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Decolourization and Stability of Ozone Oxidation in Municipal Wastewater Regeneration

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ABSTRACT

In order to understand the de-colourization effect and stability of ozone oxidation in municipal wastewater regeneration process, ozone oxidation experiment and related index analysis method are used. The results show that ozone oxidation treatment can effectively reduce the colour and true colour of secondary effluent. During the simulation of natural conditions, the true colour of water samples with different ozone dosage doesn't change significantly after 22 days, and the changes in surface colour and chlorophyll-*a* show good consistency. When the ozone dosage is less than 6 $mg \cdot L^{-1}$, the colour of water samples increases slowly within 12 days, then rise rapidly until the maximum is reached after 22 days. Low dose ozone treatment (less than 6 $mg \cdot L^{-1}$) can promote algae reproduction and increase the instability of water colour. However, when the dosage of ozone is more than 8 $mg \cdot L^{-1}$, it can obviously prolong the time of colour repetition of civilian water samples. The colour and turbidity of water samples increase slightly after 18 days. The changes in colour and turbidity are mainly caused by the living and reproduction of algae. High ozone dosage can maintain the stability of colour of water. Therefore, the ozone dosage of 8 $mg \cdot L^{-1}$ is recommended for municipal wastewater regeneration treatment.

INTRODUCTION

As an irreplaceable natural resource, water resources play an important role in economic development and people's lives. China is a country with a serious water shortage. The total amount of freshwater resources in China is 280 billion cubic meters, accounting for 6% of the world's water resources. It ranks fourth in the world after Brazil, Russia and Canada. However, its per capita water resources are only 2,200 cubic meters, which is less than one fourth of the world's average level. It is one of the 13 countries with the poorest per capita water resources in the world. There is a great disparity in the quantity of water resources in different parts of China. The uneven distribution of rainfall, runoff and time-space distribution of water resources during the year not only causes frequent floods or droughts in large areas, but also aggravates the imbalance between supply and demand of water resources. Besides, that is also extremely unfavourable to the development and utilization of water resources. There are more than 600 cities in China. By the end of the 20th century, more than 400 cities had water shortage problems, including 110 cities with serious water shortage and 6 billion cubic meters of total water shortage. The increasingly serious water pollution not only reduces the use function of water body, but also further aggravates the contradiction of water resources shortage. A large number of industrial and agricultural sewage discharges into natural water body, which results in huge environmental problems, has a serious impact on the sustainable development strategy being implemented in China, and also poses a serious threat to the water safety and health of urban residents.

Water is an important material basis for human survival and a non-renewable resource. In order to solve the problem of water shortage in modern cities, many countries and regions in the world have already opened up the secondary effluent of municipal sewage treatment plants as a new water source, and recycled municipal sewage as a way to alleviate the contradiction between supply and demand of water resources. The utilization of recycled water has become one of the important ways to solve the water shortage. Reclaimed water refers to the water that can be used usefully after proper treatment of sewage to reach a certain water quality index and meet a certain use requirement. Reclaimed water is a valuable water resource with large quantity, stable water quality and little influence by season and climate. It is internationally recognized as the second water source in cities.

Recycled water reuse is a new water resource utilization technology, which realizes the all-round sustainable utilization of water resources and provides a new method for alleviating the shortage of water resources. The utilization of reclaimed water can improve the comprehensive utilization rate of water resources and reduce water pollution. Rational utilization of reclaimed water is an important measure to implement the strategy of sustainable development. Recycling and recycling of sewage has considerable social, environmental and economic benefits, and has become an inevitable choice to solve the problem of water resources in the world. Therefore, it is of great strategic significance to recycle sewage, open up non-traditional water sources and realize sewage recycling to solve the water resources crisis.

