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Review

Progress in the Preparation of Metal Oxide Electrodes for the Electrochemical Treatment of Organic Wastewater: A Short Review

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Abstract: The direct discharge of untreated organic wastewater poses significant threats to the environment and to human health. To address these threats, electrocatalytic oxidation technology has emerged as a key solution for organic wastewater treatment. Building on research conducted over the past three years, this review highlights the considerable advantages of electrocatalytic oxidation technology in the context of organic wastewater treatment, with a particular emphasis on the application of metal oxide electrodes. The review also provides a summary of the primary methods used in the preparation of such electrodes. Subsequently, the applications of both single-metal-oxide electrodes and metal oxide composite electrodes in organic wastewater treatment are summarized. Finally, we discuss the future development of metal oxide electrodes.

Keywords: organic wastewater; human health; electrocatalytic oxidation technology; metal oxide electrodes

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1. Introduction

Organic wastewater comprises wastewater containing a large number of organic substances [1–6], typically including chemical wastewater (containing pigments, dyes, and adhesives, etc.) [7–13], agricultural wastewater (containing pesticides, herbicides, and other agrochemicals) [14–16], domestic sewage (containing excrement, detergents, food residue, etc.) [17–22] and pharmaceutical wastewater (containing drugs, antibacterial agents, and hormones) [23–25]. The substances in these wastewaters generally possess significant toxicity and the potential for bioaccumulation, posing both direct and indirect threats to the ecological environment. The impact they have on ecosystems and human health is substantial. Therefore, the treatment of organic wastewater is of paramount importance.

Currently, the common treatment technologies for organic wastewater include physical treatment technology, chemical treatment technology, biological treatment technology, and advanced oxidation technology. Physical treatment technology [26–28] usually does not involve chemical reactions or biological processes. The common physical treatment methods can be divided into two categories: the first category mainly comprises precipitation, filtration, and flotation, which are typically used as pretreatment steps. The second category mainly comprises membrane separation and depth filters, which are commonly employed in the advanced treatment stage. These methods boast several advantages, such as rapid treatment speeds and minimal impact on the natural environmental microbial community. However, these physical treatment technologies also come with limitations; they offer limited treatment effects, present unstable treatment efficiency, and may potentially generate new solid waste as a byproduct. Chemical treatment technology [29,30] uses

chemical reactions to convert organic matter in wastewater into manageable substances. Common chemical treatment methods include chemical precipitation and chemical flocculation. In the field of organic wastewater treatment, chemical treatment technology is usually used as an auxiliary method in combination with other treatment technologies to achieve better treatment effects. Notably, this technology has several strengths, such as rapid treatment speeds and a strong applicability in handling hard-to-degrade organic substances, especially those with high molecular weights. However, there are notable drawbacks to consider. Its operation entails the consumption of large amounts of chemical reagents, which may lead to secondary pollution issues. Biological treatment technology [31–33] employs microorganisms to decompose organic substances in wastewater into inorganic substances. Common biological treatment methods include the biofilm method, the activated sludge method, and aerobic and anaerobic treatments. These methods have several advantages, such as effective treatment results, low operating costs, and stable long-term operation. Additionally, the type and quantity of the microbial community can be regulated to optimize the treatment process. However, these techniques also present some challenges. They require a longer processing times compared to chemical and physical methods. They are also sensitive to fluctuations in temperature and pH, and to antibiotics and heavy metals, which can inhibit the activity of the microorganisms and thereby reduce the efficiency of the treatment process. Advanced oxidation technology [34,35] is a subset of the broader category of chemical treatment technologies, which decompose organic substances in wastewater through various chemical reactions. This technology can utilize ozone, hydrogen peroxide, and other oxidants to degrade organic substances in wastewater. It can also employ photocatalytic and electrocatalytic oxidation methods to degrade organic substances in wastewater into simple inorganic compounds. The aforementioned wastewater treatment technologies have their own characteristics, and advanced oxidation technology, represented by electrocatalytic oxidation, has been widely applied commercially in the field of organic wastewater treatment due to its operational simplicity. Compared to other advanced oxidation processes, electrocatalytic oxidation has certain advantages. It does not necessitate the addition of extra chemicals, unlike processes such as Fenton oxidation, and it has already found extensive real-world applications in wastewater treatment. Meanwhile, several other advanced oxidation technologies, exemplified by photocatalysis, are predominantly in the experimental stage, although there are instances of field applications.

In the study of electrocatalytic oxidation technology, the selection and optimization of electrode materials are key factors. At present, the electrode materials used in the related academic research and engineering practice mainly include carbon-based (such as graphite and boron-doped diamond) electrodes, precious metal (such as platinum-based alloys) electrodes, and metal oxide (such as lead oxide and ruthenium-iridium oxide) electrodes. The basic principle of the electrocatalytic oxidation treatment of organic wastewater is to accelerate the oxidation and decomposition of the organic matter in wastewater via an electrochemical reaction. Unlike the famous technique of Fenton oxidation [36,37] (which depends on the action of H_2O_2 and iron ions), electrocatalytic oxidation does not require the addition of other oxidants, but uses a current to generate redox reaction active free radicals on the electrode's surface [38–40], thus oxidizing organic compounds in wastewater. This not only prevents the residual problem of leaving potentially toxic oxidants, which is caused by some advanced oxidation technologies [41], but also saves the cost of purchasing oxidants when used in practical settings. A schematic diagram of the technical principles of using electrocatalytic oxidation for organic wastewater treatment is shown in Figure 1. The oxidation of organic pollutants mainly occurs in two ways: direct and indirect electrocatalytic oxidation. In direct electrocatalytic oxidation, organic matter is directly oxidized on the anode surface under oxidation potentials. This process transforms the organic matter into carbon dioxide, water, and other harmless substances, as depicted in Equation (1). On the other hand, indirect electrocatalytic oxidation involves the reaction of organic matter with active species, such as oxygen free radicals, hydrogen free radicals, and hydroxyl free

radicals. These active species are produced on the anode surface, leading to the generation of products similar to those produced by direct electrocatalytic oxidation.

