



Degradation of remazol golden yellow dye wastewater in microwave enhanced ClO_2 catalytic oxidation process

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ABSTRACT

Experiments were conducted to investigate the removal of remazol golden yellow dye in order to assess the effectiveness and feasibility of microwave enhanced chlorine dioxide (ClO_2) catalytic oxidation process. The catalyst used in this process was $\text{CuO}_n\text{-La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$. The operating parameters such as the ClO_2 dosage, catalyst dosage, and pH were evaluated. The results showed that microwave enhanced catalytic oxidation process could effectively degrade remazol golden yellow dye with low oxidant dosage in a short reaction time and extensive pH range compared to the conventional wet catalytic oxidation. Under the optimal condition (ClO_2 concentration 80 mg/L, microwave power 400 W, contacting time 1.5 min, catalyst dosage 70 g/L, and pH 7), color removal efficiency approached 94.03%, corresponding to 67.92% of total organic carbon removal efficiency. It was found that the fluorescence intensity in microwave enhanced ClO_2 catalytic oxidation system was about 500 a.u. which was verified that there was much hydroxyl radical produced. Compared with different processes, microwave enhanced ClO_2 catalytic oxidation system could significantly enhance the degradation efficiency. It provides an effective technology for dye wastewater treatment.

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1. Introduction

The color and high COD of effluents from dyehouse cause serious environmental contamination problems nowadays. In particular, azo dyes represent about half of the dyes used in the textile industry and, as a consequence, a relevant problem of pollution related to the release of these products in the environment is taking place [1,2]. Although there were several other technologies available for the removal of color and COD from azo dye wastewater such as biodegradation [3], sorption [4–6], electrochemical and oxidative degradation [7–11], chlorine dioxide (ClO_2) catalytic oxidation was a very attractive and useful technique for treatment of dyehouse effluents [12–14].

However, the traditional ClO_2 catalytic oxidation process has not been largely implemented for wastewater treatment because of the severe experimental conditions such as the limited range of pH, long reaction time resulting in high economical costs. Microwave enhanced catalytic process offers a potential solution to these problems. In recent years, some research progress in microwave chemistry has presented the feasibility to apply microwave in pollution control [15]. The use of microwave irradiation in catalytic

reactions, including decomposition of volatile organic compounds (VOC) [16] and non-biodegradable organics in wastewater [17–19], has given some remarkable results.

In this paper, $\text{CuO}_n\text{-La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$ has been used as the catalyst in the microwave enhanced ClO_2 catalytic oxidation process to treat synthetic wastewater containing remazol golden yellow RNL and the reaction conditions in this system have been optimized. Comparative studies on the different ClO_2 oxidation processes have been carried out. Then, the experiment to detect $\cdot\text{OH}$ in the microwave enhanced chlorine dioxide catalytic oxidation process and microwave enhanced chlorine dioxide oxidation process has been conducted.

2. Experimental

2.1. Materials

All the reagents used in our work were analytical-grade. The stock ClO_2 solution was prepared by mixing a vitriol solution and a sodium chlorite solution. The stock ClO_2 solution was stored in dark at 277 K and was standardized before using. The azo dye, remazol golden yellow RNL, was chosen as the target compound, and its chemical structure is shown in Fig. 1.

The catalyst was prepared by impregnation–deposition method, and the $\gamma\text{-Al}_2\text{O}_3$ ($\varphi=2\text{--}3\text{ mm}$) was used as the carrier. The

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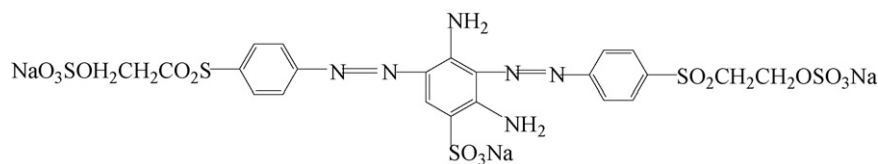


Fig. 1. Chemical structure of remazol golden yellow RNL.

$\text{CuO}_n\text{-La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$, was prepared in such a sequence that La was first loaded on the $\gamma\text{-Al}_2\text{O}_3$ by dipping 10 g of $\gamma\text{-Al}_2\text{O}_3$ in 100 mL aqueous solution containing 0.03 mol/L of La^{3+} for 12 h under room condition, and the samples were dried at 353 K for 12 h and then dried at 383 K for 2 h. The dried samples were calcined at 823 K in an oven for 4 h, so that the intermediate $\text{La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$ was obtained, and Cu was then loaded on $\text{La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$ by impregnation of $\text{La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$ with 100 mL aqueous solution containing 0.3 mol/L CuCl_2 for 24 h under room condition. After that, the Cu^{2+} was deposited with the help of precipitator [20] for 24 h, and the samples were dried at 353 K for 12 h and then dried at 383 K for 2 h. The dried samples were calcined at 723 K in an oven for 4 h to get the $\text{CuO}_n\text{-La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$ catalyst.

