Present Situation and Research Progress of Advanced Oxidation Technology

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Abstract. With the acceleration of the industrialization process, the discharge of wastewater and the types of pollutants are constantly rising, resulting in increasingly serious water pollution problems. Advanced oxidation technology, as a new sewage treatment technology in recent years, has the advantages such as strong oxidation capacity and wide application range in degradation of organic matter in water. At present, it is widely used in domestic sewage and industrial wastewater as well as other aspects. In this paper, the principle, characteristics and application status of Fenton oxidation, photocatalytic oxidation, electrochemical oxidation, ultrasonic oxidation, wet oxidation and ozone oxidation in advanced oxidation technologies are reviewed. On this basis, the progress and prospect of advanced oxidation technologies are described.

Keywords: advanced oxidation technology; sewage treatment; organic wastewater.

1. Introduction

With the rapid increase of wastewater discharge and pollutant types, some traditional sewage treatment methods cannot meet the standards of water quality treatment [1]. At present, the main methods of sewage treatment are physical methods (filtration, sedimentation, centrifugal separation and air flotation, etc.), chemical methods (neutralization, oxidation, sedimentation, coagulation and electrolysis, etc.), biological methods (activated sludge method, biofilm method, anaerobic biological treatment) and their combined processes [2]. Advanced oxidation process in chemical method is an organic wastewater treatment technology with fast reaction speed, wide application range and simple operation. It uses strongly oxidizing free radicals to degrade organic pollutants, and has been widely studied in the degradation of organic dyes, antibiotics, pesticides and emerging organic pollutants [3-4]. Advanced oxidation technologies are mainly divided into Fenton oxidation technology [5], photocatalytic oxidation technology [6], electrochemical oxidation technology [7], ultrasonic oxidation technology [8], wet oxidation technology [9], ozone oxidation technology [10] and so on.

2. Application Status and Progress

2.1 Fenton oxidation technology

The reaction mechanism of Fenton oxidation technology is that Fe^{2+} and H2O2 combine to generate hydroxyl radical (·OH), which has strong oxidation ability. Free radicals attack organic molecules, and organic molecules are mineralized until they are transformed into inorganic substances such as CO2 and H2O [11]. Fenton oxidation technology has the advantages of strong oxidation ability, mild reaction conditions, simple operation, low operating cost, low energy consumption and convenient industrial application [12], and is widely used in pretreatment and advanced treatment of landfill leachate, printing and dyeing wastewater and pharmaceutical wastewater [13]. There are some limitations in the treatment of pollutants in practical application. The catalyst of Fenton oxidation method is difficult to be reused, the reaction pH is low, the reagent consumption is large, and a large amount of sludge is generated. Also, a large amount of Fe^{2+} contained in the effluent may easily cause secondary pollution [14].

At present, many studies show that heterogeneous catalysis - Fenton oxidation technology can be used to solve the above problems. Wei Zhiyuan et al. [15] used $CoFe_2O_4$ nanoparticles as heterogeneous catalyst to study the degradation mechanism, efficiency and influencing factors of norfloxacin (NOR)