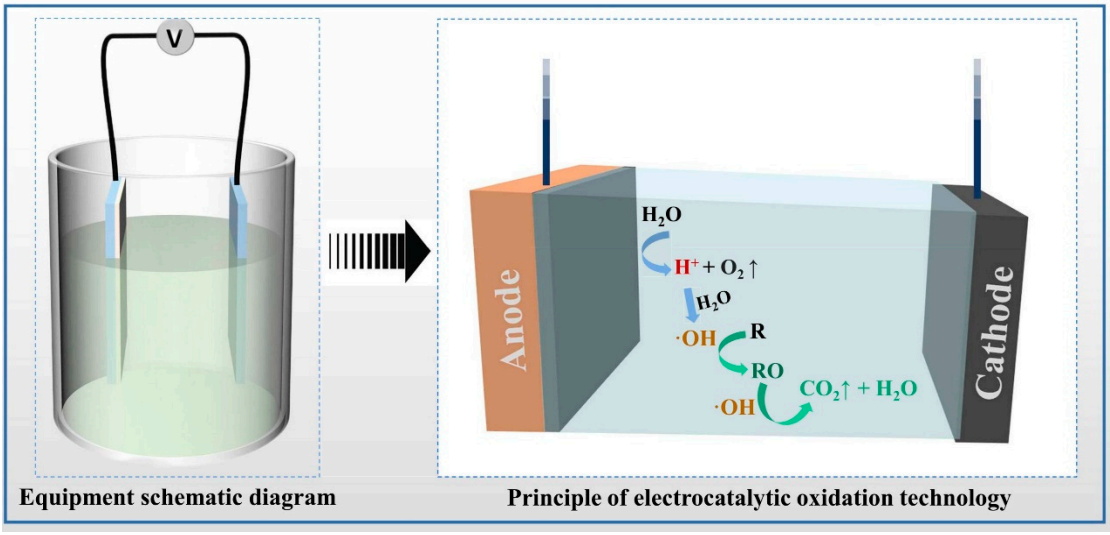

Figure 1. Schematic diagram of the equipment and related technical principles of electrocatalytic oxidation for organic wastewater treatment.

$$R + MO_X(\cdot OH) \rightarrow CO_2 + H^+ + e^- + MO_X \quad (1)$$

R: organic pollutants. MOx: active sites on the anode surface.

Due to the differences in the structure and physical and chemical properties of different active electrode materials [42–44], the efficacy of these materials in the electrocatalytic oxidation of wastewater varies. In the process of electrochemical treatment, the selection of the active electrode must usually account for the following points: the surface of the active electrode needs to have excellent catalytic performance [45–48], which means that the surface of the electrode should have a large enough specific surface area to form a large number of active sites and promote the redox reaction. The active electrode should also exhibit high conductivity and chemical stability [49–51] to ensure that it can withstand high-intensity currents while not being corroded by the chemicals in the wastewater. The cost and ease of use of active electrode materials must be reasonable [52,53]. For example, single-metal electrodes usually have the advantage of low costs, but their lower catalytic activity and narrower range of applications limit their use. In addition, it is necessary to consider the corrosion effect of different types of wastewater on the electrode [54]. For acidic or chlorine-containing wastewater, it may be more appropriate to choose active electrode materials with strong corrosion resistance.

From the above summary, it is clear that the judicious selection of active electrodes is of paramount importance for enhancing the efficiency and cost-effectiveness of electrocatalytic oxidation for wastewater treatment. Electrodes can be divided into categories based on the presence or absence of metal elements. Some carbon-based electrodes, such as graphite electrodes, lack metal elements and possess inherent conductivity and lower manufacturing costs. However, the electrocatalytic activity of these carbon-based electrodes is considerably lower than that of their metal counterparts, resulting in higher power consumption [55,56]. Furthermore, these electrodes exhibit low mechanical strength and are vulnerable to physical damage and chemical erosion, leading to unstable sewage treatment outcomes and the need for frequent replacement. Considering the limitations of current fabrication technologies, despite their excellent catalytic performance and lifespan, boron-doped diamond (BDD) electrodes still have the disadvantages of complex preparation processes and high costs [57–59]. These drawbacks add to the challenges and long-term operating costs of using carbon-based electrodes. Regarding electrodes containing metal

elements, corrosion-resistant noble metal electrodes are not widely used in sewage treatment due to their high costs. Conversely, the use of many metal-based electrodes may result in issues such as electrode corrosion and metal ion contamination [60], particularly in organic wastewater with a complex composition. In light of these challenges of using metal and carbon-based electrodes, titanium emerges as an appealing compromise. Although titanium surfaces are prone to forming dense oxide films with poor conductivity when serving as an anode [49,61], the inherent advantages of titanium in terms of its mechanical strength, electrical conductivity, and reasonable costs outweigh these drawbacks. As a result, titanium is frequently utilized as a substrate for modified electrodes, including metal oxide electrodes, in various practical applications.

Compared with the above types of electrodes, metal oxide electrodes have higher catalytic activity and electrochemical stability [62–64], which can improve the reaction rate of the electrode. They not only have a longer service life, but can also be applied in a wide range of pH values. The advantages of metal oxide electrodes include the higher number of defect sites on their surfaces, which serve as catalytic centers for more effective reaction facilitation. These electrodes possess a relatively complex structure, often comprising various metal oxides, and they typically display highly dispersed nanocrystalline grains among other forms, thereby providing a large surface area and improving the number of active sites for catalytic reactions. Additionally, most metal oxides exhibit excellent electrical conductivity and chemical stability, particularly oxidation resistance. This enhances the efficiency of electron transport and broadens the application range of the electrodes. In summary, metal oxide electrodes have the potential to be widely applied in the electrocatalytic oxidation treatment of organic wastewater, which can produce an ideal treatment effect and high stability and applicability. The main characteristics of metal oxide electrodes are compared with those of several other types of electrodes in Figure 2.

