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REVIEW ARTICLE

Application of electrode materials and catalysts in electrocatalytic treatment of dye wastewater

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Abstract The dye industry produces a large amount of hazardous wastewater every day worldwide, which brings potential threaten to the global environment. As an excellent method for removal of water chroma and chemical oxygen demand, electrocatalytic methods are currently widely used in the treatment of dye wastewater. The selection and preparation of electrode materials and electrocatalysts play an important role on the electrocatalytic treatment. The aim of this paper is to introduce the most excellent high-efficiency electrode materials and electrocatalysts in the field of dye wastewater treatment. Many electrode materials such as metal electrode materials, boron-doped diamond anode materials and three-dimensional electrode are introduced in detail. Besides, the mechanism of electrocatalytic oxidation is summarized. The composite treatment of active electrode and electrocatalyst are extensively examined. Finally, the progress of photo-assisted electrocatalytic methods of dye wastewater and the catalysts are described.

Keywords electrocatalytic oxidation, electrode, electrocatalysis, dye wastewater

1 Introduction

With the continuous progress of society and economy, the people's requirements for aesthetics are gradually increasing. Therefore, the dye industry is developing rapidly, and more and more dye wastewater is discharged into the environment. According to related reports, there are more than 100000 dyes used on the market [1–4]. Up to date, industries such as textiles, cosmetics, papermaking, leather, food and pharmaceuticals produce a large amount of high-concentration dye wastewater, which contains many kinds of organic contaminants that are threaten to environment and human health [2]. The high chromaticity and alkalinity of dye wastewater make deep treatment of dye wastewater pretty hard. Azo dyes are the most common and widely used dyes, accounting for more than 70% of their total output [5–7]. They have good structural stability and are extremely difficult to degrade [8]. Dye wastewater is hard to trade and widely exists in developing countries. For this reason, it is pretty necessary to find an efficient and low energy processing method.

Traditional wastewater treatment methods include physical methods, chemical methods, and biodegradation methods [9–11]. However, these methods are not favorable for dye wastewater treatment [12]. In the past decades, advanced oxidation methods including photochemical oxidation, catalytic wet oxidation, sonochemical oxidation, ozone oxidation, and electrocatalytic oxidation have been valued by researchers [13–21]. Among them, electrocatalytic oxidation is widely applied in the treatment and disposal of dye wastewater because it can decompose large organic molecules into small environment-friendly molecules. In addition, it also has the advantages of strong oxidation ability, simple operation, mild treatment conditions, and easy automation. It is expected to further become an efficient technology for the treatment of difficult-to-biodegradable and high-concentration organic dye wastewater [22–25].

Electrode materials and electrocatalysts are the two factors influencing the removal efficiency of electrocatalytic oxidation. At present, metal oxide anodes and synthetic boron-doped diamond (BDD) electrodes are two common high-efficiency electrode materials in the field of dye wastewater treatmentwith the advantages of high oxygen evolution potential and good stability [26–30]. While due to the limits of service life and cost, researchers have tried many methods to modify them to

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improve their performance. For dimensionally stable anodes (DSA) electrodes, modification methods mainly include doping ions, nanoparticles, introducing intermediate layers, and adjusting the microscopic morphology of the electrodes. For BDD electrodes, the main modification method is to deposite substrate on which the thin film such as Ti, Si, Nb, and Ta [31,32].

To further improve the efficiency of electrocatalytic treatment of wastewater, three-dimensional electrode have begun to be widely concerned by researchers and become a popular topic [33–38]. The three-dimensional electrode refers to the introduction of particle electrodes on two-dimensional electrodes. In the three-dimensional electrode system, the particle electrode will be polarized under the action of an electric field to form a miniature electrolytic cell. In this way, it can increase the reaction area of the cathode and anode and shorten the mass transfer distance, which can greatly increases the reaction area [39,40]. At present, the main categories of three-dimensional electrodes include carbon materials, metal oxides, and minerals. The three-dimensional electrode catalyst mainly refers to supporting some catalytically active substances on the three-dimensional electrode to further improve the reaction efficiency of electrocatalytic oxidation [41,42].

The specific process of degrading organic matter such as dyes using electrocatalytic oxidation is very complicated. Up to now, many researchers believe that hydroxyl radicals (·OH) with strong oxidation ability generated on the surface of the electrode, which decomposes refractory organic contaminants into carbon dioxide and water. According to the different ways of electron transfer in the system, electrocatalytic oxidation is generally categorized into direct oxidation and indirect oxidation. Direct oxidation means that the organic matters directly gain or lose electrons on the electrode surface and are reduced or oxidized. Indirect oxidation refers to the generation of strong oxidant matters on the electrode surface, such as ·OH, dissolved Cl_2, HClO or ClO^-, etc., which will decompose organic pollutants into carbon dioxide or other simple compounds [43–46]. In the process of treating dye wastewater, both processes exist theoretically.

Comninellis et al. deeply studied the oxidation-reduction mechanism of organic matter on the electrode surface and came up with a model (shown in Fig. 1). The reaction process is as follows [47–50]. In the first step, the water discharges at the anode, and the formed hydroxyl radicals adhereto the electrode surface (1). Secondly, the oxygen from adsorbed hydroxyl radicals at the electrode surface convert to the lattice (2). Therefore, there are two states of active oxygen in the solution. The one is the oxygen in the hydroxyl radicals adsorbed on the electrode surface, and the other is the oxygen in the metal oxide lattice. When organic matter exists, two kinds of active oxygen react with it to degrade the organic matter into carbon dioxide and water or some other simple small molecules (3,4). If there is no organic matter in the solution, the two active oxygen species will undergo an oxygen evolution reaction (5,6).

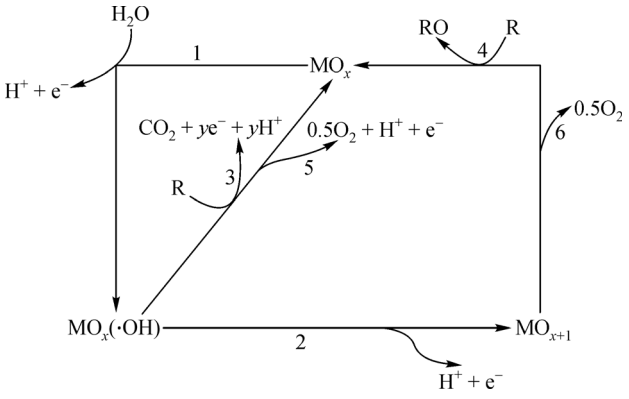

Fig. 1 Schematic diagram of electrocatalysis principle [45–48].

$$MO_x + H_2O \rightarrow MO_x[OH] + H^+ + e^- \quad (1)$$

$$MO_x[\cdot OH] \rightarrow MO_{x+1} + H^+ + e^- \quad (2)$$

$$R + MO_x[\cdot OH] \rightarrow CO_2 + MO_x + yH^+ + ye^- \quad (3)$$

$$R + MO_{x+1} \rightarrow RO + MO_x \quad (4)$$

$$MO_x[\cdot OH] \rightarrow 0.5O_2 + MO_x + H^+ + e^- \quad (5)$$

$$MO_{x+1} \rightarrow 0.5O_2 + MO_x \quad (6)$$

Most of the dye molecules are aromatic compounds, and their structure is generally complex with many functional groups. Taking auramine-O dye as an example, the hydroxyl radicals and metal cations in the system will first attack the –N = N– bond and –C = N– bonds to cause them to crack. Next, the oxygen-containing functional group is attacked and oxidized. After that, the most stable benzene ring is attacked to obtain the final oxidation product, CO_2 and H_2O [51].