The formation of CuO, Cu_2O and La_2O_3 loaded on the surface of $\gamma\text{-Al}_2\text{O}_3$ is characterized by X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS). The component amounts of Cu and La in $\text{CuO}_n\text{-La}_2\text{O}_3/\gamma\text{-Al}_2\text{O}_3$ catalyst determined by using XRF are 8.12% and 1.14% respectively.

2.2. Experimental method

At the beginning of each test run, 100 mL of synthetic wastewater containing 200 mg/L remazol golden yellow dye was placed in a sealed reactor. Certain amount of the catalysts and stock ClO_2 solution were added. The reactor was shaken sufficiently to mix the stock ClO_2 solution and the wastewater, and subsequently loaded in a reconstructive commercial microwave oven which was operating at 2450 MHz with the continuous power (50–750 W). The time was recorded by the stopwatch. After been irradiated in the microwave oven for sometime, the sealed reactor was taken out and the reagent of sodium thiosulfate ($\text{Na}_2\text{S}_2\text{O}_3$) was added to dechlorinate ClO_2 residuals.

The temperature of the wastewater was measured using a thermocouple inserted into the wastewater. The pH value was determined by a pH-3C pH meter. The concentrations of ClO_2 , ClO_2^- , ClO_3^- and Cl^- in the 'stabilized chlorine dioxide' solution were measured by the method of continuous iodimetry and precipitation titration, respectively. The absorbance and total organic carbon (TOC) of the suspensions were analyzed to investigate the efficiency of the system. The visible light absorbance at 430 nm was measured using a 721-type UV-vis spectrophotometer. TOC measurement was carried out with a TOC-VCPN Shimadzu TOC analyzer.

For evaluating the catalytic activity of catalysts, both color removal and TOC removal were calculated as shown below:

$$X(\%) = \frac{C_0 V_{\text{wastewater}} - C_t (V_{\text{wastewater}} + V_{\text{ClO}_2} + V_{\text{Na}_2\text{S}_2\text{O}_3})}{C_0 V_{\text{wastewater}}} \times 100\% \quad (1)$$

where C_0 and C_t are the initial and final absorbances of remazol golden yellow dye, or the initial and final TOC, respectively; $V_{\text{wastewater}}$, V_{ClO_2} and $V_{\text{Na}_2\text{S}_2\text{O}_3}$ are the volumes of wastewater, ClO_2 , and $\text{Na}_2\text{S}_2\text{O}_3$ solutions, respectively.

It was known that hydroxyl radicals ($\cdot\text{OH}$) reacted with terephthalic acid and generated 2-hydroxyterephthalic acid which could emit fluorescence, as shown in Fig. 2. So the terephthalic acid was employed as the hydroxyl radical capturer to detect the formation of $\cdot\text{OH}$ in the microwave enhanced catalytic oxidation system. The process was conducted under the following conditions: 20 g/L of

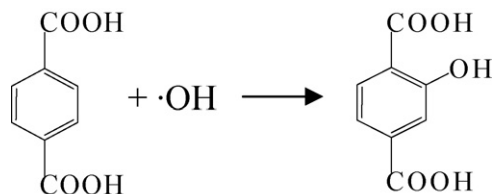


Fig. 2. Reaction between hydroxyl radical and terephthalic acid.

the catalysts and 40 mg/L of ClO_2 solution were added into 100 mL of 0.3 mmol/L terephthalic acid aqueous solution in the reactor, then irradiated the reactor in the microwave oven with the continuous power of 50 W. The time was recorded by a stopwatch. The water samples were taken from the reactor to determine the fluorescence spectrum. The fluorescence spectrum of generated 2-hydroxyterephthalic acid was measured on a Jasco FP-6500 fluorescence spectrophotometer. The slit widths were set at 5.0 nm. Sequential scans of the emission spectra were carried out between 400 nm and 600 nm at different excitation wavelengths ranging from 240 nm to 400 nm. The spectra were recorded at every 5 nm intervals.