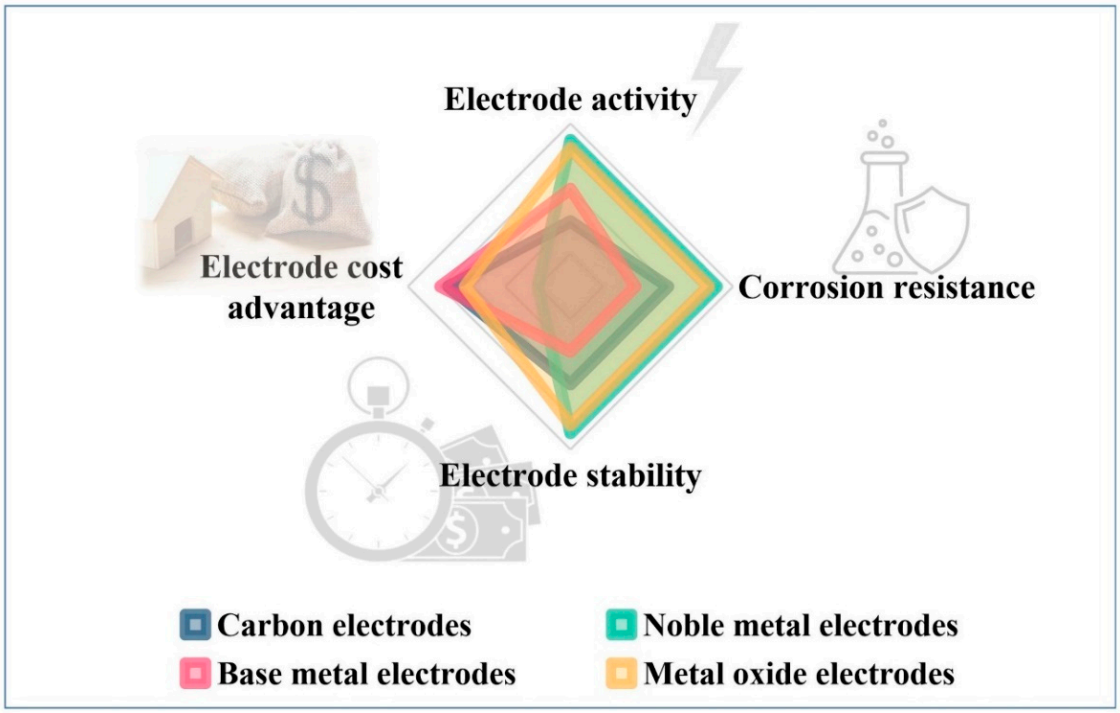

Figure 2. Comparison of the main characteristics of various electrodes that are suitable for organic wastewater treatment.

There are numerous types of metal oxide electrodes, but they can mainly be categorized into the following two groups: single-metal-oxide electrodes [65–67] (represented by lead oxide electrode), and metal oxide composite electrodes [68–70] (various types, such as RuO_2-IrO_2 electrodes and SnO_2-Sb_2O_5 electrodes). These electrodes have been

widely used in the treatment of organic wastewater. This work summarizes the types and hazards of organic wastewater, highlights the significant advantages of using metal oxide electrodes in the electrocatalytic oxidation treatment of organic wastewater, and introduces the primary preparation methods for metal oxide electrodes, as well as the application of single-metal-oxide and metal oxide composite electrodes in organic wastewater treatment. The information presented in this paper will be useful to researchers and practitioners working in the field of wastewater treatment.

2. Preparation of Metal Oxide Electrodes: Fundamentals, Challenges, and Progress

2.1. Fundamentals of Preparing Metal Oxide Electrode Materials

Based on the composition of the electrodes, metal oxide electrodes can be divided into two main types: pure metal oxide electrodes and surface-modified metal oxide electrodes. Pure metal oxide electrodes often possess high degrees of hardness and brittleness. However, surface-modified metal oxide electrodes can demonstrate varying properties, including differences in conductivity and the number of catalytic active sites, which are dependent on the specific metal oxide used and the applied modification technique. In certain applications, surface-modified metal oxide electrodes have attracted more attention due to the potentially superior characteristics they offer. These types of electrodes are also the main subject of the subsequent discussion. The preparation of metal oxide electrode materials typically involves the following elements. First, the selection of the preparation method, whereby the appropriate preparation method is selected based on the desired electrode morphology and application scenario. Second, the selection of raw materials: suitable metal ion sources and other additives (such as stabilizers, surfactants, etc.) are used. Third, the control of the reaction conditions, which involves regulating the characteristics of the prepared material through controlling the temperature, reaction time, and other parameters. Among these processes, the choice of preparation method has the greatest influence on the preparation of the metal oxide electrode, because it has a direct impact on the morphology, structure, and physical and chemical properties of the materials. The main methods for preparing metal oxide electrodes include: the thermal decomposition method [71–73]; the chemical vapor deposition (CVD) method [74–76]; the electrochemical deposition method [77,78]; the sol–gel method [79–81]; the hydrothermal method [82,83], and the physical vapor deposition method [84,85]. Figure 3 lists the basic processes and characteristics of these preparation methods. These include: (1) the thermal decomposition method. As shown in Figure 3a, when using this method, metal ions are dissolved in organic matter/other solutions, coated on the surface of a good conductor (substrate electrode), and decomposed by high temperatures to obtain metal oxide electrodes. The preparation method is simple and fast, but it also has some drawbacks, such as the need for high temperatures, high vacuum conditions, and organic residues. (2) The chemical deposition method. As shown in Figure 3b, the most typical method is chemical vapor deposition (CVD), where metal ions are reduced to metal atoms and deposited on the substrate electrode surface. These atoms are then oxidized to produce a metal oxide electrode. This preparation method can produce high-quality electrodes, has simple preparation conditions, and allows for good control. (3) The electrochemical deposition method. As shown in Figure 3c, the electrochemical deposition method electrolyzes metal ions in a solution, reduces them into metal atoms, and deposits them on the substrate electrode's surface, finally obtaining a metal oxide electrode. This method has the advantages of simple preparation conditions, good control, and the ability to produce a uniform thin-film electrode. (4) The sol–gel method. As shown in Figure 3d, metal organic salt and a solvent are mixed to form a sol solution (with a catalyst added), and metal oxide materials are obtained through drying and calcination. This method can produce electrode materials with a large specific surface area, and the preparation process is relatively simple. However, some residual additives used in the process may affect the electrode's performance. (5) The hydrothermal method. As shown in Figure 3e, by subjecting a mixture of the appropriate metal ion sources to high temperatures and pressures, a metal oxide electrode material is