The novelty of this paper focuses on the electrocatalytic treatment of dye wastewater, summarizing several of the most widely used electrode materials and the performance of three-dimensional electrode catalysts in the field of dye wastewater treatment. In addition, the synergistic treatment of multiple advanced oxidation methods is extensively considered and critically reviewed. Such a combination is relatively uncommon in previous reviews in this field [52–55]. Finally, our team also speculates on future research directions based on our previous relevant studies and projects, proposing that photoelectric composite non-homogeneous catalysis will have a larger scope for development in the future.

2 Anode materials in electrocatalytic oxidation

In electrocatalytic oxidation, the oxidizing substances produced by the anode play an important role in the degradation of pollutants. Howerer, oxygen evolution reaction occurs at the anode increases the energy consumption of the reaction at the same time, which requires the anode material to have higher oxygen evolution potential, good electrocatalytic performance, and stability [56–58]. Pt has stable chemical properties, high reaction activity, and high oxygen evolution potential, but the use of Pt is limited due to the high cost. Graphite electrodes have good conductivity, chemical inertness, and low price, but their mechanical strength is relatively low [59]. Moreover, graphite electrodes are easy to swell and peel in acidic media. In recent years, new electrode materials represented by lead dioxide electrode materials, DSA and BDD electrodes have been extensively studied. Their chemical properties are stable but the price is not cheap, especially BDD electrodes. They are currently the two most widely used anode materials in electrocatalytic oxidation. Lastly, there are newly developed electrodes based on substoichiometric titanium oxide electrode which have been applied for electrochemical wastewater treatment of different classes of organic pollutants. Although the Ti_4O_7 electrode is less capable of mineralising organic matter such as dyes than the BDD electrode, it is still significantly better than the DSA and Pt electrodes. In addition, its cost is also advantageous [60,61].

2.1 Metal electrode

Metal active electrodes have always been considered as a good choice for electrocatalytic degradation of dye wastewater. Pt, Ru and Ir are commonly applied as metal electrodes materials [62–71]. In the past 15 years, doped and undoped PbO_2 and DSA electrodes has been widely studied and applied [72–89]. The following section summarizes the corresponding research program.

2.1.1 PbO_2 electrode

Pure PbO_2 has a high oxygen evolution potential. While the durability of it limits the widely application due to surface corrosion. Sometimes the highly toxic Pb^{2+} is released when PbO_2 act as the electrode corrodes, causing serious secondary pollution [90,91]. The the new doped PbO_2 electrode proved to be more stable and active [75,92]. Therefore, the use of doped PbO_2 anodes for the electrocatalytic oxidation of dyes has received extensive attention from the scientific community in the past 10 years [55].

Samarghandi et al. used electrocatalytic precipitation to prepare a graphite doped PbO_2 anode. The experiment results exhibited that the removal efficiency of G/β-PbO_2 for methylene blue is 96.2% at the optimal reaction conditions (pH of 5.75, reaction time is 50 min, current density is 10 $mA \cdot cm^{-2}$, and electrolyte concentration of Na_2SO_4 is 78.8 $mg \cdot L^{-1}$). Noteworthy, it was much higher than the removal efficiency of ordinary graphite electrodes. Scanning electron microscopy (SEM) and X-ray diffraction (XRD) analysis showed that graphite matrix was uniformly covered by β-PbO_2 film with the form of pyramidal clusters. The results showed that this highly dense structure of β-PbO_2 does help to improve the catalytic oxidation ability of the electrode [93].

In addition to the modification method of doping carbon-based materials into PbO_2, many scholars have also tried to dope active metal elements in PbO_2 electrode to improve its electrocatalytic efficiency. For example, a novel Ce-PbO_2/C electrodes prepared by Hu et al. showed excellent catalytic activity in the electrocatalytic treatment of acid red B wastewater. By characterizing the target electrodes, they found that the doping process of Ce in PbO_2 films can reduce the crystal size of the anode, increase the specific surface area of the anode and improve the oxidation capacity of the electrode. Cyclic voltammetry experiments showed that the oxidation peak potential of the Ce-PbO_2/C electrode was smaller than that of the pure PbO_2 electrode. The Ce-PbO_2/C electrode had a wider oxidation peak and a stronger ability to generate reactive groups. Under the optimal reaction conditions, the chemical oxygen demand (COD) removal rate in solution can reach 90.17% and the decolorisation rate can reach 99.98%. The gas chromatography and mass spectrometry (GC-MS) analysis showed that the final oxidation products of acid red B are carbon dioxide and water, which can fully meet the requirements of green chemistry [94]. Yang et al. prepared Nb/PbO_2 electrodes by electrodeposition and used them as anodes for the electrocatalytic degradation of methyl orange. Under the conditions of pH = 6.0, temperature of 45 °C, current density of 50 $mA \cdot cm^{-2}$, electrolyte (Na_2SO_4) concentration of 0.08 $mol \cdot L^{-1}$ and electrolysis time of 30 min, the decolorisation and COD removal rate reached 99.6% and 72.6%, respectively. A series of characterization showed that the Nb/PbO_2 electrode surface was dense and homogeneous, and mainly composed of β-PbO_2 and a small amount of α-PbO_2 [95].

Different power supply methods will have a certain impact on the degradation of dye wastewater. Lei et al. studied the removal efficiency of acid red G and crystal violet on Ti/Sb-SnO_2/α-PbO_2/β-PbO_2 electrode using pulsed electrocatalytic oxidation technology. The results showed that the current density and pulse duty cycle had a positive effect on the pulse electrocatalytic degradation of acid red G and crystal violet. Besides, the pulse constant current power supply method had better COD removal efficiency than that of the direct current constant current power supply method [96].

Other interesting results have been reported by Elaissaoui et al. who improved the removal efficiency of dyes by

changing the structural composition and properties of lead dioxide electrodes. They fabricated a stainless steel/SiO_x/PbO_2 electrode with a SiO_x intermediate layer, and compact structure and high interconnection of oxide particles by a continuous cross-flow method. By improving the structural composition and hydrophobicity of the PbO_2 electrode, the efficiency of electrocatalytic degradation capability of PbO_2 was improved [97]. Some authors have attempted to cover the electrode surface with polymeric organic substances, which could alter the nucleation of crystals on the surface of the substrate. Jin et al. found that the Ce-PVP-PbO_2 electrode had smaller crystal particles and a denser structure compared to that of PbO_2, Ce-PbO_2 and PVP-PbO_2 electrodes. It also had lower energy consumption and better electrocatalyticl oxidation performance. As Fig. 2 shown below, the removal rates of COD and decolorization methyl orange solution (100 mg·L^{-1}) by electrolysis for 2 h were 91.8% and 99.9%, respectively [98].