3. Results and discussion

3.1. Effect of the ClO_2 concentration on color removal

The cost of ClO_2 was the primary factor contributing to the chemical costs of microwave enhanced ClO_2 catalytic oxidation treatment, so it was important to minimize the required amount of ClO_2 . Therefore, investigation of the initial ClO_2 concentration on the degradation of remazol golden yellow dye was conducted with varying ClO_2 concentration from 0 mg/L to 110 mg/L. The other experimental conditions were conducted as below: microwave power, 50 W; contact time, 15 min; catalyst dosage, 50 g/L. The results were presented in Fig. 3.

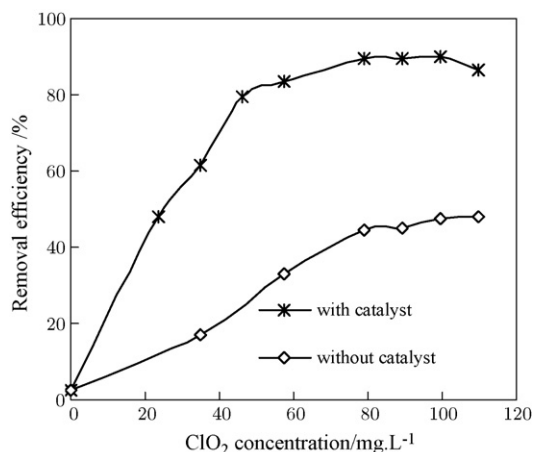


Fig. 3. Effect of ClO_2 concentration on removal efficiency ([Dye] = 200 mg/L; microwave power = 50 W; time = 15 min; catalyst dosage = 50 g/L).

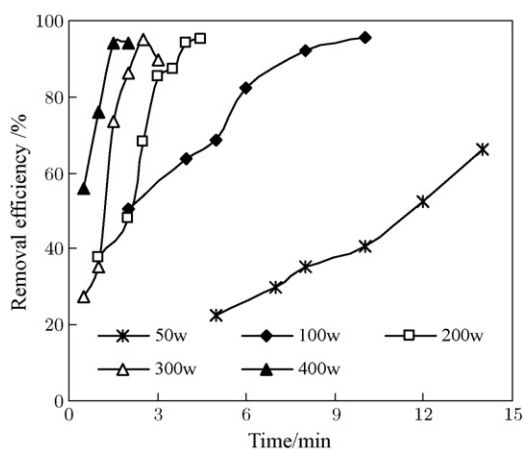


Fig. 4. Effect of microwave power and irradiation time on removal efficiency ([Dye]=200 mg/L; [ClO₂]=80 mg/L; catalyst dosage = 50 g/L; pH 5.6).

As shown in Fig. 3, the dosage of ClO₂ had an important influence on the degradation of remazol golden yellow dye. However, the color removal increased non-linearly with the increasing chlorine dioxide dosage. In the presence of ClO₂, firstly, the color removal increased with the ClO₂ concentration rapidly. This proved that the effect of increasing ClO₂ concentration was first positive to the degradation of remazol golden yellow dye. However, with the continuous increase in the initial ClO₂ concentration, the color removal increased slightly. In addition, the color removal in the catalytic oxidation system was much higher than that in the oxidation system. In the microwave enhanced catalytic oxidation process, when the concentration of the ClO₂ solution was 80 mg/L, the color removal was 89.5% while the color removal was only 44.47% without catalyst. This indicated that the catalysts of CuO_n-La₂O₃/γ-Al₂O₃ had obvious catalytic ability in microwave enhanced ClO₂ catalytic oxidation process.

3.2. Effect of microwave power and irradiation time on color removal

The degradation of remazol golden yellow RNL at different reaction times under different microwave powers was shown in Fig. 4. The other experimental parameters were conducted as below: ClO₂ concentration, 80 mg/L; catalyst dosage, 50 g/L; pH, 5.6.

In our study, it could be observed from Fig. 4 that microwave irradiation could obviously shorten the reaction time. Reaction time could be shortened from 90 min in the catalytic wet oxidation to 1.5 min and the azo dye could be effectively eliminated, and would greatly reduce the economical costs of the catalytic oxidation process.

As expected, the color removal of the remazol golden yellow RNL regularly enhanced with the microwave power increasing. Under the same power, the color removal increased obviously with the running time at the initial stage, and then the increase slowed down by prolonging the run time. This could be attributed to the relative high concentrations of remazol golden yellow RNL and oxidants resulted in rapid reaction rate in the reactor at the beginning. Based on the result, the reaction time for the microwave enhanced ClO₂ catalytic oxidation of remazol golden yellow RNL was determined to be 1.5 min under 400 W microwave irradiation.