in wastewater under the condition of adding a small amount of peroxymonosulfate (PMS). The results showed that under the optimal reaction conditions of 0.5mmol /L PMS concentration, 100 mg/L $CoFe_2O_4$ mass concentration, pH=7, and the temperature is 25 ℃, the degradation efficiency of NOR could reach 95% within 30 min, and the reaction rate constant was about 0.1054 min^{-1}. After four times of cycling experiments, the degradation rate of NOR can still reach 65%. Chen [16] et al. studied the effect of pyrite catalyzed Fenton-like oxidation on the tertiary treatment of printing and dyeing wastewater. The experimental results showed that pyrite could effectively catalyze H_2O_2 to oxidize the refractory pollutants in the biological effluent of printing and dyeing wastewater. When the initial pH is 3~6, H_2O_2 is 10 mM and pyrite is 24 g/L, the removal rate of chemical oxygen demand (COD) and color rendering rate are about 76% and 97% respectively in 180 min, while the removal rate of COD in the traditional Fenton system can only reach 65% under the optimal reaction conditions. The improvement of Fenton-like oxidation treatment effect of pyrite is mainly due to the appropriate conversion rate of H_2O_2 into hydroxyl radicals compared with traditional Fenton oxidation. Quynh et al. [17] synthesized Fe_3O_4 catalyst supported by iso-Fenton molecular sieve by impregnation-precipitation method. The catalyst used heterogeneous photoFenton process to degrade azo dye methylene blue (MB) under ultraviolet irradiation in aqueous solution. Under the conditions of initial methylene blue concentration of 50 mg/L, pH=3, catalyst dosage of 0.6 g/L, H_2O_2 concentration of 66 mg/L, and UV lamp power of 6 W, the removal rate was close to 100% within 5 min. In addition, Fe_3O_4/ Zeolite A catalyst (Fe/ZA) showed good stability in photo - Fenton degradation of methylene blue after four consecutive cycles, indicating that Fe/ZA catalyst may be a potential choice for wastewater treatment. The improved Fenton oxidation method significantly improves the treatment efficiency of high-concentration refractory organic wastewater, and makes up for the shortcomings of using the Fenton method alone, such as long treatment time, harsh reaction conditions, large number of chemical reagents and serious secondary pollution of iron mud, and has a good application prospect [18].

2.2 Photocatalytic oxidation technology

Photocatalytic oxidation technology is a chemical reaction of pollutants under the combined action of light and catalyst, which generates reactive groups to oxidize harmful compounds, mineralizes them, and decomposes harmful compounds into carbon dioxide, water and inorganic salts. The mechanism is that when the incident energy is not less than the band gap, the valence band electrons absorb light energy and jump to the conduction band. Meanwhile, holes are generated in the valence band, and photogenerated electrons migrate to the surface of semiconductor particles and are oxidized for redox reaction [19]. Photocatalytic oxidation technology has the advantages of strong oxidation ability, high selectivity, mild conditions and no secondary pollution. It is widely used in water pollution control, water decomposition, air purification, anti-bacteria, organic synthesis and other fields. After decades of development, photocatalysis has become one of the hot research fields worldwide. Its disadvantages lie in that the photocatalytic oxidation efficiency of industrial catalysts at present is not high, the utilization of solar energy is not sufficient, and there are few industrial photoreactors available at present [8]. This is because the hydroxyl radicals produced by a single photocatalytic oxidation treatment are limited, and semiconductor catalysts are often added to the reaction solution or used in combination with other methods, which produce more hydroxyl radicals under ultraviolet irradiation and have higher degradation efficiency of pollutants.

In recent years, researchers can expand the range of light energy utilization by doping metals or nonmetals and compounding with other semiconductor materials, thus improving catalytic activity. Liu Yajie [20] et al. successfully prepared magnetic graphene oxide/titanium dioxide composite photocatalyst (MgO/TiO_2) by hydrothermal synthesis method, and studied the adsorption and photocatalytic degradation of aflatoxin B1(AFB_1) by MgO/TiO_2. Adding 10 mg of catalyst to 10 mL of AFB_1 solution with a mass concentration of 2.5 μ g/mL, and irradiating it with 250 W high-pressure mercury lamp for 120 min, the photocatalytic degradation rate of AFB_1 can reach 96%. The best 0.2 M Co_3O_4/g-C_3N_4 composite photocatalyst synthesized by Suganthi et al. [21] has higher degradation

efficiency of organic pollutants methyl orange (M_O, 65%) and methylene blue (MB, 87%) in 210 min under light than that of naked g- C_3N_4. After five consecutive cycles, Co_3O_4/g-C_3N_4 composite still had good photocatalytic recycling stability. Wang Xun et al. [22] used titanium tetraisopropoxide and glucose as raw materials to prepare a brand-new CQDs/TiO_2 composite material and used it to degrade tetracycline wastewater. The results showed that the removal rate of tetracycline by CQDs/TiO2 could reach 93% with the condition of pH=5.6, catalyst dosage 1 g/L, tetracycline solution concentration 10 mg/L and illumination for 120 min, and the degradation efficiency of the composite material was greatly improved. Zhang Qiuyun et al. [23] synthesized TiO_2 @ UIO-66 photocatalyst by one-pot hydrothermal method. Under the irradiation of visible light, 0.05g of TiO_2 @ UIO-66 material was put into 40 mg/L rhodamine B solution, and its degradation rate was 87% after 120 min of photocatalysis. The photocatalytic oxidation technology of composite materials is much more effective than the single photocatalytic oxidation technology, and its treatment efficiency is higher.