Lead dioxide electrodes have shown relatively satisfactory results in the treatment of actual wastewater, which provides ideas for possible industrial treatment. For instance, Aquino et al. conducted an experiment on the degradation of actual dye wastewater by the β-type PbO_2 electrode. The results suggested that Ti-Pt/β-PbO_2 electrode had higher catalytic oxidation efficiency, and higher COD removal rate than that of pure mass-transport controlled process [69]. Comparing with DSA electrode, energy consumption was close, but the degradation efficiency was better. Mukimin et al. tried to use Ti/PbO_2 paired with a stainless steel cathode in the electrocatalytic tube reactor to degrade dye wastewater. Under the optimal reaction conditions, the biochemical oxygen demand in the water can be degraded by 95%, but the COD degradation rate was only 59% [99]. Common electrode materials in the field of dye wastewater treatment were shown in the Table 1. Through comparison, it was found that under neutral conditions, Ce-PbO_2/C and Ce-PVP/PbO_2 had the best effect.

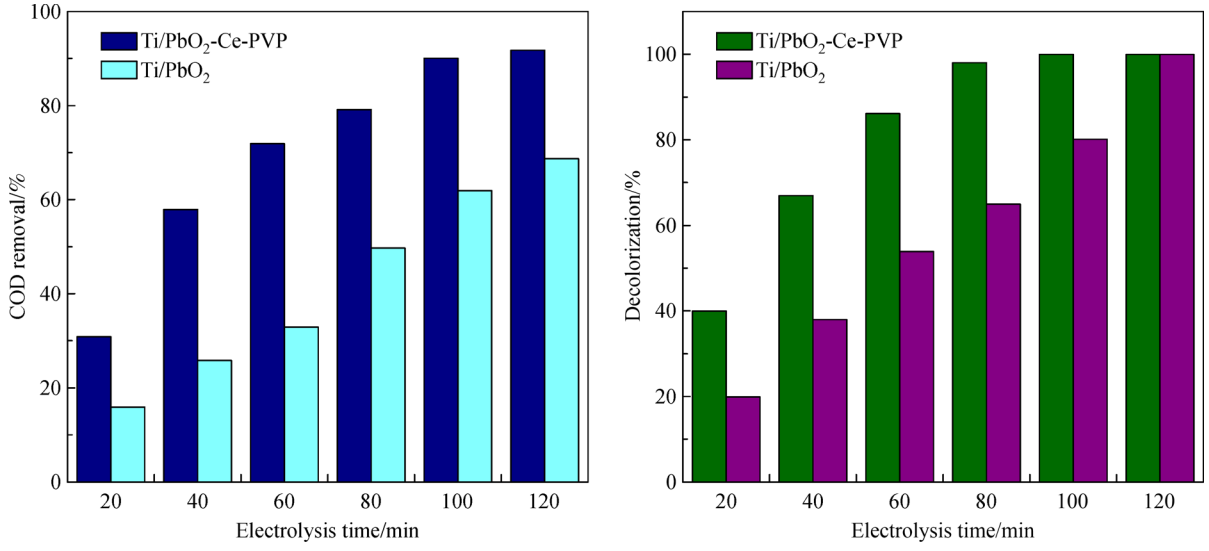

Fig. 2 The COD and decolorization of methyl orange as a function of electrolysis time on different PbO_2 anodes. Condition: temperature = 25 °C; j = 50 mA·cm^{-2}; [C_{14} H_{14} N_3 SO_3 Na] = 100 mg·L^{-1}; [Na_2SO_4] = 0.05 mol·L^{-1} [98].

Table 1 Different kinds of modified PbO_2 electrodes

Anode type	Targeted dye	C_0 /(mg·dm^{-3})	j /(mA·cm^{-2})	pH	Color decay/%	COD decay/%	Ref.
Ti/Sb-SnO_2/α-PbO_2/β-PbO_2	Acid red G	200	50	–	100	50	[96]
	Crystal violet	200	50	–	100	50	[96]
Ti/β-PbO_2 (Cylinder)	Industrial wastewater	–	5.6 [b]	6	–	59	[99]
Ce-PbO_2/C	Acid red B	1000	10 [b]	6.5	99.9	90.2	[94]
Ce-PVP/PbO_2	Methyl orange	100	50	–	100	91.8	[98]
Nb/PbO_2	Methyl orange	30	50	6	99.6	72.6	[95]
G/β-PbO_2	Methylene blue	60	10	5.8	96.4	–	[93]
SS/SiO_x/PbO_2	Amaranth	0.015 [a]	25	<7	100	84	[97]
Ti-Pt/β-PbO_2	Industrial wastewater	–	75	>7	–	88	[69]

a) mmol·L^{-1} concentration; b) voltage.

2.1.2 DSA electrode

Indirect electrocatalytic oxidation has been widely used in recent years and has gradually become an efficient alternative method. As a specific electrode material of indirect electrocatalytic oxidation, DSA electrode is formed using titanium metal as the substrate and platinum group metal oxide as the main active component on surface. Active coatings mainly include Ti, Ru, Ir, Sn, Ta and Sb.

Feng et al. prepared Ti/RuO_2-Pt anode and applied it to degradate acid orange 7 (AO7). The results exhibited that Ti/RuO_2-Pt anode electrolysis can effectively decompose and mineralize AO7 dye. Under the conditions of current density of 10 mA·cm^{-2}, pH of 6.8, and NaCl concentration of 0.001 mol·L^{-1}, the removal rate of all AO7 can reach 100% within 4 h, which implied that Ti/RuO_2-Pt anode had strong anti-passivation ability and the potential for continuous application [100]. In addition, many studies have also reported that similar DSA electrodes have good corrosion resistance. For example, Xu et al. used sol-gel and electrodeposition methods to prepare rare earth cerium-doped Ti/nano TiO_2/PbO_2 electrodes (Ti/Ce-nano TiO_2/Ce-PbO_2). SEM images indicated that the surface of Ti/Ce-nanoTiO_2/Ce-PbO_2 electrode was more uniform and had smaller particles [101].