EARLIER STUDIES

The development and utilization of reclaimed water has effectively alleviated the water crisis in water-scarce areas around the world, and has unique advantages in terms of economy, environmental protection and sustainable use. The United States, Japan, Israel, Australia, Singapore and other countries are in the leading position in recycled water reuse technology. The development of sewage recycling technology in these places is earlier and more mature. The technology has been widely used in all walks of life. Recycled water allocation research starts in the United States, and the initial prototype is a simple reclaimed water reuse system. Other system analysis methods are applied to establish a linear programming model for the reuse and transportation of reclaimed water. The model takes the minimum cost of water supply as the objective function, takes the balance of water distribution of multiple sources as the constraint condition, and fully considers the quantity of reclaimed water, the arbitrariness of reclaimed water reuse, seasonal differences and other factors, resulting in better allocation results. However, the linear programming mathematical model has certain limitations because it doesn't take into account the impact of treatment process, water quality and other factors on the reuse of reclaimed water (Sun et al. 2018, Haroon et al. 2018, Hailing 2017, Md Mahmudul 2018). A study establishes a linear programming model with the lowest water allocation cost of water resources from different sources as objective function and water quality and quantity as constraints. The model is flexible and can meet different water quality standards (Dong et al. 2018). In a recent research, the non-linear programming model of regional water supply is developed and applied to Texas. In this way, the regenerated water resources allocation scheme of the region is obtained (Jing et al. 2018). Incorporate sewage into the water resources allocation system and solve the water supply in different years and seasons. Environmental pollution has become a key factor to restrict the rational allocation of reclaimed water. A previous study has put the water quality requirements of reclaimed water and the discharge of water resources pollution into the allocation study and put forward a dynamic programming allocation model of reclaimed water (Wang et al. 2018, Sahadeb et al. 2019, Abija et al. 2018). A study proposes a system method of reclaimed water allocation based on health risk assessment and user acceptance (Chhipi-Shrestha et al. 2017, Chuanlei et al. 2018, Kadharsha et al. 2018). A comprehensive model of reclaimed water reuse including discharge, treatment, transportation, storage and so on is put forward by a study (Jiao et al. 2017, Ullah et al. 2018, Roslee 2018, Thiruchelvam et al. 2018). The model takes into account many factors such as supply and demand, environmental pollution, health risk, technical treatment degree and so on. A research thinks that not only the factors put forward by Oron, but also the cost of recycled water reuse should be considered. Therefore, the concept of cost-benefit should be introduced into the allocation of recycled water according to the specific situation of Israel. After the 21st century, the allocation method of renewable water resources is more mature (Gu et al. 2017, Gu et al. 2018). Based on a study, a simulation model to simulate Mexico's surface water, groundwater, reclaimed water and other water use conditions is used, and an economically viable urban water use planning is proposed (Carre et al. 2017, Nwankwoala et al. 2018, Duy et al. 2018). Besides, an economic and technological combination model to plan and dispose municipal wastewater treatment and evaluate the planning and disposal scheme based on environmental impact and economic and technological standards is used (Godenzoni & Perraton 2017, Elmnifi et al. 2018, Hanafiah 2018). In another study, set up cost function and GIS to obtain data were used to study the reclaimed water project in Tokyo area, and obtain a more economical water distribution scheme (Wang et al. 2017, Kamarubahrin et al. 2019).

MATERIALS AND METHODS

Ozone oxidation experiment: The ozone oxidation test is completed in the pilot plant. The pilot plant process mainly includes air compression and purification device, air condenser, oxygen making device, ozone generator, ozone reaction tower (stainless steel), ozone concentration detector and catalytic ozone tail gas treatment device. Air enters the discharge chamber of ozone generator through compressor and purification device to produce mixing of ozone-containing gases. The ozone mixture gas is evenly distributed into the ozone reactor through the Qin metal aeration device, and the gas-water counter current is carried out in order to fully mix the reaction. The residual ozone enters the ozone tail gas destroyer and is catalytically decomposed. Ozone concentration is detected by on-line monitor (Hare EG-00, Jitsugyo Japan). The effective volume of ozone reactor is 250 L, the contact time between ozone and water is 10 minutes, and the inflow water flow is 1.5 m^3h^{-1}. By adjusting ozone concentration and inlet gas flow rate of ozone generator, the dosage is controlled at 2, 4, 6, 8 and 10 $mg \cdot L^{-1}$.