2.3 Electrochemical oxidation technology

Electrochemical oxidation technology can be divided into direct oxidation and indirect oxidation. In the process of direct electrochemical oxidation discharge, pollutants diffuse to the anode surface, where they act as electron acceptors for direct electron transfer, which leads to the formation or complete mineralization of small organic molecules. Indirect electrochemical oxidation mainly relies on generating strong oxidants on the electrode surface and then transferring them to the bulk solution to attack and degrade organic pollutants [24]. The advantage of this technology is that it does not need to add additional chemicals, only consumes electric energy, and produces strong oxidation active substances [25]. In recent years, electrochemical oxidation has been widely used in the field of non-biodegradable pollutants such as landfill leachate, printing and dyeing wastewater and refinery wastewater. However, electrochemical oxidation method has some shortcomings, such as low energy conversion efficiency, high energy consumption, poor chemical stability and short electrode life, which limit the wide popularization of electrochemical oxidation technology in refractory organic wastewater [26]. Only relying on one single technology to treat specific wastewater has limitations, and it can be combined with other traditional wastewater treatment methods such as adsorption, biological treatment and membrane filtration as the pretreatment or advanced treatment stage. Therefore, the combination of various water treatment technologies is an important direction for future development. This method can improve sewage treatment efficiency and reduce energy consumption by taking advantage of the strengths of different technologies [27].

Wang Yu et al. [28] used macroporous resin adsorption and electrochemical oxidation to treat dye wastewater. The chemical oxygen demand concentration ρ (COD) of the wastewater before treatment was about 22 g/L, and the removal rate of ρ (effluent COD) after treatment by resin adsorption technology was 3.3 ~ 3.6 g/L, reaching 84%. Then, the organic matter, which is difficult to be adsorbed by resin, was further treated by electrochemical oxidation with titanium-based lead dioxide as electrode, and finally ρ (effluent COD) was less than 0.1 g/L, and the total removal rate of dye wastewater can reach 99%. He et al. [29] used tower bipolar electrode flocculation-oxidation reactor (BEF-O) and electrochemical biofilm reactor (EBR) to effectively treat mature landfill leachate, and studied the removal efficiency and mechanism of various pollutants by BEF-O reactor. The combined device can reduce the concentration of COD from 6410 mg/L to 338 mg/L and the concentration of NH4+-N from 4065 mg/L to 4 mg/L, and the economic energy consumption was 32.02 kWh / (kg COD) and 54.04 kWh / (kg NH4+-N) respectively. Popat et al. [30] degraded mixed industrial wastewater by combining electrochemical advanced oxidation process (EAOPs) with biological treatment. Before biological treatment, Electro-Fenton process and persulfate were used to improve the biodegradability of wastewater. The experimental optimization conditions are pH=3, with effective area of 25 cm^2, electrode spacing of 1 cm, and voltage of 10 V, dosage of persulfate 200 mg/L and dosage of catalyst 10 mg/L. After electrolysis for 1 hour, the COD removal rate can reach 60%. Combined with EAOPs and biological treatment, the overall COD removal rate was as

high as 94%. Therefore, the combined process is an effective treatment scheme for the degradation of organic pollutants.

2.4 Ultrasonic oxidation technology

Ultrasonic oxidation method uses a certain range of ultrasonic radiation to generate ultrasonic cavitation in the solution, and generates strong oxidative free radicals through physical and chemical processes, and the free radicals react with organic pollutants to realize the degradation of organic pollutants in the solution [31]. Ultrasonic oxidation has the advantages of strong oxidation ability, simple operation of oxidation equipment, fast oxidation speed and no secondary pollution. It is widely used in the purification of high-salinity wastewater, decolorization of dye wastewater, regeneration of activated carbon, sludge dehydration and so on. Ultrasonic oxidation technology alone has high treatment cost, high energy consumption, poor treatment effect on hydrophilic and nonvolatile organic matter, and low efficiency in degrading pollutants. A lot of literature analysis shows that the combination of ultrasonic oxidation and other advanced oxidation technologies has more obvious advantages and is the key development direction in the future [32].