3.3. Effect of the dosage of catalyst on color removal

The catalyst played an important role in the microwave enhanced ClO₂ catalytic oxidation process. The effect of catalyst dosage on dye removal was investigated by the addition of differ-

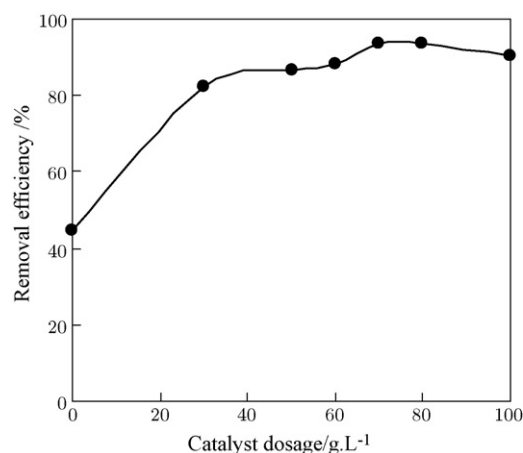


Fig. 5. Effect of the catalyst dosage on removal efficiency ([Dye]=200 mg/L; [ClO₂]=80 mg/L; microwave power = 400 W; time = 1.5 min).

ent amounts of catalyst into 100 mL of synthetic wastewater, and the results were shown in Fig. 5. The other experimental conditions were conducted as below: ClO₂ concentration, 80 mg/L; microwave power, 400 W; contact time, 1.5 min.

As the catalyst dosage increased, an increase in the color removal was observed. When the addition of catalyst reached 70 g/L, the removal efficiency could come up to over 90%. Nevertheless, the final removal did not change, showing that the excess amount of the catalyst could not improve the efficiency in the microwave enhanced ClO₂ catalyst oxidation.

3.4. Effect of pH on color removal

The initial pH is an important parameter influencing the performance of ClO₂ catalytic oxidation process [12]. To examine its effect, the initial pH of the samples was adjusted to 2–11 with the addition of HCl or NaOH. The color removal of dye solutions in various initial pH under the same reaction conditions was shown in Fig. 6. The other experimental parameters were conducted as below: ClO₂ concentration, 80 mg/L; microwave power, 400 W; contact time, 1.5 min; catalyst dosage, 70 g/L.

The results presented in Fig. 6 showed that the color removal in the microwave enhanced ClO₂ oxidation was lower than 50% and

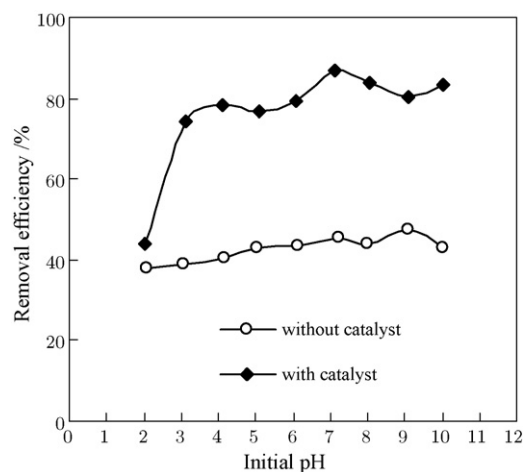


Fig. 6. Effect of the initial pH value on removal efficiency ([Dye]=200 mg/L; [ClO₂]=80 mg/L; microwave power = 400 W; time = 1.5 min; catalyst dosage = 70 g/L).

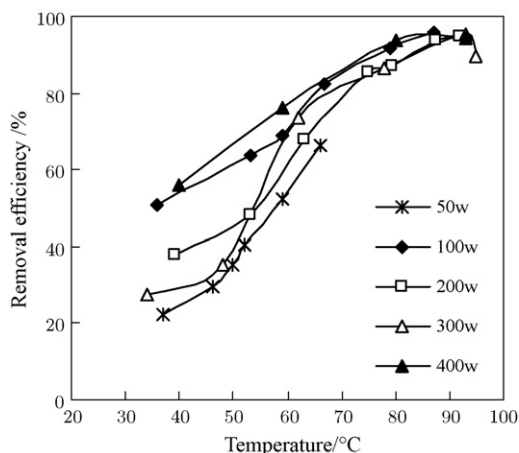


Fig. 7. Effect of wastewater temperature on removal efficiency ([Dye] = 200 mg/L; $[\text{ClO}_2]$ = 80 mg/L; catalyst dosage = 70 g/L).

changed slightly with the initial pH. But in microwave enhanced ClO_2 catalytic oxidation process, the color removal varied obviously from initial pH 2 to pH 3. It could be concluded that the optimum initial pH value in catalytic system ranges from 3 to 11. The color removal of catalytic system in this pH range was much higher than that in the oxidation system, which indicated that the catalyst associated with microwave could improve the color removal of system and adapt to extensive pH range, suggesting this process had advantages for the engineering application.