De-colourization stability research: After ozone oxidation, 6 L water samples are taken from high temperature sterilized bottles and cultured in a constant temperature climate chamber. The culture conditions are as follows: Day length is 14 h, temperature is 25°C, with 35% RH and 20% light; night length is 10 h, temperature is 25°C, with 60% RH.

On the first day of culture, a certain volume of water is taken to determine the colour and turbidity. The water samples are filtered with 0.45 μm filter membranes to check the chroma, NH_4^+-N, soluble orthophosphate, UV_{254}, TOC and DOC. The content of chlorophyll a is determined after 6-8 hours freeze-drying.

Analysis method: Water quality parameters are determined by Nessler's reagent colorimetry; TOC and DOC are measured by Analytikjena Multi N/T 2100 TOC/TN analyser. UV_{254} is measured by UV-2401 (SHIMADZU) ultraviolet/visible spectrophotometer. Surface and true colorimetry are determined by SD-9012A colorimeter. Turbidity is determined by 2100 AN desktop turbidity analyser.

Chlorophyll a is determined by three-wavelength method. The specific method is as follows: A certain volume of algae liquid is filtered by cellulose acetate microporous membrane (pore size 0.45 um). Then the filter membrane is placed in a 10 mL centrifugal tube and 90% acetone 10 mL is added to the filter. Then a scroll mixer is used to fully oscillate the sample. The chlorophyll a is extracted in a refrigerator for 24 hours, then centrifuged in a freezing centrifuge at 4500 r/min^{-1} for 10 minutes. The supernatant is removed at a cm ratio. The absorbance at 750, 663, 645 and 630 nm is determined by spectrophotometer. The chlorophyll a content is calculated by formula 1 with 90% acetone solution as reference.

$$\text{Chl-}a = ((11.64\times D_{633}\text{-}D_{750})\text{-}2.16\times(D_{645}\text{-}D_{750})+0.1 \times(D_{633}\text{-}D_{750}))\cdot V1)/V\cdot\delta \quad \ldots(1)$$

In the formula, Chl-a is the concentration of chlorophyll a ($mg\cdot m^{-3}$), V is the volume of water sample (L), D is the absorbance. V_1 is the volume of the extract after constant volume. δ is the light path of the colorimetric dish (cm).

RESULTS AND DISCUSSION

Effect of different ozone dosage on colour and true colour removal: The colour is mainly produced by dissolved substances and insoluble suspended substances, while the true colour is only produced by dissolved substances. The variation of colour and true colour with ozone dosage is shown in Fig. 1. When the ozone dosage exceeds 4 $mg\cdot L^{-1}$, the removal rate of colour and true colour tends to be stable, which may be due to the fact that some of the chromogenic substances in water are still difficult to be removed by ozonation. When the ozone dosage is more than 6 $mg\cdot L^{-1}$, the removal rate of chroma tends to be flat. When the dosage of ozone is 10 $mg\cdot L^{-1}$, the surface colour decreases from 20 degrees to 5 degrees and the removal rate is 75%. The true colour value decreases from 13 degrees to 2 degrees, and the removal rate is 84.6%. It can be concluded that ozone has a good oxidation effect on dissolved organic matter and insoluble suspended matter in water.

Effect of different ozone dosage on removal of UV_{254} and turbidity: As shown in Fig. 2, with the increase of ozone dosage, UV_{254} decreases from 0.097 to 0.056, indicating that ozone oxidation has a certain ability to remove aromatic ring compounds. In addition, when the ozone dosage is 10