obtained after a period of time. The method makes it easy to control the shape and size of the product. However, this method has stringent requirements in terms of the equipment and operating conditions. (6) The physical vapor deposition (PVD) method. As shown in Figure 3f, utilizing physical vapor deposition technology, metals or metal oxides are evaporated and then deposited on the substrate electrode's surface, forming a uniform oxide film. Metal oxide electrodes prepared using this physical deposition method exhibit high uniformity and controllable thickness. However, this method is associated with higher costs.

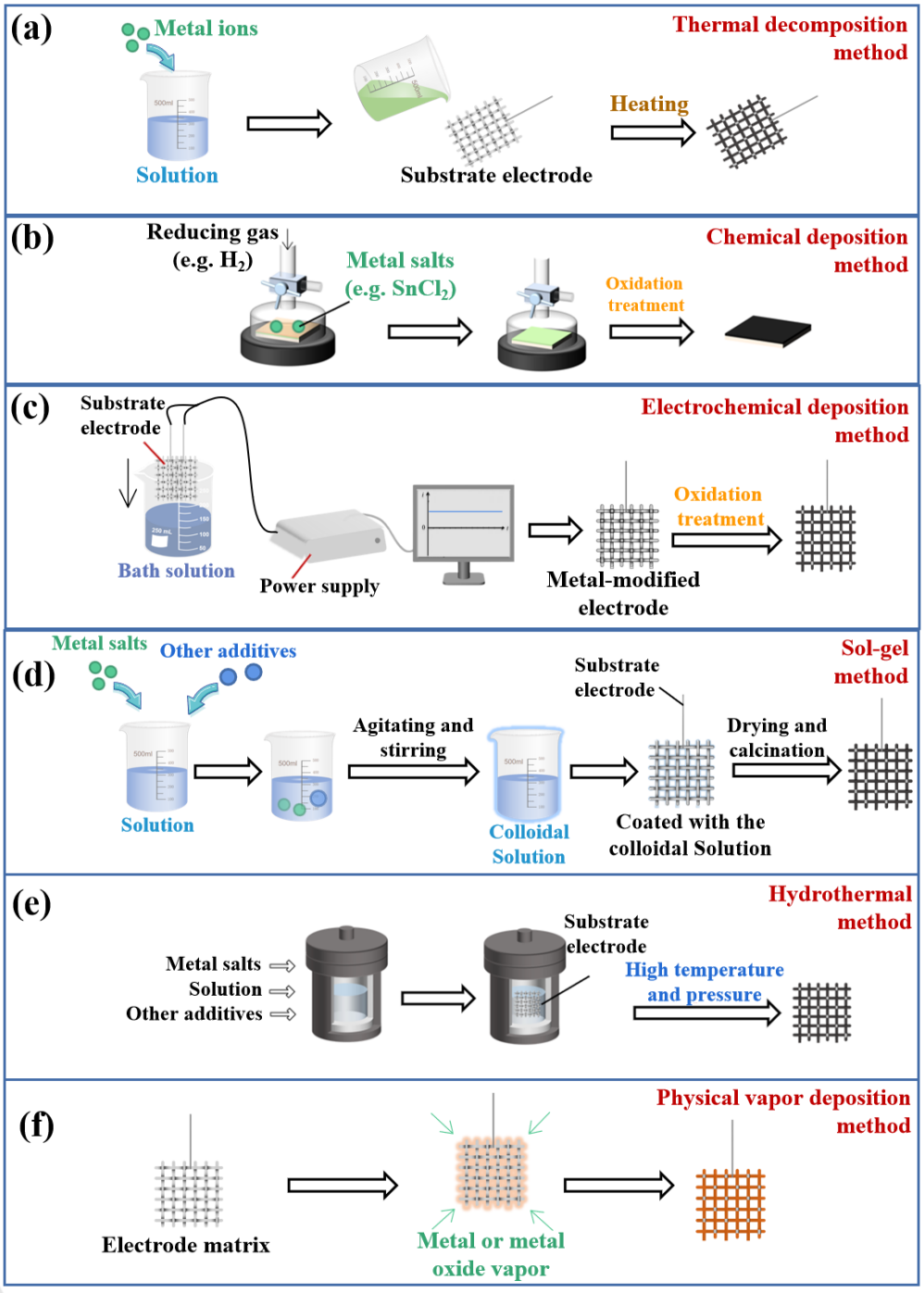

Figure 3. Main metal oxide electrode preparation methods. Thermal decomposition method (**a**); chemical deposition method (**b**); electrochemical deposition method (**c**); sol–gel method (**d**); hydrothermal method (**e**); physical vapor deposition method (**f**).

2.2. Challenges and Research Progress in the Preparation of Metal Oxide Electrodes

Despite its many advantages in practical applications, there are still some challenges in the preparation of metal oxide electrodes. One of the main issues is that some manufacturing processes, such as physical vapor deposition and chemical vapor deposition, require high standards for equipment and production environments, which may not be feasible for all research or industrial settings. Moreover, at the microscopic scale, effectively controlling the size, morphology, and distribution of metal oxide particles remains a persistent challenge, especially when scaling up to large-scale industrial electrodes. Although enhancing the specific surface area of the metal oxide layer benefits the progression of electrocatalytic oxidation reactions, it may also exacerbate the problems of layer shedding and corrosion in the oxide layer.