Among the many types of DSA electrodes, except for Ru and Pt, Ir and Pb can also be used as catalytic active component. Isarain-Chávez et al. compared the degradation efficiency of several different DSA electrodes on methyl orange. The results showed that Ti/Ir-Pb had the best COD removal effect under the same conditions. Basing on the characterization of energy dispersive X-ray spectroscopy, XRD and transmission electron microscope, it was found that the electrode contained a variety of oxides such as IrO_2, Pb_2O_3 and Pb_3O_4 [102]. A possible conjecture that metal oxides on electrodes can enhance catalytic degradation was put forward. Similarity, the experiment results obtained by Baddouh et al. confirmed the judgement. They studied the oxidation effect of rhodamine B (RhB) dye on the Ti/RuO_2-IrO_2 electrode, and the best efficiency was obtained at a pH of 6.5 and a temperature of 25 °C. After 90 min of electrolysis, the removal rates of chroma and COD were 100% and 61.7%. Compared with the SnO_2 electrode, it had better efficiency and cost-effectiveness. The reason may be that more strong oxidant species such as ClO^- ions can be produced in the electrolysis process. However, it was not as efficient as the Ti/Ir-Pb electrode just mentioned. The phenomenon showed from the side that the presence of metal oxides was not the main factor in improving the catalytic efficiency of the electrode [103].

It was found that Ti/$Ru_{0.3}Ti_{0.7}O_2$ was widely mentioned as an efficient electrode material due to the environment-friendly nature. Silva et al. tested the electrocatalytic oxidation of two reactive dyes on RuO_2 DSA® electrodes. Ti/$Ru_{0.3}Ti_{0.7}O_2$ is the most active material for oxidation of organic compounds [104]. Because of its outstanding characteristics, Baddouh et al. further studied the electrocatalytic performance of this electrode in an electrocatalytic flow battery system. The effects of several factors on the degradation rate were investigated, such as supporting electrolyte, current density, electrolysis time and temperature. As shown in Fig. 3, the electrocatalytic efficiency was higher when sodium chloride solution was used as the electrolyte solution than sodium sulfate solution. When the applied current density was 3.9 mA·cm^{-2} and the reaction time was 180 min, the COD removal rate reached 93.0%, and the energy consumption was 2.98 kWh·m^{-3} [105]. Based on these studies, some scholars further explored the optimal reaction conditions for this electrode. A 2^3 full factorial design combined with response surface methodology was investigated by Santos et al. Using DSA electrodes (Ti/$Ru_{0.3}Ti_{0.7}O_2$) for the degradation of dye wastewater. It was found that the removal efficiency was higher in the presence of sodium chloride fraction, probably due to the generation of active chlorine during the electrolysis process increasing the reaction kinetics [106].

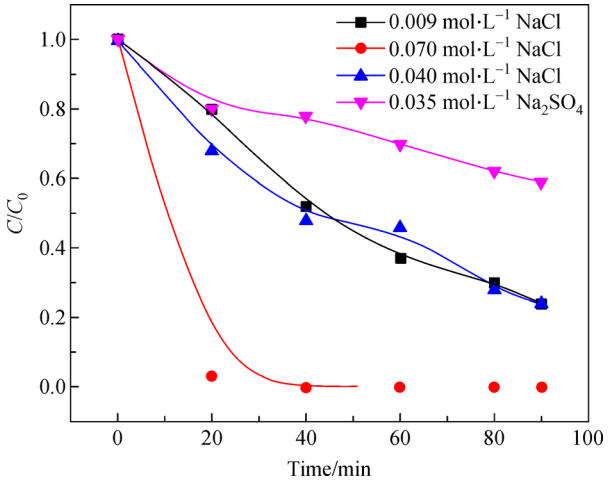

Fig. 3 Effect of NaCl and Na_2SO_4 and their concentration on the dye removal. Conditions: current density, 5.4 mA·cm^{-2}; [RhB], 80 mg·L^{-1}; *T*, 25 °C; flow rate, 30 L·h^{-1} [105].

DSA electrodes can also be used for the treatment of actual wastewater, which is a guide for potential industrialization. Faridayunus et al. used precious metal coated titanium mesh as a modified DSA electrode to degrade rhodamine 6G. Under the optimal experimental conditions, a decolorization efficiency of over 99.5% and a low energy consumption of 1.58 kWh·m^{-3} can be achieved. In addition, it was proved that NaCl is the electrolyte solution with the best degradation effect among sodium salts. The RuO_2 coated Ti mesh anode had good recyclability and longevity, and it was expected to be

further made in scale for the comprehensive treatment of industrial dye-containing wastewater [107].

DSA electrodes still have some shortcomings and limitations. Compared with other types of electrodes, the degradation efficiency of DSA electrodes is lower. For example, Wächter et al. studied the removal efficiency of acid yellow 49 dye with three different anodes, BDD, β-PbO_2, and DSA® Anodes. The results showed that BDD anodes had the best performance and DSA® anodes had the worst performance. Because of the low efficiency and high energy consumption of DSA electrodes, the current study of electrocatalytic anode materials is a tendency toward BDD anodes for the treatment and disposal of dye wastewater [108]. As shown in Table 2, the authors compared different types of DSA electrodes and found that Ti/nano TiO_2/PbO_2 electrode had the best degradation efficiency and Ti/RuO_2-Pt electrode was more energy efficient and had good degradation effect at the same time.

2.2 BDD electrodes

Pure diamond is not electrically conductive. Whereas, doping B elements can enhance the conductivity of diamond. The conductivity of the electrode is related to the content of B doping [109]. Since Carey first patented BDD in 1995, the advantageous BDD can be used as a new potential anode material for the electrocatalytic oxidation of organic matter in wastewater [110]. BDD is considered as an ideal electrode for electrocatalytic oxidation with a high ·OH yield through the generation of hydroxyl radicals by spin trapping [111]. At present, BDD coated electrodes have been widely used in various industries such as agro-pharmaceutical, printing and dyeing and other high concentration, difficult to degrade organic pollutant wastewater treatment, and the electrode can effectively mineralise the organic matter in the wastewater to achieve the ideal degradation treatment effect. There are some advantages of BDD electrodes for wastewater treatment. First, it can produce intermediate substances with strong oxidation, which further decomposes the pollutants in water into carbon dioxide without producing secondary pollution. Secondly, it has strong electrocatalytic stability to prevent the contamination of electrode surface, resulting in high activity of the electrode and current efficiency for a long time even in some acidic and alkaline solutions. Finally, it does not dissolve compared to transition metal oxide electrodes such as PbO_2 and SnO_2, and is more resistant to corrosion [112,113].

Ramírez et al. studied the reaction process of removing methyl orange azo dye using the BDD electrode. Liquid chromatograph-mass spectrometer analysis revealed the formation of seven oxidation products deriving from the cleavage of the –N5N– group of the dye, followed by deamination, formation of a nitro group and desulfonation of the resulting aromatics [114].

2.2.1 Ti/BDD

The effective electro-oxidation of dyes with small Ti/BDD anodes has beeninvestigated by Migliorini et al. In the article, two different doping levels of Ti/BDD electrodes were prepared by Hot filament chemical vapor deposition and the electrocatalytic activities for remove dye were studied. SEM and Raman measurements showed that both electrodes can effectively degrade dye substances. The electrocatalytic efficiency of the Ti/BDD electrode was higher at current densities above 75 $mA \cdot cm^{-2}$, which may be due to the smaller grain size. In addition, the doping level can also have effect on the efficiency of the diamond electrode [115]. Palma-Goyes et al. studied the degradation of crystal violet by Ti/BDD electrodes. The study investigated the effects of different experimental parameters on the results and intermediate by-products in the degradation process. The intermediate substances were mainly *N*-methylaniline, *N*,*N*-dimethylaniline, 4-methyl-*N*, *N*-dimethylaniline, and other aromatic compounds detected by GC-MS. The experiment results showd that the degradation pathway of crystal violet was related to the free radicals generated around the electrode [116].