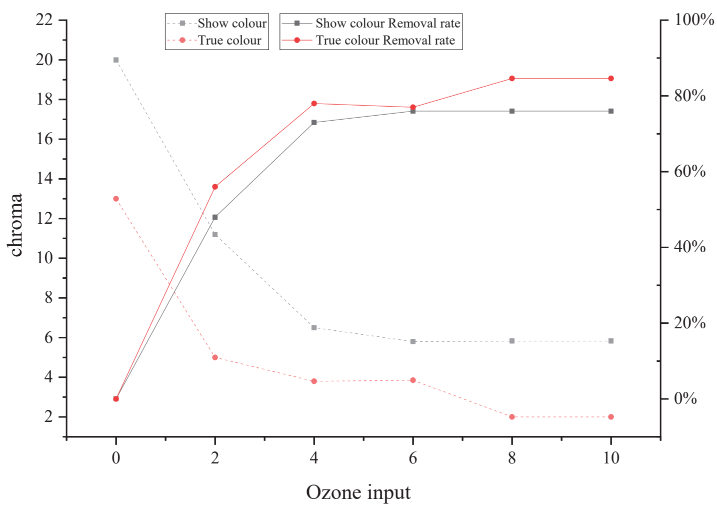

Fig. 1: Variation curves of colours and real colours with ozone input.

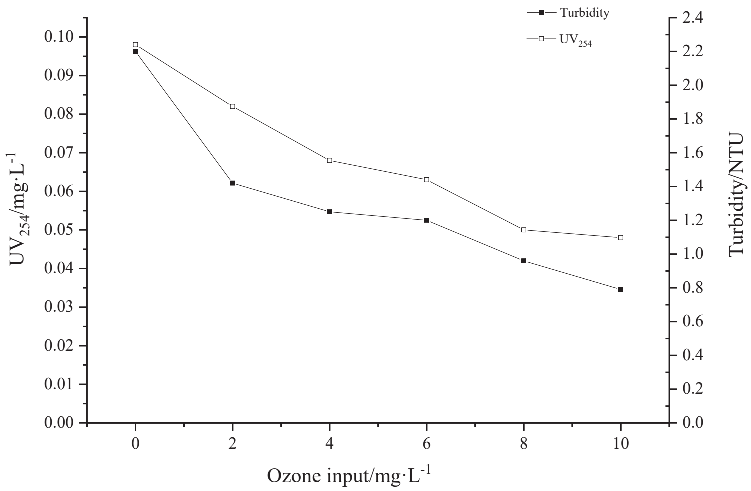

Fig. 2: Changes of UV_{254} and turbidity with ozone addition.

mg·L^{-1}, the turbidity decreases from 2.08 NTU at the beginning to 0.77 NTU at the end, and the turbidity decreases by 1.31 NTU.

Changes of algae and water quality under simulated natural conditions: From the appearance, the water samples oxidized by ozone dosage of 2, 4, 6, 8, 10 mg·L^{-1} are relatively clear at first, and there are no suspended substances. Only the raw water without ozone treatment is light green, and there is a small amount of suspended substances. When the natural conditions are simulated for 15 days, the water samples without ozone oxidation become green, the water body becomes turbid and the suspended substances increase. The dosage of 2, 4, 6 mg·L^{-1} is added again. Reclaimed water changes from colourless to light yellow, then green. The regenerated water with 4, 6 mg·L^{-1} ozone dosage change to green in a short time, while the regenerated water with 8 and 10 mg·L^{-1} ozone dosage has no obvious change in colour and turbidity. After 24 days, the regenerated water samples with dosage of 0, 2, 4, 6 mg·L^{-1} turns dark green, and with the increase of ozone dosage, the turbidity of water samples increases, especially in 4, 6 mg·L^{-1} water sample. The phenomena are closely related to the growth of algae. When algae begin to appear in the water body in the early stage, they evenly disperse in the whole water body and gradually become turbid. In the later stage, algae begin to accumulate on the surface and bottom of the water body, and the water samples with the dosage of

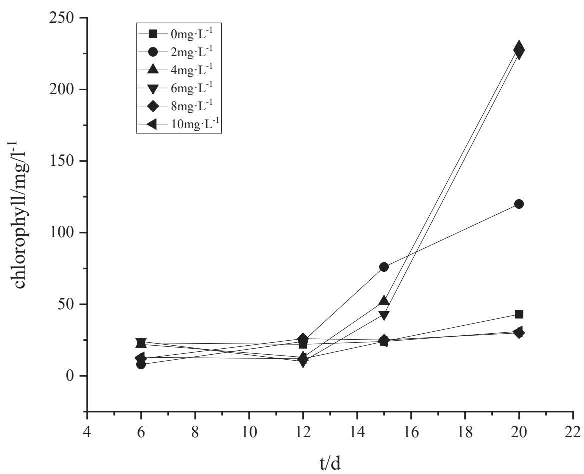

Fig. 3: Changes of chlorophyll *a* concentration with time.