3.5. Effect of the wastewater temperature on color removal

The effect of wastewater temperature on dye removal was investigated under different microwave powers. The other experimental parameters were conducted as below: ClO_2 concentration, 80 mg/L; catalyst dosage, 70 g/L.

Fig. 7 demonstrates the effect of the wastewater temperature on the color removal. As expected, the temperature of wastewater was one of the effect factors. The color removal changed with the variation of temperatures and microwave powers. At the beginning, the removal efficiency increased obviously by increasing the temperatures. However, the final removal even decreased by increasing the system temperatures. As a result, changes in the color removal were not linearly related with temperatures in the microwave enhanced ClO_2 catalytic oxidation process. Under the same temperature, the different microwave powers got the different color removal efficiencies, which showed that the microwave in the process acted as an induced factor.

3.6. Effect of initial dye concentration on color removal

The effect of initial concentration of synthetic dye wastewater on its degradation was illustrated in Fig. 8 with varying remazol golden yellow RNL concentration from 100 mg/L to 400 mg/L. The other experimental conditions were conducted as below: ClO_2 concentration, 80 mg/L; microwave power, 400 W; contact time, 1.5 min; catalyst dosage, 70 g/L.

The results showed that the color removal of remazol golden yellow RNL decreased with increasing the initial remazol golden yellow RNL concentration. This could be attributed to the relative reduction of ClO_2 /dye molar ratio as remazol golden yellow RNL concentration increased. Thus, the effect of remazol golden yellow RNL initial concentration on its degradation was similar to that of ClO_2 dosage, which was stated above.

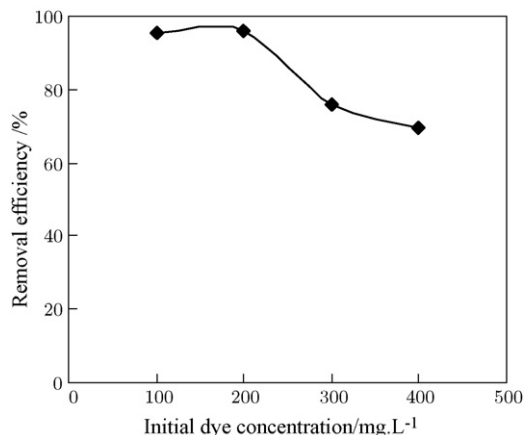


Fig. 8. Effect of initial concentration of synthetic dye wastewater on removal efficiency ($[\text{ClO}_2]$ = 80 mg/L; microwave power = 400 W; time = 1.5 min; catalyst dosage = 70 g/L).

3.7. Comparative study on the different treatment processes

In order to check the microwave enhanced effect, the color removal in different treatment processes was compared. The temperature of thermostatic system was 350 K. The other operation conditions of thermostatic system were the same as the microwave enhanced system, such as ClO_2 concentration 80 mg/L; contact time 1.5 min; catalyst dosage 70 g/L. Experimental results were shown in Fig. 9.

The reaction between the ClO_2 and remazol golden yellow RNL was an endothermic reaction, but as shown in Fig. 9, the color removal in the microwave ClO_2 oxidation was higher than traditional ClO_2 oxidation even at the same temperature. It was indicated that the microwave could improve the efficiency of traditional process not only because of its calefaction but also because of its induced function. The catalyst can also enhance the efficiency because of its function of sorption and catalysis. However, when the microwave and catalyst were used together, the removal increased obviously and the increase in the removal was more than that of summation of the respective increase, which confirmed the hypothesis mentioned above that there would be microwave induced functions.