To further improve the catalytic activity of Ti/BDD

Table 2 Common DSA electrodes

Anode type	Targeted dye	C_0/($mg \cdot dm^{-3}$)	j/($mA \cdot cm^{-2}$)	Color decay/%	COD decay/%	Ref.
Ti/RuO_2-IrO_2	RhB	50	40	100	61.7	[103]
Ti/RuO_2-Pt	Acid orange 7	100	10	100	79.5	[100]
Ti/RuO_2-Ta_2O_5	Reactive blue 4	100	50	100	80	[103]
	Reactive orange 16	100	50	100	80	[103]
Ti/nano TiO_2/PbO_2	Methyl orange	50	50	100	96.6	[101]
Ti/SnO_2-Sb-CNT	Acid red 73	1.0 [a)]	50	100	80.1	[102]
Ti/$Ru_{0.3}Ti_{0.7}O_2$	Reactive yellow 145	0.75 [a)]	10	48.1	–	[106]
	Reactive blue 19	0.75 [a)]	10	78	–	[106]
	Reactive red 195	0.75 [a)]	10	75	–	[106]

a) $g \cdot L^{-1}$

electrodes, some scholars tried to modify their surfaces, such as porous, granular, etc. He et al. used porous Ti/BDD electrode to degrade triphenylmethane dye xylenol orange. The results showed that the porous Ti/BDD electrode had a higher removal efficiency for the target dye than that of the flat BDD electrode. It was speculated that the porous BDD membrane can provide more active sites for hydroxyl radicals and the active chlorine in the solution, and further promoted the removal of COD and chromaticity [117].

Multiple process mixing is a viable approach to treating dye wastewater, which can improve the removal efficiency for target contaminants. Juang et al. designed an innovative Ti/BDD electro-oxidation-microfiltration (EO/MF) hybrid system to treat acid yellow 36 dye wastewater. The results showed that EO/MF had a good COD removal effect on dye wastewater, and the removal rate of chroma and turbidity was over 90%. It can be seen that MF did not affect electrocatalytic oxidation, and the process had good application prospects [118]. Daghrir et al. compared the degradation effects of different anode materials on dye wastewater. The results showed that the electrocatalytic activity of the Ti/BDD anode was higher than that of the other two DSA electrodes (Ti/IrO_2 and Ti/IrO_2-RuO_2), which implied that BDD electrode was a alternative choice for treating dye wastewater [119].

Mei et al. obtained high-quality 3D-BDD electrodes with uniformly distributed pores on nickel foam using chemical vapor deposition at a low current. The diameter of the hole was 1 mm, and the width of the skeleton was 200 μm. A connected channel was formed on the electrode for the passage of liquid phase. The peak current of the 3D-BDD electrode was much higher than that of the 2D-BDD electrode. In addition, the resistance of 3D-BDD electrode was much lower than that of the 2D-BDD electrode. In the process of treating reactive blue 19, it showed exciting catalytic activity [120].

2.2.2 Nb/BDD

Brito et al. comparatively studied the performance of Nb/BDD and Si/BDD anodes for the acid violet 7 dye removal. The study found that Nb/BDD had a slightly higher oxygen evolution reaction potential than Si/BDD, indicating that more ·OH was produced on its surface. 100% decolorization can be completed within 120 min at a suitable current density on both electrodes [121]. Moreover, Brito et al. also investigated the effect of different reactors on textile wastewater using Nb/BDD electrodes. The results showed that the parallel I–O flow cell had a higher COD removal rate than the perpendicular I–O flow reactor when all other reaction conditions being equal. This phenomenon may be due to that there may be some stagnation in the lateral region of the electrodes, which facilitated a longer contact time between the pollutants and the anode [122]. Fajardo et al. studied the removal effect of Ti/IrO_2-Ta_2O_5 and Nb/BDD anodes on amaranth dye. For the Nb/BDD anode, the total chromaticity was eliminated after 30 $mA \cdot cm^{-2}$ treatment for 60 min, and the COD removal rate was 49.1%. Under the same conditions, after 360 min of electrolysis time, the decolorization rate of Ti/IrO_2-Ta_2O_5 electrode was 98.5% and the COD removal rate was 43.2%. As shown in Fig. 4, Nb/BDD degraded 50% initial concentration of COD at 30 $mA \cdot cm^{-2}$ current density for three hours. The effect was better than the electrolysis effect of Ti/IrO_2-Ta_2O_5 electrode at 60 $mA \cdot cm^{-2}$ current density. It can be seen that the Nb/BDD anode had better mineralization ability than the Ti/IrO_2-Ta_2O_5 electrode [123]. Compared to other types of BDD electrodes and DSA electrodes, the Nb/BDD electrode has a better ability to mineralise organic matter. However, due to the high cost of Nb, it is difficult to be produced on an engineering scale.

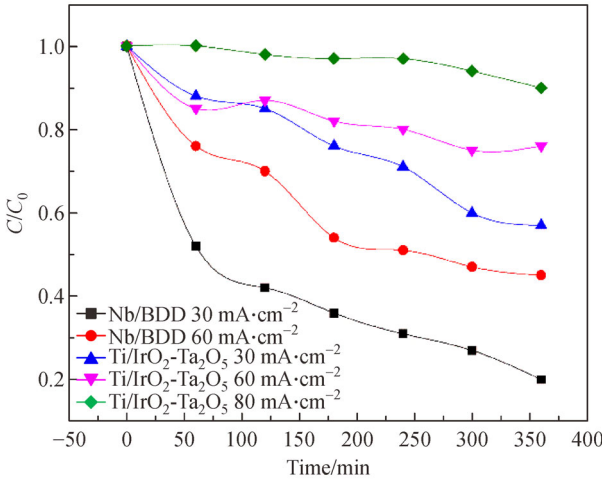

Fig. 4 Oxidation of the amaranth dye using a single cell. Effect of the anode material on COD concentration. Operating conditions: 100 $mg \cdot L^{-1}$ of amaranth dye, 20 $g \cdot L^{-1}$ of Na_2SO_4, current density = 30, 60 and 80 $mA \cdot cm^{-2}$, t = 25 °C [123].

Soni et al. investigated the electrocatalytic activities of Nb/BDD, Si/BDD and DSA electrodes for Reactive black 5 removal. The results showed that the amount of wastewater treated by the continuous process was much larger than that of treated separately. Besides, the dual effect of two different electrodes enhanced the removal rate of total organic carbon (TOC). In addition, the mineralization efficiency of continuous operation was higher than that of batch process. The results also implied that the combined effect of DSA and BDD electrodes was more effective. Therefore, industrial-scale dye removal can be achieved at a lower cost using the dual effects of mixed metal oxides and BDD electrodes [124].