8 and 10 mg·L^{-1} gradually become light green, with a small amount of suspended matter coming out.

Since all algae contain chlorophyll a, it can directly reflect the growth of algae cells as an indicator of microbial primary productivity in water. Fig. 3 shows the variation curve of chlorophyll a with time under different ozone dosage. It can be seen that the growth of algae has gone through several stages, such as slow growth, logarithmic growth and stable growth. Chlorophyll-a of water samples with ozone dosage of 2 ,4, 6 mg·L^{-1} begin to increase gradually after a delay of about 6 days in the initial growth stage of algae. After 12 days, algae begin to grow suddenly and sharply more than those of water samples without oxidation treatment. This may be due to the fact that at low concentration, ozone can only destroy a small number of algae cells and promote the release of N, P and other elements into the water, which on the contrary promotes the growth of algae. However, when the ozone dosage is 8, 10 mg·L^{-1}, the appearance of water samples remains unchanged. This is because ozone kills most of the algae and ensures the biological stability of water samples at high doses.

Fig. 4 shows that DOC increases by 5.64 mg·L^{1} from 5.03 mg·L^{-1} with ozone dosage of 2, 4, 6 mg·L^{-1}, and decreases by 5.38 mg·L^{-1} with ozone dosage of 8 and 10 mg·L^{-1}. It can be explained that ozone molecule oxidation destroys cell wall and leads to cell split and death. Cell content substances are released into water; thus, DOC rises. However, with the further increase of ozone dosage, the released cell content is mineralized into CO_2. When the natural environment is simulated after 6 days, a large number of bacteria multiplies and utilizes part of the organic matter in the water, and DOC decreases significantly. After 24 days, with the algae in the water entering the decay period, the intracellular substances begin to release, which leads to the increase of DOC. As shown in Fig. 4b, TUC in water samples doesn't change much without oxidative pre-treatment and high ozone dosage (more than 8 mg·L^{-1}), while TUC in water samples increases significantly with low ozone dosage, which is consistent with the results of chlorophyll analysis. It indicates that low dose ozone treatment can promote algae reproduction instead.

Fig. 5 shows the variation of ammonia nitrogen and soluble phosphate over time. It can be seen from Fig. 5a that, the concentration of ammonia nitrogen decreases significantly in the former 6 days, and then tends to be flat. This is due to the growth of algae and the utilization of ammonia nitrogen in water. From Fig. 5b, it can be seen that the concentration of phosphate drops rapidly in the initial 15 days. The ozone dosage of 2, 4, 6 mg·L^{-1} drops fastest, then slowly drops, and finally tends to be stable. In the first days, algae grow rapidly by using nutrients such as N and P in water, and the algae grows most vigorously in the water sample with the ozone dosage of 6 mg·L^{-1}. At the later stage, phosphate is depleted, algae also enter the decline period, and the number of algae decreases rapidly.

CONCLUSIONS

Firstly, ozone treatment can effectively remove the colour and true colour of secondary effluent. When the dosage of ozone is 6 mg·L^{-1}, the removal rate of colour and true colour exceeds 75%.

Secondly, in the process of simulating natural conditions, no significant change is found in true colour of all water samples, but the trend of change of chlorophyll a, turbidity