To investigate the oxidation degree of remazol golden yellow RNL, the TOC of treated wastewater was tested. The result showed that the color removal was 94.03% while the TOC removal was only 67.92%. It suggested that the remazol golden yellow RNL in this

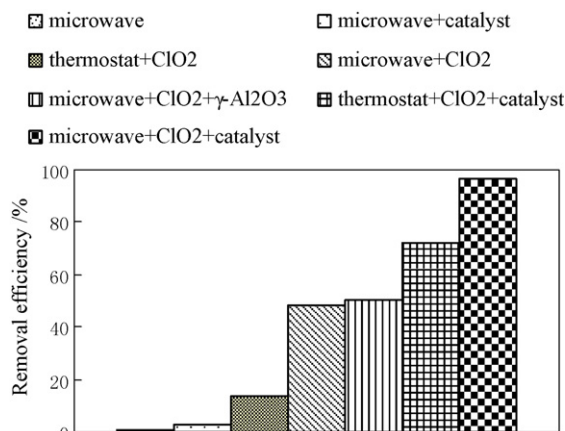


Fig. 9. Color removal efficiency in different treatment processes ([Dye] = 200 mg/L; $[\text{ClO}_2]$ = 80 mg/L; catalyst dosage = 70 g/L; time = 1.5 min; T_w = 350 K).

process was not entirely oxidized into carbon dioxide. So, much effort should be given to the detection of the new products which remazol golden yellow RNL is oxidized into and improve the TOC removal in further study.

3.8. Evaluation of $\cdot\text{OH}$ in microwave enhanced ClO_2 catalytic oxidation system

The mechanism of the ClO_2 catalytic oxidation was not very clear. It was generally reported that the oxygenous group over the catalyst can initiate the ClO_2 to be changed into radicals, such as $\cdot\text{OH}$. Microwave irradiation could be cooperative with catalyst to initiate the more formation of free radicals in the presence of ClO_2 .

To verify the above mentioned theory, the fluorescence technology was applied to detect the hydroxyl radicals ($\cdot\text{OH}$) produced in microwave enhanced ClO_2 catalytic oxidation system with the catalyst dosage of 20 g/L. (If the optimal catalyst dosage of 70 g/L was selected, the fluorescence intensity exceeded the detection range.) First, sequential scans were conducted to verify the product in the microwave enhanced ClO_2 catalytic oxidation system. The result was shown in Fig. 10.

Fig. 10 shows that the fluorescence peak of product of reaction between ClO_2 and terephthalic acid in microwave enhanced ClO_2 catalytic oxidation system was located in the vicinity of excitation/emission wavelength pair 315 nm/425 nm (± 2 nm). It was the same with the fluorescence characteristic of 2-hydroxyterephthalic acid reported in the article [21]. Therefore, perhaps, the fluorescent product formed during microwave enhanced ClO_2 catalytic oxidation was due to the reaction between $\cdot\text{OH}$ with terephthalic acid. It was indicated that the $\cdot\text{OH}$ was produced in the microwave enhanced ClO_2 catalytic oxidation system.

Influence of irradiation time on fluorescence spectrum in microwave enhanced ClO_2 catalytic oxidation process was shown in Fig. 11. It could be observed that the fluorescence intensity of microwave enhanced ClO_2 catalytic oxidation system was increasing with the time and the increase was fast at the beginning. It probably could be attributed to the fact that the relative high concentrations of terephthalic acid and ClO_2 would result in rapid reaction rate in the reactor at the initial stage. As the oxidation reactions proceeded, on the other hand, it gave rise to the appearance of 2-hydroxyterephthalic acid which would inhibit the reaction.

The fluorescence intensity of different systems was shown in Fig. 12. It was found obviously that the fluorescence intensity of

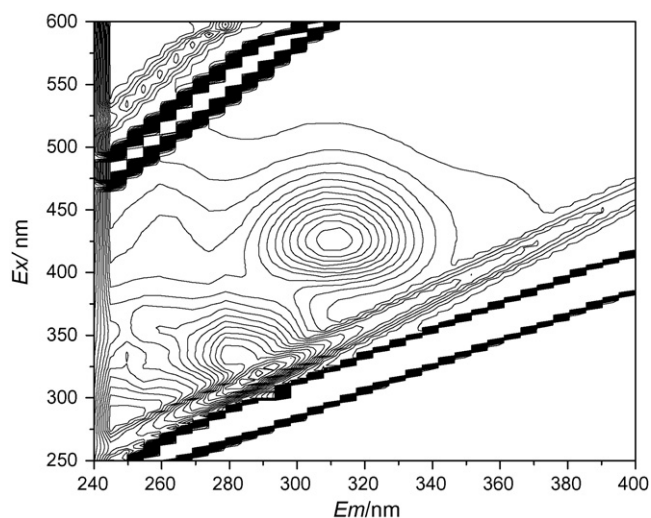


Fig. 10. Contour plot of three-dimensional fluorescence spectra of degradation product ($[\text{ClO}_2] = 40$ mg/L; [terephthalic acid] = 0.3 mmol/L; microwave power = 50 W; time = 15 min; catalyst dosage = 20 g/L).