2.2.3 Si/BDD

Si/BDD anode is the most widely used diamond material in the field of electro-oxidation of dyes. Bogdanowicz et al. prepared a series of novel electrodes with different B-doping levels. By analyzing the Raman spectrum, it was found that boron atoms were introduced into the diamond lattice to obtain the desired doping concentration. The results indicated that the higher the doping concentration, the higher the removal efficiency of dye. Noteworthly, as the concentration of boron atom doping increased, the amounts of small-diameter crystallites on the diamond surface increased, which can enhance the performance of electrocatalytic oxidation [125].

Santos et al. studied the electrocatalytic oxidation reaction of methyl red on Pb/PbO_2, Si/BDD and Ti/Sb doped SnO_2 anodes. The results showed that methyl red can be oxidized on all three electrodes. However, there was a significant difference between the oxidation level and the oxidation efficiency. Among them, the oxidation efficiency of the Si/BDD electrode was the highest. It only took 15 min to completely oxidize methyl red to carboxylic acid. The oxidation rate on Pb/PbO_2 and Ti/Sb doped SnO_2 electrodes was slow, and many organic intermediate products such as 2-aminobenzoic can be detected. From this comparison, Si/BDD electrode exhibited the excellent electrocatalytic oxidation efficiency [126]. Zhou et al. studied the removal efficiency of methyl orange on Si/BDD electrode and mixed metal oxide electrode. The effects of different factors including current density, pH, and electrolyte solution on the removal efficiency of the two electrodes were investigated [81]. Under the same current density conditions, Si/BDD electrode was obviously more efficient to decolorize than mixed metal oxide electrodes. The Si/BDD electrode was not sensitive to pH changes, and its performance was slightly better under acidic condition. Different electrolyte solutions had a greater impact on the removal efficiency of methyl orange. In sodium sulfate solution, the color removal rate can only reach 10%, but in sodium chloride electrolyte solution, the color removal rate can be close to 100%. However, the removal efficiency of COD showed a difference. The removal rate in sodium chloride solution was much higher than that in sodium sulfate solution. These facts confirmed that the presence of NaCl not only promoted decolorization, but also promoted mineralization.

To improve the catalytic oxidation efficiency of dye wastewater, scholars tried to combine a variety of advanced oxidation technologies to achieve efficient degradation of organic dye wastewater. For example, Zhu et al. studied the ultrasonic-enhanced electrocatalytic oxidation of a typical alizarin red S (ARS) dye on a BDD anode. The results showed that in the ultrasound enhanced electrochemical oxidation (US-EO) system, the removal efficiency of COD was inversely proportional to temperature, and the removal rate of COD reached the highest value of 86.07% when the pH was 4.97. The GC-MS analysis showed that ARS treatment with US-EO for 5 min resulted in the intermediate products 1,2-dihydroxyanthraquinone and 9,10-anthracenedione. If the reaction time was further extended, COD can be completely removed. The synergistic effect of sonochemistry and electrocatalytic treatment on the organic matter is a problem worthy of research [127].

As shown in Fig. 5, several different sets of comparisons show that the performance and energy consumption levels of the BDD series electrodes for COD removal are better than most DSA and PbO_2 electrodes [80,119,123,125,126]. Among the different types of BDD electrodes, Nb/BDD is the most effective, but is expensive and difficult to apply to the treatment of actual dye wastewater. Taking all factors into account, Si/BDD electrodes are currently the most widely used electrode material. In addition to the modification of the electrodes themselves, researchers are now focusing on the synergistic treatment of wastewater with different types of electrodes, aiming to maximise the dye explanation within the allowed energy and cost limits.

As shown in Table 3, the Nb/BDD electrode had the best performance, and the COD removal rate was basically maintained above 90%. The Ti/BDD electrode was also ideal for dye removal, which was largely better than the Si/BDD electrode.

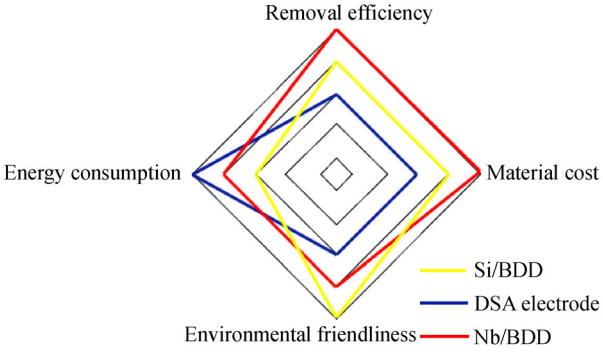

Fig. 5 Comparison of the performance of different types of electrodes.

3 Three-dimensional electrode

The three-dimensional electrode method is adding particle electrodes between the anode and cathode electrodes of the two-dimensional electrode reactor, and apply an appropriate voltage to charge the surface of each particle electrode to form the third electrode of the reaction system. The mechanism of the three-dimensional electrode is shown in Fig. 6. The charged particles added to the system increase the surface area of the electrode, which can

Table 3 Common BDD electrodes

Anode type	Targeted dye	C_0/(mg·dm^{-3})	j/(mA·cm^{-2})	Color decay/%	COD decay/%	Ref.
Ti/BDD	Xylenol orange	200	30	100	79	[117]
Si/BDD	Reactive black 5	100	50	99	90	[124]
Nb/BDD	Reactive black 5	100	50	99	90	[124]
BDD	Crystal violet	250	2.5	100	–	[116]
BDD	Methyl orange	100	31	94	–	[114]
Si/BDD	Acid violet 7	200	15	100	48	[121]
Nb/BDD	Acid violet 7	200	15	100	60	[121]
Ti/BDD	Domestic wastewater	–	16.5	89.5	78.2	[119]
Nb/BDD	Amaranth	100	30	100	49.1	[123]
3D-BDD	Reactive blue 19	400	–	100	84.5	[120]
Ti/BDD	Reactive orange 16	50	75	98	–	[115]
Ti/BDD	Acid yellow 36	20	30	>90	100	[118]

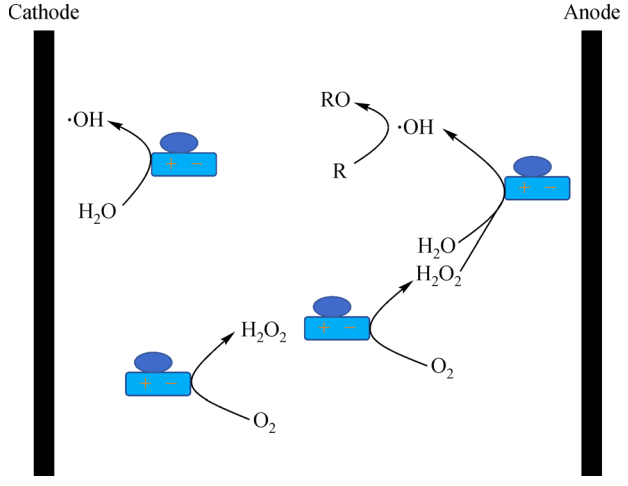

Fig. 6 Mechanism of the three-dimensional electrode reactor.

produce more oxidizing substances such as ·OH and H_2O_2, and contribute to the efficient electrocatalytic oxidation. Compared with the traditional two-dimensional flat electrode, the electrocatalytic oxidation method of the three-dimensional electrode has the advantages of large volume, large processing capacity per cell, and high current efficiency. In the 1960s, Backhurst et al. first proposed three-dimensional electrodes. In the 1990s, Brown et al. explored the application of three-dimensional electrodes in wastewater treatment. In recent years, it has been widely used in dye wastewater treatment. In the treatment of dye wastewater, the common three-dimensional electrodes materials are carbon-based materials, metal oxides materials, ceramics and minerals.