Electrochemical deposition is a relatively cost-effective method for electrode fabrication, which offers easy control over the reaction process. Guo and colleagues [86] managed to effectively control the surface morphology and structure of PbO_2 electrodes by adjusting the power supply parameters, including the pulse frequency, duty ratio, average current density, and electrodeposition time. Using this approach, they successfully fabricated porous titanium-based PbO_2 electrodes that possess a large number of active sites. By adjusting the elemental composition of the electrode surface, the grain size on the microscale and the actual electrochemical active area can be effectively controlled. Wang et al. [87] fabricated PbO_2 electrodes doped with various rare earth elements using the electrochemical deposition method, which significantly improved the degradation effect of the metal oxide electrodes on p-nitrophenol; the pseudo-first-order kinetic constant increased from 0.19 to 0.41.

Titanium-based metal oxide electrode deposition technology currently faces challenges such as poor coating quality, a weak bonding ability between the coating and substrate, insufficient research on electrodeposition kinetics theory, and limited industrial applications. In response, Wu et al. [88] systematically studied iridium oxide electrodeposition preparation technology, covering the formulation of the deposition solution, substrate selection and treatment, the electrodeposition method, and the deposition time. They proposed several strategies for tackling the issue of coating quality, including optimizing the electrodeposition, combining multi-deposition processes, and co-deposition with metals such as tantalum and lanthanum. In related research, Zhou et al. [89] compared common preparation methods for lead dioxide and examined the electrochemical oxidation performance of lead dioxide in wastewater containing various contaminants. Their work provided a comprehensive reference for operating conditions and offered valuable insights for researchers in the field. Meanwhile, Guo et al. [49] addressed the key points and difficulties in the preparation of titanium sub-oxide electrodes. They suggested future research directions including process optimization, material modification, and application expansion.

Overall, studying the optimization methods and theory of metal oxide electrode preparation technology is of significant value for improving wastewater treatment efficiency and developing more advanced materials.

3. Single-Metal-Oxide Electrodes: Applications and Enhancements in Organic Wastewater Treatment

3.1. Application in Organic Wastewater Treatment

Metal oxide electrodes are crucial electrocatalytic materials that exhibit good chemical stability, electrochemical activity, and photocatalytic performance. Single-metal-oxide electrodes are those composed of a single type of metal oxide, such as lead oxide, copper oxide, and titanium sub-oxides. Each of these electrodes plays a different role in the treatment of organic wastewater. Table 1 presents several typical single-metal-oxide electrodes, along with their advantages and drawbacks. The lead oxide electrode, with its excellent performance, simple preparation process, and strong electrocatalytic oxidation abilities, is highly favored for organic wastewater treatment in the current industrial applications.

Table 1. Typical metal oxide electrodes and their main advantages and drawbacks.

Electrode Types	Main Advantages	Main Drawbacks	Refs.
Lead oxide electrodes (PbO_2 electrode, etc.)	Lead oxide electrodes have good electrical conductivity, corrosion resistance, and catalytic activity, and their preparation cost is relatively low.	Lead oxide electrodes have the potential to gradually release lead ions into the water, which can lead to heavy metal contamination.	[90–93]
Titanium sub-oxide electrodes (Ti_4O_7 and Ti_5O_9 electrodes, etc.)	They have good electrocatalytic activity and stability, and their potential toxicity is very low.	The manufacturing cost of the electrode is relatively high, and it is easily deactivated by some external pollutants in practical applications.	[49,94,95]
Electrodes based on copper oxides	Copper oxides demonstrate remarkable catalytic activity, providing an effective solution to the degradation of some resistant organic substances, particularly antibiotics.	Compared with lead oxide electrodes, their preparation costs are relatively high and the preparation technology is complex.	[96,97]
Electrodes based on iron oxides	Iron oxides can enhance the electrode's capacity to degrade organic wastewater. Furthermore, they can elevate the electrode's oxygen evolution potential and reduce the electrode's inherent resistance in certain application environments.	Iron oxides are susceptible to corrosion in highly acidic environments.	[98–100]

3.2. Optimization Strategy Based on Adding Intermediate Layers

Our previous research [86,101,102] confirmed that metal oxide electrodes, represented by lead oxide electrodes, possess strong cost-effectiveness advantages, and exhibit impressive performance in treating various types of organic wastewater. However, there are potential risks associated with the dissolution of toxic lead elements into the solution as lead ions, which potentially reduce the lifespan of the electrode and lead to secondary pollution.

The electrochemically active modified layer on the surface of a single-metal-oxide electrode is less stable and may release heavy metal ions into the water. Adding an intermediate layer between the surface-active layer and the substrate can effectively improve the electrode's stability and can also potentially enhance the electrocatalytic performance. As shown in Figure 4a,b, Man et al. [92] first employed electrophoretic deposition to prepare a highly conductive MXene material as an intermediate layer for the fabrication of a novel PbO_2 electrode. The electrochemical performance of the PbO_2 electrode was significantly improved by the $Ti_3C_2T_x$ interlayer. The charge-transfer resistance was reduced by nearly 90%, the internal active site increased by 5.21 times, and the ability to generate ·OH radicals increased by 4.07 times compared to the control anode. The new electrode could achieve the almost complete degradation of basic fuchsin within three hours. Similarly, Dai et al. [93] prepared a novel PbO_2 electrode (GNP-PbO_2) by adding graphene nanosheets (GNP) as an intermediate layer to the β-PbO_2 active layer and used it to degrade the antibiotic enrofloxacin (ENO); Figure 4c shows the electrochemical treatment reactor. The GNP intermediate layer increased the oxygen evolution potential of the PbO_2 electrode (2.05 V vs. SCE), which was conducive to inhibiting oxygen precipitation and promoting the production of ·OH, indicating that GNP-PbO_2 is a promising material for the treatment of organic wastewater. The lifespan of such active electrodes is often limited, especially when dealing with wastewater with complex compositions (for example, some electroplating wastewater contains a lot of organic matter and heavy metal ions). As shown in Figure 4d, Rong et al. [103] prepared a bismuth-doped lead oxide (PbO_2-Bi) electrode for the degradation of organic wastewater using the electrodeposition method; they demonstrated that the new structure of the electrode lasted 2.5 times longer than an ordinary PbO_2 electrode.

