that the dissolved oxygen in the water, thereby reducing the oxidation activity.

#### Conclusion

Antibiotic wastewater treatment by nano-CuO and  $H_2O_2$  was investigated. The results showed that copper-catalyzed hydrogen peroxide oxidation catalytic properties of the system alone significantly higher hydrogen peroxide, hydrogen peroxide and for individual nano-copper oxide catalytic system, increasing the amount of catalyst, hydrogen peroxide dosage, pH, temperature increase the catalytic activity promote the decomposition of antibiotic wastewater. Greater than 80 % COD removal from antibiotic wastewater was achieved *via* nano-CuO and H<sub>2</sub>O<sub>2</sub>.

### REFERENCES

- 1. R.P. Schwarzenbach and B.I. Escher, Science, 313, 1072 (2006).
- L. Birošová, T. Mackulak, I. Bodík, J. Ryba, J. Škubák and R. Grabic, Sci. Total Environ., 490, 440 (2014).
- R. Sidrach-Cardona, M. Hijosa-Valsero, E. Marti, J.L. Balcázar and E. Becares, *Sci. Total Environ.*, 488-489, 220 (2014).
- X. Yu, J.E. Zuo, R.X. Li, L.L. Gan, Z.X. Li and F. Zhang, *Ecotoxicol. Environ. Saf.*, **106**, 40 (2014).
- P.X. Liu, H.M. Zhang, Y.J. Feng, F.L. Yang and J.P. Zhang, *Chem. Eng. J.*, 240, 211 (2014).
- 6. E.S. Elmolla and M. Chaudhuri, J. Hazard. Mater., 192, 1418 (2011).
- 7. E.S. Elmolla and M. Chaudhuri, Desalination, 285, 14 (2012).
- S.K. Behera, H.W. Kim, J.E. Oh and H.S. Park, *Sci. Total Environ.*, 409, 4351 (2011).
- N. Dorival-García, A. Zafra-Gómez, S. Cantarero, A. Navalón and J.L. Vílchez, *Microchem. J.*, **106**, 323 (2013).
- 10. E.S. Elmolla and M. Chaudhuri, Desalination, 272, 218 (2011).
- 11. K.D. Brown, J. Kulis, B. Thomson, T.H. Chapman and D.B. Mawhinney, *Sci. Total Environ.*, **366**, 772 (2006).
- 12. A.K. Shaw and Z. Hossain, Chemosphere, 93, 906 (2013).
- 13. L. An, S.C. Liu, Z. Yang and T. Zhang, Toxicol. Lett., 213, 220 (2012).
- 14. F. Bayansal, H.A. Çetinkara, S. Kahraman, H.M. Çakmak and H.S. Güder,
- Ceram. Int., 38, 1859 (2012).
  15. O. Karvan, A. Sirkecioglu and H. Atakül, Fuel Process. Technol., 90, 1452 (2009).
- A.K. Shaw, S. Ghosh, H.M. Kalaji, K. Bosa, M. Brestic, M. Zivcak and Z. Hossain, *Environ. Exp. Bot.*, **102**, 37 (2014).