Liu et al. used foamed nickel as a particle electrode to degrade RhB [128]. The novel three-dimensional electronic Fenton system can degrade 99% of RhB, much higher than the group without three-dimensional electrodes. The phenomenon might be due to that the foamed nickel particle electrode can activate molecular oxygen to produce $\cdot O_2^-$, which can effectively oxidize organic material. Based on this study, some articles have tried to load active metals on nickel foam to further improve its performance. Liu et al. modified the foamed nickel and loaded Pd and Fe on Ni to form a new particle electrode. The evaluation results showed that this particle electrode can quickly generate hydroxyl radicals. The removal performance on the target substance was excellent, and the particle electrode exhibited good stability and activity. The two loaded elements played important roles in accelerating electron transfer [129]. He et al. used kaolin doped with iron molybdate as the third pole to treat methyl orange wastewater. When the methyl orange concentration was 100 mg·L^{-1}, the pH was 4.34, the particle electrode mass concentration was 6.6 g·L^{-1}, the electrolyte concentration is 0.05 mol·L^{-1}, and the current density of the electrode per square centimeter is 68 mA, the dye wastewater was almost completely decolorized, and the COD removal rate can reach 92% [130]. Zhang et al. studied the degradation performance of dye wastewater by the coordinated ordered mesoporous carbon (OMC) three-dimensional electrode loaded with Ni (Ni/OMC) and Ce (Ce/OMC) and the ozone oxidation method. As shown in Fig. 7, the particle electrode loaded with Ni and Ce accompanied with ozone aeration had the highest degradation efficiency of COD in wastewater, which was higher than that of the catalyst loaded with one metal alone. The high performance of $Ni_{0.2}$-$Ce_{0.2}$/OMC catalyst may be due to the better dispersion degree of metal on the OMC carrier, which can provide more active sites, increasing the number of oxygen vacancies and facilitating electrocatalytic and ozonation reactions [41].

Li et al. prepared carbon (GAC) coated with SnO_2-Sb

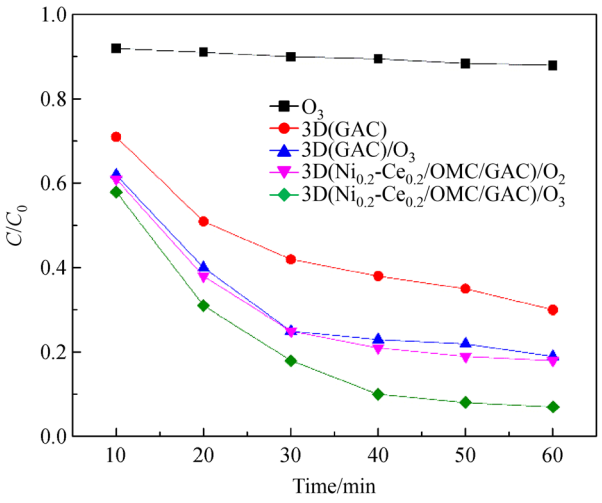

Fig. 7 COD concentration by different processes [41].

doped TiO_2 (GAC/ATOT) by the sol-gel method as a three-dimensional electrode to treat RhB dye wastewater. The results showed that compared to TiO_2 coated GAC (GAC/T), Sb doped SnO_2 coated GAC (GAC/ATO), and pure GAC, GAC/ATOT electrode can effectively increase the removal rate of COD in wastewater and reduce the energy consumption of the reaction which mainly due to that GAC/ATOT can greatly increase the yield of ·OH. It was proved that the high yield of ·OH can improve the electrocatalytic oxidation performance of organic compounds [131]. Wei et al. prepared a high-impedance Sb-doped SnO_2 porous ceramic particle electrode and observed its performance in the process of catalytic degradation of the non-biodegradable reactive brilliant red X-3B dye. Compared with the two-dimensional electrode system, when the voltage was 13 V and the treatment time was 60 min, the three-dimensional particle electrode system increases the COD removal rate of reactive brilliant red X-3B wastewater by 32.9% and reduces energy consumption by 33.3%. Cyclic voltammetry curve analysis showed that the contribution of direct oxidation in the three-dimensional electrode system was weaker than that of the two-dimensional electrode system. Under the excitation cell voltage, the amounts of hydroxyl radicals in the three-dimensional electrode system are more than that in the two-dimensional electrode system [132].

Yue et al. used Fe_2O_3/γ-Al_2O_3 as a three-dimensional electrode Fenton-like catalyst to study the mineralization and degradation effects of acid red 3R in dye wastewater under different experimental parameters. Under the conditions of an external voltage of 20.0 V, a pH of 3.0, an airflow of 0.24 $m^3 \cdot h^{-1}$, and an electrode spacing of 3.0 cm, the decolorization efficiency can reach 77.6% after 100 min. The catalyst can effectively catalyze the generation of ·OH from hydrogen peroxide, and reach the complete mineralization of dye pollutants with the generation of CO_2 and H_2O [133].

Three-dimensional electrodes are more efficient to process and are relatively easy to recycle. Such advantages enable it can be potential for large-scale industrialisation. A lot of basic research has been done on it, and the next research should still focus on the study of loaded three-dimensional electrodes. How the reaction efficiency can be improved and ensuring environmental friendliness are should be worth considering. The common three-dimensional electrode in the field of dye wastewater treatment were shown in Table 4. By comparison, it was found that the particle electrode with kaolin as the carrier and the particle electrode with OMC as the carrier had the best catalytic effect, and the active components were often metallic elements such as iron, nickel, cerium, etc.

4 Integrated electrocatalytic technology in dye wastewater treatment

To further improve the degradation efficiency of dye wastewater, some researchers combined electrocatalytic oxidation with other catalytic technologies in a reaction system for synergistic catalysis, such as photoelectric catalysis.