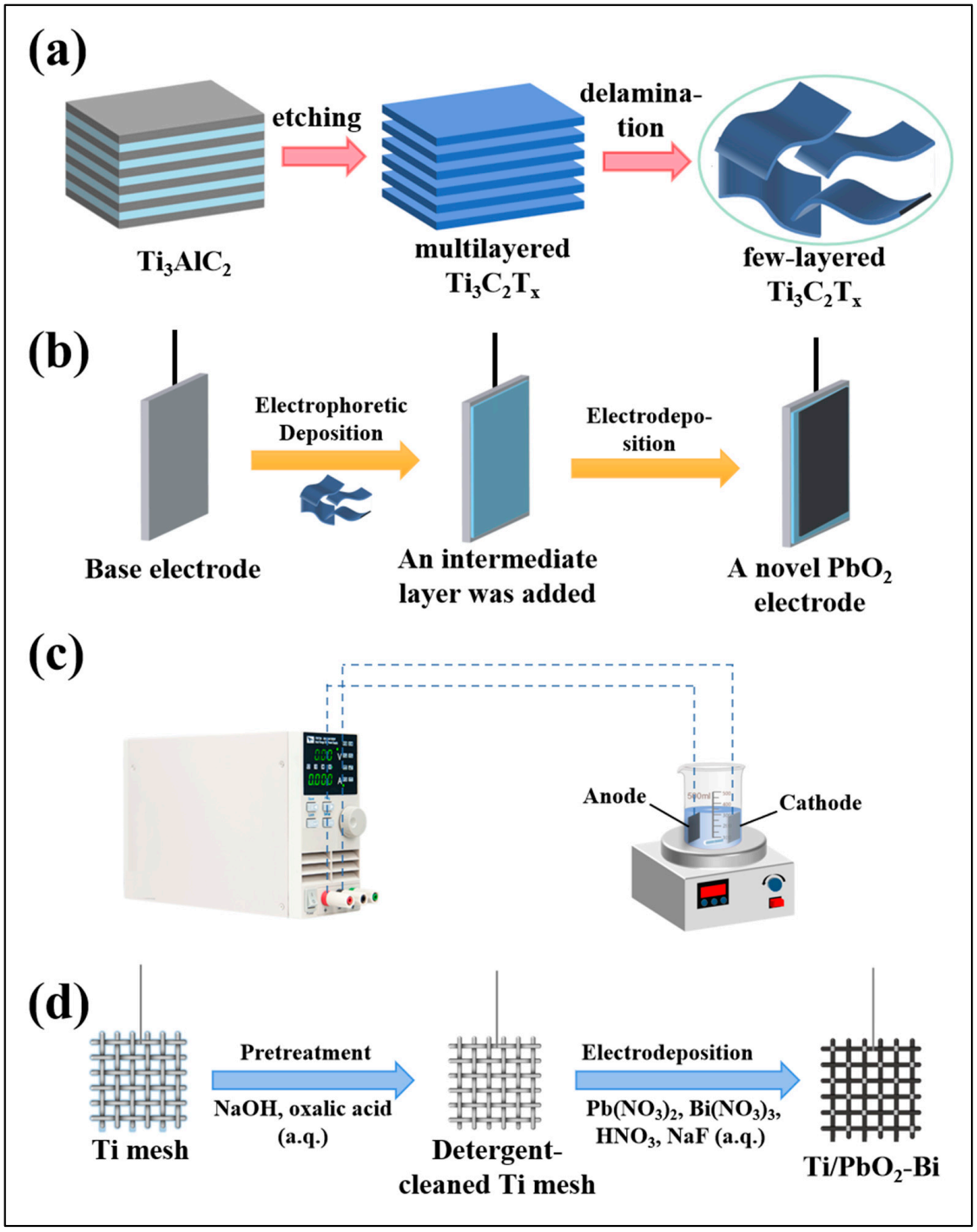

Figure 4. The preparation procedure of few-layered $Ti_3C_2T_x$ (**a**) and a novel PbO_2 electrode with a MXene material as the intermediate layer (**b**) [92]. The electrochemical treatment reactor for a GNP-PbO_2 electrode (**c**) [93]. The fabrication procedure for the PbO_2-Bi electrode (**d**) [103].

The aforementioned studies demonstrated that the introduction of an intermediate layer between the electrode substrate and the active surface layer, or the doping of a single metal oxide, can markedly enhance the stability and longevity of the electrode, which is also always accompanied by an improvement in the catalytic oxidation performance of the electrode material. The above methods can improve the electrochemical performance of the electrode by increasing the number of internal active sites, improving its capacity to generate free radicals, and reducing its charge-transfer resistance.

However, when dealing with organic wastewater with a complex composition (especially electroplating wastewater, which contains a great deal of organic matter and heavy metal ions), involving factors such as chemical corrosion, electrochemical corrosion, and the destruction of the original crystal structure, the lifespan of single-metal-oxide electrodes tends to be limited, and the efficiency of the wastewater treatment can also be relatively low.

4. Metal Oxide Composite Electrodes: Relative Advantages and Progress in Electrochemical Wastewater Treatment

4.1. Relative Advantages

Although single-metal-oxide electrodes have certain advantages in terms of catalytic performance, compared with single-metal-oxide electrodes, metal oxide composite electrodes exhibit the following advantages in the electrochemical treatment of organic wastewater. First, they have better catalytic activity potential. Metal oxide composite electrodes are superimposed with different kinds or forms of metal oxides. This interaction can lead to the enhancement of catalytic activity, which significantly improves the efficiency of the wastewater treatment. They also exhibit better corrosion resistance; based on a specific composition of multiple metal oxides, the corrosion resistance of electrode materials can be improved by appropriate design choices, which allow them to function for longer in acid-based environments. Additionally, they have lower use costs; metal oxide composite electrodes exhibit higher catalytic activity and more lasting stability, which can reduce the frequency of electrode replacement and regeneration, thus reducing the use cost. Therefore, compared with single-metal-oxide electrodes, metal oxide composite electrodes have better catalytic activity, better corrosion resistance, a wide application range, and strong stability in the electrochemical treatment of organic wastewater with complex compositions. Like single-metal-oxide electrodes, metal oxide composites are often loaded on a substrate with excellent electrical conductivity and mechanical strength (such as a Ti electrode) rather than used alone; this not only makes the electrode more stable and reduces its likelihood of fracturing or succumbing to fatigue and other phenomena, but also effectively reduces the electrode production cost.