Xiong Ying et al. [33] studied the effects of ozone oxidation (O_3), ultrasonic oxidation (US) and ozone and ultrasonic combined oxidation (O_3+US) on reducing the COD of shale gas fracturing flowback fluid. The results showed that O_3+US had a better effect on reducing the COD because it could produce more free radicals. The mass concentration of ozone was 42 mg/L, the pH was about 2.5, the ultrasonic power was 800 W, the addition of catalyst MnO2 was 0.45 g/L, and the reaction time was 100 min. After treatment, the COD was reduced by about 68%. Ran et al. [34] used the combined action of ultrasonic dissolution and ozone oxidation to remove the organic matter in the waste liquid, and determined that the optimum technological conditions of the combined treatment system of ultrasonic dissolution and ozone were reaction time of 7h, ozone concentration of 7.65 g/h and ultrasonic power of 600 W. The removal rate of total organic carbon (TOC) was 60%, and the decolorization rate was 87%. Amarzadeh et al. [35] used ultrasonic oxidation to assist electro-Fenton oxidation to remove penicillin G and ciprofloxacin from aqueous solution. The results showed that the removal rates of penicillin G and ciprofloxacin by ultrasonic-electro-Fenton process are about 96% and 98% respectively. Under the optimized operation conditions, the removal rates of COD and TOC by ciprofloxacin were 73% and 62% respectively, and those by penicillin G were 61% and 72% respectively. Therefore, in order to improve the application effect and expand the application scope of ultrasonic oxidation technology, the future research of ultrasonic oxidation technology must focus on the joint application with advanced oxidation technology.

2.5 Wet oxidation technology

Wet oxidation method refers to a treatment method that uses oxygen or air to oxidize organic matter and other reducing substances in water under high temperature and pressure to generate CO_2 and H_2O. This method can unselectively and efficiently oxidize high-concentration organic wastewater, and the reaction time is short. At present, it is applied to the degradation of pollutants in industrial wastewater, sludge disposal, recycling of papermaking waste liquid and so on. At present, the popularization of wet oxidation method is limited, and it is difficult to achieve good treatment effect because of the harsh reaction conditions. Organic acids will be produced during the whole treatment process, so the selected equipment should be resistant to high temperature and high pressure and have strong corrosion resistance [37]. In the process of sewage treatment, the equipment cost is high, which can only be used in small-scale wastewater treatment. Wet oxidation method will also produce toxic substances when used, so it is rarely used in industrial production. Wet oxidation technology has strict requirements on reaction conditions. Strengthening wet oxidation technology by combining it with other processes can reduce the requirements on reaction conditions and improve the reaction efficiency to a certain extent.

Lu Xiaoyang et al. [38] used two-stage process of wet oxidation (WAO) - catalytic wet oxidation (CWAO) to treat photographic fixing wastewater. The optimum reaction conditions of WAO were

temperature 160℃, oxygen partial pressure 1 MPa, reaction time of 2h and influent pH=4.8. Under these conditions, the COD_{Cr} removal rate reached 79% and effluent pH=1.4. When CWAO treated WAO effluent, the selected reaction conditions were: pH=12.9, temperature 250℃, oxygen partial pressure 3 MPa, and reaction time of 2h. When CWAO and WAO were combined to treat photographic fixing wastewater, the removal rate of COD_{Cr} was as high as 99%, and the removal rate of ammonia nitrogen reached 97%, with pH = 5.6. Pariente et al. [39] studied the coupling treatment of non-biodegradable agrochemical wastewater by heterogeneous catalytic wet hydrogen peroxide oxidation (CWHPO) and rotating biological contactor (RBCs). Under the best operating conditions, the effluent of CWHPO was co-treated by RBCs with simulated municipal sewage with different concentrations of CWHPO. Red blood cells showed high stability when treating CWHPO wastewater, and TOC and total nitrogen (TN) decreased by about 78% and 50% respectively. Gout et al. [40] studied the coupling of membrane process and wet air oxidation to treat industrial membrane concentrate of organic pollutants in wastewater, and achieved the best removal rate at 300℃. The removal rate of total organic carbon was between 75% and 98%, the removal rate of chemical oxygen demand was between 82% and 99%, and the fluorescence footprint was completely removed.