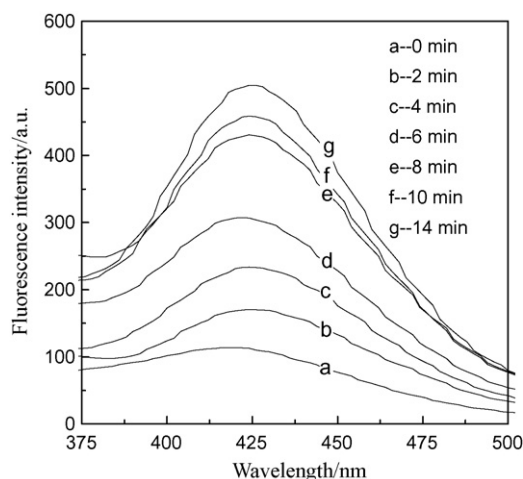


Fig. 11. Influence of irradiation time on fluorescence spectrum in microwave enhanced ClO_2 catalytic oxidation process ($[\text{ClO}_2] = 40$ mg/L; [terephthalic acid] = 0.3 mmol/L; microwave power = 50 W; catalyst dosage = 20 g/L).

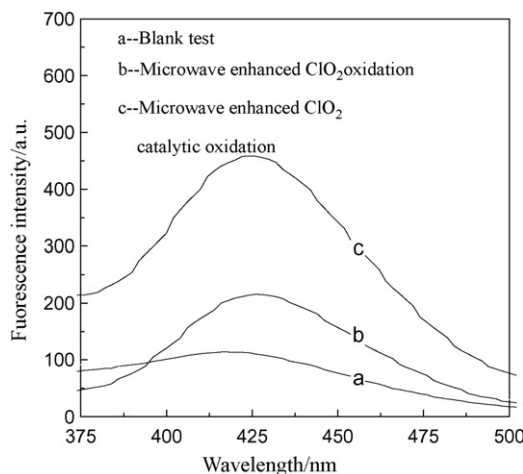


Fig. 12. Fluorescence spectrum of degradation product in different processes ($[\text{ClO}_2] = 40$ mg/L; [terephthalic acid] = 0.3 mmol/L; microwave power = 50 W; time = 10 min; catalyst dosage = 20 g/L).

catalytic oxidation system was about two times that of the oxidation system. It was indicated that the quantity of $\cdot\text{OH}$ generated in the microwave enhanced system increased by the addition of catalyst. Perhaps, in the above microwave enhanced ClO_2 catalytic oxidation process, $\cdot\text{OH}$ could be generated by a free radical chain auto-oxidation process.

4. Conclusions

The microwave enhanced chlorine dioxide catalytic oxidation process was a novel technique for the treatment of wastewater. When the remazol golden yellow RNL initial concentration was 200 mg/L, the color removal efficiency could reach 94.03%, corresponding to 67.92% of TOC. The optimum experimental conditions were as follows: volume of synthetic wastewater was 100 mL, ClO_2 concentration was 80 mg/L, contact time was 1.5 min, microwave power was 400 W and the solution pH was 7. The fluorescence technology verified that there was $\cdot\text{OH}$ produced in microwave enhanced ClO_2 catalytic oxidation system and the formation quantity of $\cdot\text{OH}$ increased with irradiation time. The formation rate of $\cdot\text{OH}$ in catalytic oxidation process was about two times of that in oxidation process.

The improvement of traditional ClO_2 oxidation process has been turned into realization by combining microwave technology and modified Al_2O_3 as catalyst. On the basis of achieving the same treatment efficiency, the improved process has more advantages such as lower oxidant consuming, wider pH range and shorter reaction time. As a developing process, the microwave enhanced ClO_2 catalytic oxidation process would provide a novel treatment method for the refractory wastewater and would have a broad prospect.