At present, the main metal oxide composite electrodes are as follows: the Ti/SnO_2-Sb_2O_5 electrode, the Ti/RuO_2-IrO_2 electrode, the Ti/PbO_2-SnO_2 electrode, the Ti/IrO_2-TiO_2 electrode, the Ti/SnO_2-TiO_2 electrode, etc. Their advantages and drawbacks are shown in Table 2. It can be seen that, compared with single-metal-oxide electrodes (such as PbO_2 electrodes), metal oxide composite electrodes generally have the following advantages: (1) high electrochemical activity. These electrodes possess exceptional reactivity on their surfaces, enabling them to catalyze reactions at low potentials. (2) High electrochemical stability: this kind of electrode material generally has good corrosion resistance, allowing for stable operation in a variety of corrosive media, including acidic and alkaline media, fluorine-containing compounds, and chlorides. (3) A long service life. Due to their ability to withstand higher current densities and their superior chemical stability, these types of electrodes can often be used for extended periods without requiring replacement.

Table 2. Typical metal oxide composite electrodes and their advantages and drawbacks.

Electrode Types	Main Advantages	Main Drawbacks	Refs.
Ti/SnO_2-Sb_2O_5 electrode	High catalytic activity, excellent corrosion resistance, and stable electrochemical performance.	High price and brittleness.	[104–106]
Ti/RuO_2-IrO_2 electrode	High catalytic activity, excellent corrosion resistance, stable electrochemical performance, and can withstand high current densities.	High preparation costs, and the treatment effect of some organic wastewater types is not good.	[107]

Table 2. *Cont.*

Electrode Types	Main Advantages	Main Drawbacks	Refs.
PbO_2-SnO_2 electrode	High catalytic activity, good corrosion resistance, and a long service life.	High preparation costs, and the treatment effect of some organic wastewater types is not good.	[108–110]
ZnO/CuO electrode	High catalytic activity, good chemical stability, and low manufacturing costs.	The requirements in terms of water quality are relatively high (e.g., ion content, pH value).	[111]

4.2. Progress in the Design of Metal Oxide Composite Electrodes

The combination of various types of metal oxides can effectively improve the electrocatalytic activity of the electrode. As shown in Figure 5a, Sun et al. [112] prepared a CuO-SnO_2-SbO_x electrode with Ti as the base. The experimental results show that the prepared composite electrode exhibits high catalytic degradation activity and an oxygen evolution potential that is 0.4 V higher than that of the widely used Ti/SnO_2-SbO_x electrode. The complete mineralization of toxic p-nitrophenol can be achieved within four hours. Man et al. [113] used a hydrothermal method to add a three-dimensional Sb-doped SnO_2 nanoflower intermediate layer onto a Ti substrate; the corresponding preparation process is shown in Figure 5b. The electrode, which has a three-dimensional structure, exhibits excellent electrical conductivity. Moreover, the preparation method employed in this study avoids environmental pollution and the short electrode lifespan associated with traditional thermal decomposition methods. This was demonstrated through an accelerated lifetime test. Compared with traditional PbO_2 anodes, the novel electrode material has a tighter electrode surface with a high specific surface area, and it can effectively degrade three typical organic compounds: methylene blue (MB), norfloxacin (NOR), and p-dihydroxybenzene (p-DHB). In addition, the stability and safety of the anode are improved by this three-dimensional interlayer, and its life is extended by 2.70 times. This study provides a useful method for preparing PbO_2 anodes with high catalytic activity and good stability for wastewater treatment. The number of active sites on the surface of the electrodes is crucial for the electrocatalytic oxidation efficiency, while methods to enhance the electrochemical activity of metal oxide electrodes are not limited to modifications of the electrodes themselves. Shao et al. [91] constructed a magnetic assembly electrode (MAE) system based on magnetic carbon nanotubes (CNTs) on the surface of metal oxide electrodes, as shown in Figure 5c. This method effectively increased the number of active sites and improved the efficiency of the organic wastewater treatment. Through systematic research, Shao et al. filled the gaps in the understanding of the mechanism of MAE. The use of MAE to increase the number of active sites on the electrode surface was also validated by Zhang [114] et al.

Metal oxide composite electrodes are composed of a variety of metal oxides. First, metal oxide composite electrodes have high catalytic activity because they have abundant electronic states and surface-active sites, which can adsorb and activate reactive substances to improve the reaction rate and efficiency. Additionally, the catalytic performance of the metal oxide composite electrode can be optimized and upgraded by adjusting its material structure and composition to meet the needs of different reactions. Moreover, the metal oxide composite electrode can be effectively compensated and stabilized by its compatibility with the substrate material, thus improving its reliability and lifespan in long-term operations. In summary, metal oxide composite electrodes are excellent materials with good catalytic activity, electrode stability, and corrosion resistance. They have a wide range of application prospects in the field of the electrocatalytic oxidation of organic wastewater.