2.6 Ozone oxidation technology

The principle of ozone oxidation method is to destroy the ozone double bond through the action of ozone, and decompose organic matter into smaller molecules by stages. This method can be divided into direct reaction and indirect reaction [41]. Direct reaction is the reaction between ozone and organic matter, which has strong selectivity and slow reaction speed. Indirect reaction refers to the decomposition of ozone in water to generate hydroxyl radicals, which participate in the oxidation reaction. This method has no selectivity, fast reaction speed and complete degradation of organic matter [42]. Ozone oxidation method has strong oxidation ability and can be decomposed quickly in water. It is generally used in municipal water supply, municipal sewage, industrial wastewater, flue gas denitrification, fine chemicals, swimming pool disinfection, space disinfection, beverages and foods, etc., and has a good effect on wastewater decolorization, deodorization, sterilization and degradation of pollutants [43]. Because of the shortcomings of single ozone oxidation method in wastewater treatment, such as low ozone solubility, high power consumption when ozone is generated, unstable reaction, corrosion of equipment and pipes, and high operating cost, ozone oxidation method is often combined with other methods to improve the degradation rate of pollutants in actual wastewater treatment [44]. For example, O_3/UV, O_3/H_2O_2, O_3/Fe (II), O_3/metal oxide catalyst, O_3/activated carbon, O_3/ultrasound, O_3/Fenton, photocatalytic ozone oxidation and other combined processes [45].

Meng Ning et al. [46] used O_3, hydrogen peroxide/ozone oxidation (H_2O_2/O_3) combined process to treat the oil production wastewater in an oil field in order to reduce the COD in the effluent and improve the biodegradability of the oil production wastewater. The results showed that when O_3 was used alone to treat the oil production wastewater in an oil field, the COD removal rate was 28% and the B/C rose from 0.08 to 0.248, under the conditions that O_3 was 20 mg/L, the reaction time was 60 min and the wastewater pH was 8.5. The treatment effect of H_2O_2/ O_3 combined process is more remarkable. Under the optimum conditions of O_3 30 mg/L, reaction time of 60 min, H_2O_2 dosage 0.24 g/L and wastewater pH=8.5, the COD removal rate reaches 55% and the B/C is increased to 0.44. Wang et al. [47] designed and applied the UV/O_3 pressurization process to treat the actual high-salinity textile wastewater. Under the conditions of O_3 dosage of 200 g/t and pressure of 0.2 MPa, the decolorization rate of UV/O_3 pressurization process was up to 85% and the COD_{Cr} removal rate was up to 43%. Compared with ordinary ozone oxidation, UV/O_3 pressurization process improved the solubility of O_3 in wastewater and gas-liquid mass transfer. The research of Asgari et al. [48] mainly focused on the removal of ceftazidime (CFT) from aqueous solution by photocatalytic ozonation with T_iO_2 magnetic nanoparticles (T-MNPs). Under the optimum conditions (CFT concentration of 10 mg/L, pH=11, catalyst dosage of 1 g/L and ozone flow rate of 0.22 g/h), and after 15min of treatment, the removal rate of CFT was very high, the mineralization rate was 75%, and the reusability of the

photocatalyst was evaluated, showing that the removal performance decreased by 5% after 6 consecutive cycles.

3. Conclusion

Advanced oxidation technology, as a new sewage treatment technology, has the advantages of strong oxidation ability, fast reaction speed, wide application range and high efficiency in degrading organic pollutants, and has good treatment effect in organic wastewater. However, due to its rich and diverse technical types and different application conditions, the ideal sewage treatment effect cannot be achieved by using one technology alone. In recent years, the main application direction of advanced oxidation technology is technology combination, such as using electrochemical oxidation-biological treatment to degrade mixed industrial wastewater, which not only reduces the COD of wastewater, but also effectively improved the biodegradability of wastewater. Whether advanced oxidation technology is combined with traditional technology or advanced oxidation technology, it is necessary to ensure that the functions of combined technology are not coincident and their advantages are complementary, so as to improve the oxidation capacity and the efficiency of degrading organic pollutants in wastewater.

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