Acknowledgements

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References

- [1] C. Baiocchi, M.C. Brussino, E. Pramauro, A.B. Prevot, L. Palmisano, G. Marci, Characterization of methyl orange and its photocatalytic degradation products by HPLC/UV–VIS diode array and atmospheric pressure ionization quadrupole ion trap mass spectrometry, *Int. J. Mass Spectrom.* 214 (2002) 247–256.
- [2] C.L. Hsueh, Y.H. Huang, C.C. Wang, C.Y. Chen, Degradation of azo dyes using low iron concentration of Fenton and Fenton-like system, *Chemosphere* 58 (2005) 1409–1414.
- [3] S.Y. Kim, J.Y. An, B.W. Kim, The effects of reductant and carbon source on the microbial decolorization of azo dyes in an anaerobic sludge process, *Dyes Pigments* 76 (2008) 256–263.
- [4] A. Mittal, A. Malviya, D. Kaur, J. Mittal, L. Kurup, Studies on the adsorption kinetics and isotherms for the removal and recovery of methyl orange from wastewaters using waste materials, *J. Hazard. Mater.* 148 (2007) 229–240.
- [5] A.P.P. Cione, M.G. Neumann, F. Gessner, Time-dependent spectrophotometric study of the interaction of basic dyes with clays. III. Mixed dye aggregates on SWy-1 and laponite, *J. Colloid Interf. Sci.* 198 (1998) 106–112.
- [6] C. Namasivayam, D.J.S.E. Arasai, Removal of congo red from wastewater by adsorption onto waste red mud, *Chemosphere* 34 (1997) 401–417.
- [7] B. Mounir, M.N. Pons, O. Zahraa, A. Yaacoubi, A. Benhammou, Discoloration of a red cationic dye by supported TiO_2 photocatalysis, *J. Hazard. Mater.* 148 (2007) 513–520.
- [8] G.M. Colonna, T. Caronna, B. Marcandalli, Oxidative degradation of dyes by ultraviolet radiation in the presence of hydrogen peroxide, *Dyes Pigments* 41 (1999) 211–220.
- [9] I.A. Salem, M.S. El-Maazawi, Kinetics and mechanism of color removal of methylene blue with hydrogen peroxide catalyzed by some supported alumina surfaces, *Chemosphere* 41 (2000) 1173–1180.
- [10] N. Daneshvar, A.R. Khataee, A.R. Amani Ghadim, M.H. Rasoulifard, Decolorization of C.I. Acid Yellow 23 solution by electrocoagulation process: investigation of operational parameters and evaluation of specific electrical energy consumption (SEEC), *J. Hazard. Mater.* 148 (2007) 566–572.
- [11] N. Mohan, N. Balasubramanian, C. Ahmed Basha, Electrochemical oxidation of textile wastewater and its reuse, *J. Hazard. Mater.* 147 (2007) 644–651.
- [12] C.T. Wang, Q.H. He, P. He, Catalytic oxidation treatment of reactive brilliant red dye wastewater by chlorine dioxide, *Environ. Sci. Technol.* 29 (2006) 65–67 (in Chinese).
- [13] C.T. Wang, Q. Wang, Q.H. He, Z.F. Zhou, A study on the treatment of direct scarlet dye wastewater by chlorine dioxide catalytic oxidation on active carbon carrier, *Dyestuffs Coloration* 43 (2006) 49–55.
- [14] L.S. Shi, H.M. Zhu, Y.Y. Dong, Y.Y. Li, Study on the catalytic oxidation of simulated wastewater containing naphthol green B with chlorine dioxide as oxidant, *J. Shandong Univ. (Eng. Sci.)* 36 (2006) 91–95.
- [15] P. Wang, *Environmental Microwave Chemistry Technology*, Chem. Ind. Press, Beijing, 2003.
- [16] T. Hideaki, L.L. Ren, K. Yoshinori, Catalytic decomposition of TCE under microwave, *Catal. Commun.* 5 (2004) 317–319.
- [17] G. Hong, P. Wang, G.Y. Zhang, Microwave induced oxidation process for dye wastewater treatment with modified attapulgite as catalyst, *J. Microwaves* 21 (2005) 62–65.
- [18] G.Y. Zhang, P. Wang, S.P. Jiang, Treatment of ARGAZOL BF-3B150% by microwave induced oxidation process, *Environ. Sci.* 25 (2004) 52–55.
- [19] G.Y. Zhang, P. Wang, Y. Shi, Microwave-induced catalytic oxidation process for treatment of phenol in water with $\text{Fe}_2\text{O}_3/\text{Al}_2\text{O}_3$ catalyst, *Chin. J. Catal.* 26 (2005) 597–601.
- [20] P. Wang, X.Y. Bi, H. Jiang, Preparation method of $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ using in the microwave-induced ClO_2 catalytic oxidation process, China, CN 1907562.
- [21] T. Hirakawa, Y. Nosaka, Properties of $\cdot\text{O}_2^-$ and $\cdot\text{OH}$ formed in TiO_2 aqueous suspensions by photocatalytic reaction and the influence of H_2O_2 and some ions, *Langmuir* 18 (2002) 3247–3254.