Shen et al. prepared nano-TiO_2 catalyst and Co-Bi-PbO_2/Ti anode to treat dye wastewater together. The influence of electrolyte concentration, temperature, stirring, voltage and other factors on the decolorization efficiency were investgated. Generally speaking, in a certain range, the removal rate is proportional to each influencing factor. The results showed that when the voltage was 3 V and the electrolyte concentration was

Table 4 Common three-dimensional electrode

Anode type	Targeted dye	C_0/(mg·dm^{-3})	pH	Color decay/%	COD decay/%	Ref.
$Fe(MoO_4)_3$-Kaolin	Xylenol orange	100	4.34	100	92.48	[130]
Foam nickel	Reactive black 5	5	6.20	99	–	[128]
Ni-Ce/OMC	Reactive black 5	–	7.50	–	93.70	[41]
GAC/ATOT	Crystal violet	150	7.00	–	70.00	[131]
Fe_2O_3/γ-Al_2O_3	Methyl orange	600	3.00	77.6	58.60	[133]
Sb-SnO_2/ceramic	Acid violet 7	–	–	–	67.30	[132]

0.01 mol·L^{-1} Na_2SO_4, the degradation effect was the best. In addition, compared with the blank control group, both the nano-TiO_2 catalyst and the Co-Bi-PbO_2/Ti anode can increase the decolorization rate by 0.15 times. But when they were combined together to treat dye wastewater, the removal efficiency can be increased by 0.22 times. The comparison showed that the removal of COD from the wastewater was somewhat improved, but the cost of the investment was too high and the improvement does not reach the expected level [134].

Titanium dioxide has a very high photovoltaic conversion efficiency and is a very widely used low cost photocatalyst today. Add a titanium dioxide photoelectric catalyst to the reactor to produce ·OH which can efficiently oxidize and degrade dye pollutants in water. Esquivel et al. developed a modified surface of optical fiber and deposited a TiO_2 layer on it. Compared with other types of TiO_2 layers, the content of photogenerated ·OH was nearly 50% higher. Under suitable conditions, the photoelectric catalytic process can achieve complete decolorization of 15 mg of azo orange II dye within 60 min, and the TOC removal rate can reach 75% [135]. Other similiar results were also reported by Bessegato et al. [136] who used boron-doped-TiO_2 nanotubes photoelectrodes as photoanode under UV-visible spectrophotometer irradiation and bias potential. They further designed a full factor experiment to explore the mechanism of photoelectric catalytic treatment. The independent variables including pH, bias potential and counter electrode material were investigated to explore the catalytic performance. Response surface methodology was used to monitor the TOC and camel rate of dye wastewater to determine the best conditions. The results showed that the optimal conditions for photocatalytic degradation of acid red 151 dye were pH 2.0 and potential 2.0 V. The complete mineralization of pollutants can be completed after 90 min of reaction.

In addition to titanium dioxide loading on the electrode surface, titanium dioxide doping in three-dimensional electrodes for photocatalysis is also a viable method. Pan et al. studied the synergistic effect of photocatalytic oxidation (PCO) and electrocatalytic oxidation (ECO) under different anode bias voltages in the presence of a TiO_2 immobilized on columnar activated carbon (TiO_2/CAC) three-dimensional electrode. When the anode bias voltage was 1.0 V, the degradation efficiency of methyl orange by the photoelectrocatalytic oxidation process reaches 98.76% at 35 min, which is much higher than that of the two processes alone. When acting alone, the COD removal efficiency of the PCO process was 62.43%, and the removal efficiency of the ECO process was 33.93%. The new type of three-dimensional electrode reactor had a good degradation effect of photoelectrocatalytic oxidation and had great application potential in wastewater treatment [137].

The degradation of solutions with 0.260 mmol·L^{-1} of Congo Red was learned by Solano et al. They studied different electrocatalytic advanced oxidation methods, such as anodic oxidation with electrogenerated H_2O_2(AO-H_2O_2), electro-Fenton and photoelectro-Fenton. The results showed that the highest oxidation capacity was photoelectro-Fenton, and the lowest was AO-H_2O_2. In all the electrochemical advanced oxidation processes, increasing current densityenhanced the degradation process, but with a loss of mineralization current efficiency and higher energy consumption. Optimum conditions were found for 0.260 mmol·L^{-1} of congo red with 0.50 mmol·L^{-1} Fe^{2+} at 100 mA·cm^{-2}, yielding almost complete mineralization in 240 min with about 49% current efficiency [138].

In exception to titanium dioxide, some other materials also show superior photocatalytic properties, such as tungsten trioxide and bismuth tungstate. Although the photocatalytic performance of single-component tungsten trioxide is not good. However, its catalytic performance can be enhanced by semiconductor compounding and metal ion doping. Compared with commonly used photocatalysts such as TiO_2 and ZnO, tungsten trioxide has a smaller forbidden band width and a larger light absorption range, and can more effectively utilize visible light, which accounts for nearly half of the energy of solar radiation. Umukoro et al. studied the use of tungsten trioxide and flake graphite (EG) as photoanodes to degrade orange II dye using electrocatalytic and photo-assisted electrocatalytic methods. Based on a series of characterization and testing, it was found that WO_3 nanoparticles with different average particle diameters were uniformly dispersed on the EG. In a 0.1 mmol·L^{-1} Na_2SO_4 solution, orange II was degraded at a current density of 10 mA·cm^{-2}. Compared with the EG electrode, the WO_3-EG nanocomposite electrode had a higher removal rate, and the dye removal rate can reach 95% [139].

5 Conclusions

Environment are closely related to human health and sustainable ecological development. In recent years, many public health safety problems are caused by the insufficient purification of industrial wastewater. Therefore, it is necessary to develop new methods to degrade organic pollutants, such as organic dyes, through a new class of advanced oxidation processes. In recent years, electrocatalytic technology catch many scholars attentions to treat dye wastewater. In addition to the research on traditional electrocatalytic techniques, many research groups have also worked to clarify the advantages of electrochemical advanced oxidation processes as potential substitutes for the removal of dyes in synthetic and industrial wastewater. Compared with other treatment methods, electrocatalytic technology has a reservoir effect, is generally more cost-effective, and requires less maintenance. This review

discusses the anode materials in detail that perform well in electrocatalytic oxidation reactions, such as DSA and BDD, and heterogeneous three-dimensional electrode catalysts. Among the many active anodes, PbO_2 electrodes always have the problem of toxic ion release, DSA electrodes are less effective in removal than BDD electrodes, which are currently the most environmentally friendly and best performing active anodes, but they are more expensive and difficult to recycle. The non-homogeneous catalytic degradation of dye wastewater, represented by the three-dimensional electrode, has a better treatment effect. It can increase the surface area of the electrode, shorten the mass transfer distance and has the potential for industrialisation. At the same time, there is still much room for improvement in how to suppress side reactions such as hydrogen and oxygen precipitation. Ultimately, the synergistic catalysis of multiple advanced oxidation methods is found to be theoretically more effective, such as photoelectrocatalytic oxidation. Electrocatalytic reactions are rarely carried out in dark rooms, which allows visible light to be used to further improve removal efficiency and keep costs within limits. At this stage, photocatalysis is mainly based on TiO_2 as the main catalyst, but cannot make use of solar energy. The next stage of research in this field should be to find photocatalysts that can make efficient use of visible light, and bismuth tungstate is currently showing some potential.

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