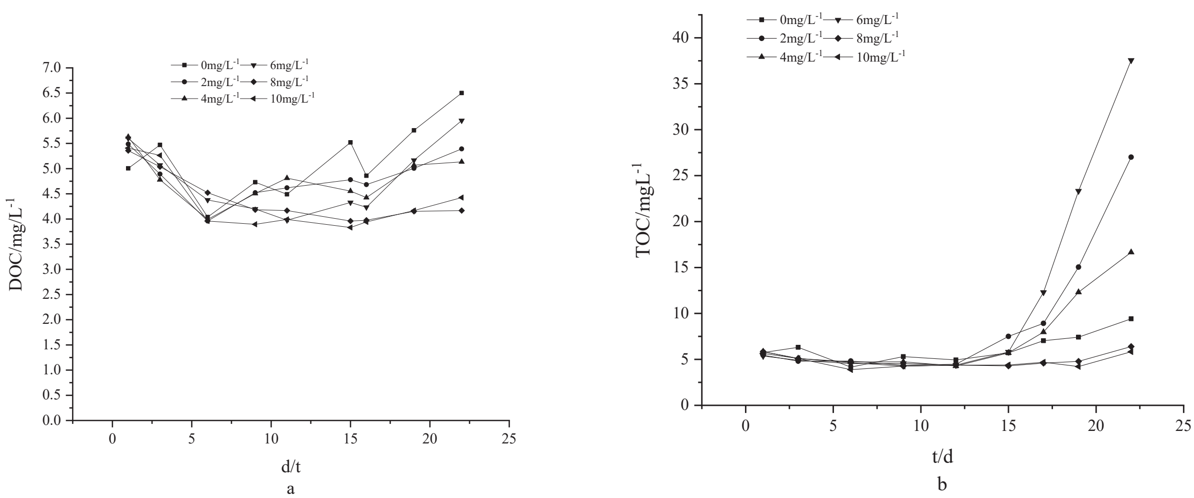

Fig. 4: Changes of DOC (a) and TOC (b) over time.

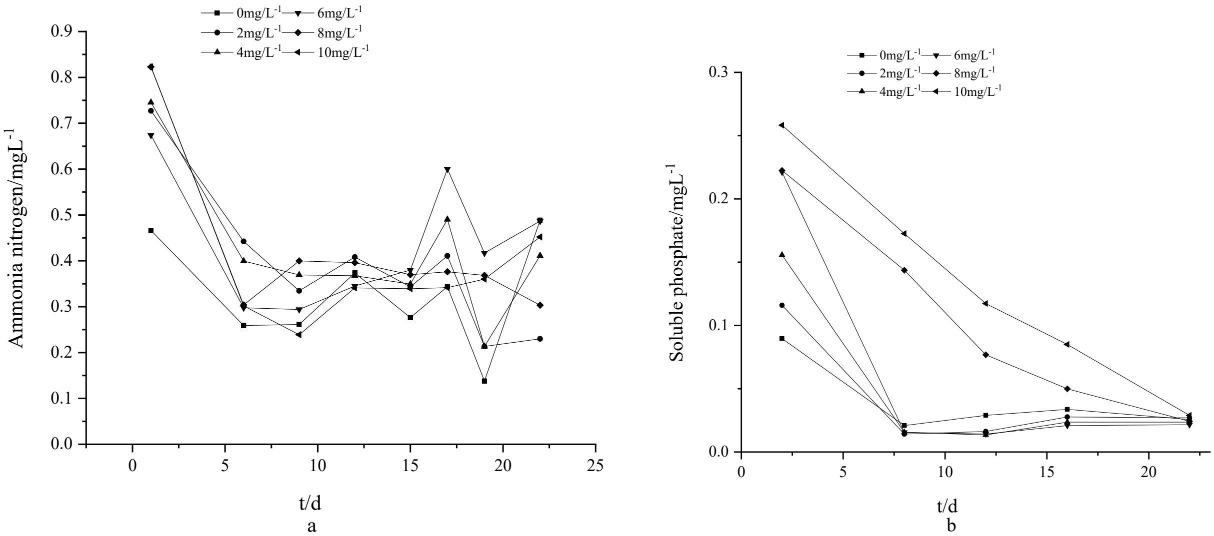

Fig. 5: Changes of ammonia nitrogen (a) and phosphate (b) concentrations with time.

and surface colour is very similar. There are two stages of slow increase and rapid increase. In the former 12 days, each index increases slowly, and then it begins to increase rapidly and reaches its maximum on the twenty-second day.

Thirdly, after ozone oxidation, the water colour is mainly due to algae reproduction. Increasing ozone dosage can prolong the water colour time and maintain the colour stability of the water. In the advanced treatment process of reclaimed water, the dosage of ozone is recommended to be 8 $mg \cdot L^{-1}$.

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