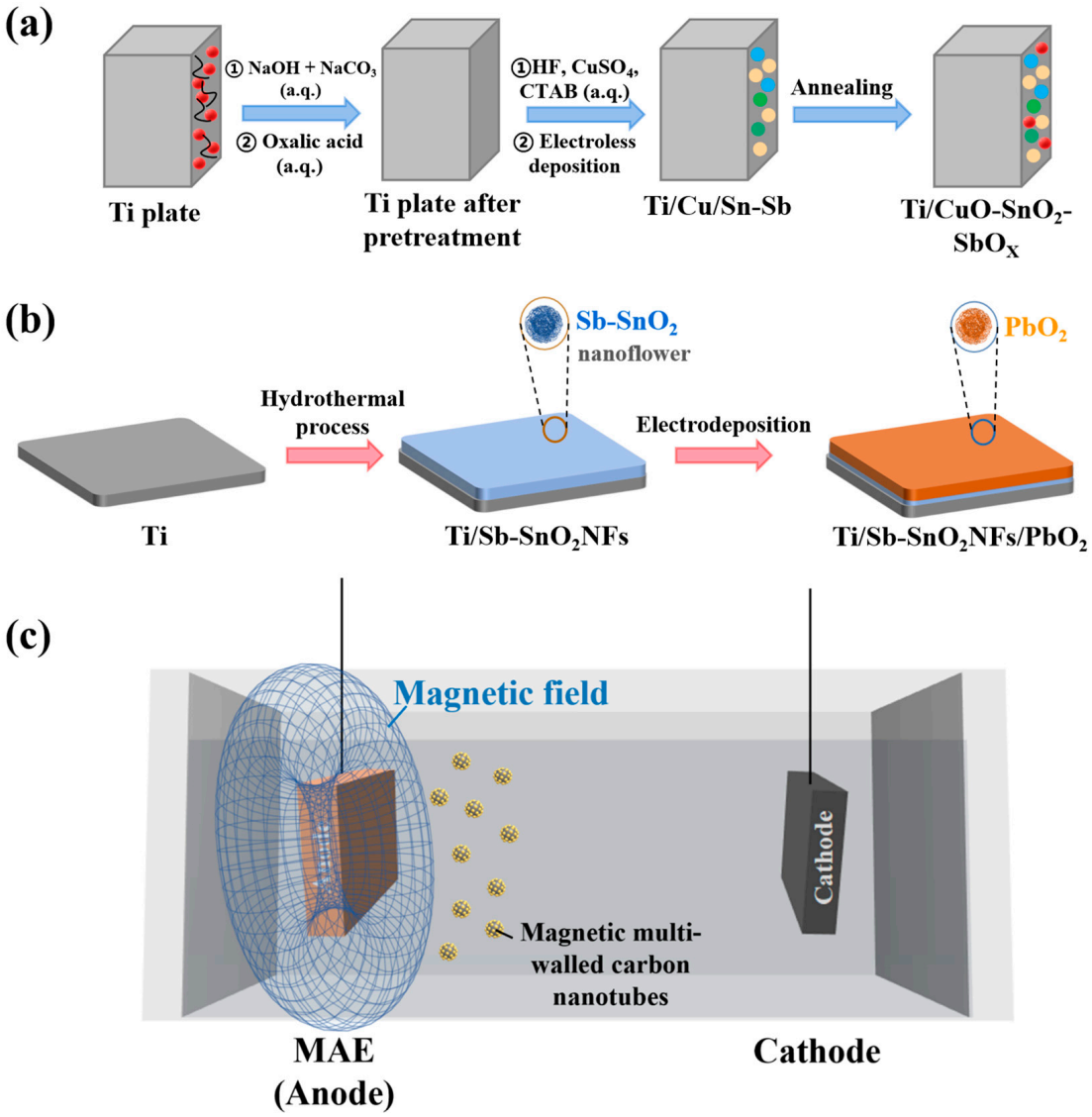

Figure 5. The preparation procedure for Ti/CuO-SnO_2–SbO_X electrodes (**a**) [112]. The preparation procedure for the Ti/Sb-SnO_2 NFs/PbO_2 electrode (**b**) [113]. A schematic diagram of the MAE system (**c**) [91].

5. Summary and Prospects

5.1. Summary

This paper systematically explored the detrimental impacts of organic wastewater and presented an overview of the main wastewater treatment technologies that are currently in use. Special emphasis was placed on the application of electrocatalytic oxidation technology in this domain, with a particular focus on the benefits associated with the use of metal oxide electrodes. The main methods for preparing metal oxide electrodes were explained in detail, including physical, chemical, and electrochemical approaches. Additionally, the primary classifications and characteristics of single-metal-oxide electrodes and metal oxide composite electrodes were introduced, along with the related research and their applications in the field of organic wastewater treatment. From a comprehensive review of the literature, it can be observed that the traditional preparation process for single-metal-oxide electrodes is relatively simple. However, it tends to be constrained by factors such as catalytic activity and the electrode's lifespan. Current research on this type of electrode is primarily evolving towards the use of composite materials. The preparation process for metal oxide composite electrodes, while more complex compared to that for traditional

single-metal-oxide electrodes, often results in higher catalytic activity and relatively low usage costs. Based on the descriptions provided in this paper, it can be concluded that metal oxide electrodes have broad application potential in the electrocatalytic oxidation treatment of organic wastewater. However, it is still necessary to further strengthen research in areas such as electrode manufacturing costs, catalytic degradation efficiency, and electrodes' service life. It is essential to fit the appropriate electrode and degradation program to the actual situation, in order to effectively improve the treatment efficiency and quality of organic wastewater.

5.2. Prospects

5.2.1. Challenges and Potential Solutions in Using Electrocatalytic Oxidation Technology for Organic Wastewater Treatment

Compared with traditional wastewater treatment methods, electrocatalytic oxidation technology has advantages including high removal efficiency, short treatment times, simple operations, and no need for the addition of chemical agents, thus avoiding secondary pollution. Therefore, electrocatalytic oxidation technology has a wide range of potential applications in organic wastewater treatment. However, the electrocatalytic oxidation system centered around metal oxide electrodes still presents numerous challenges, primarily in the following areas. First, cost-effectiveness is still lacking; high power consumption in electrocatalytic oxidation treatment leads to elevated costs when treating organic wastewater using this method. Second, its catalytic capabilities are insufficient. The application scope of the catalytic oxidation method is limited; it can only treat organic wastewater of a certain concentration, making it challenging to handle large volumes of high-concentration wastewater. Third, the material stability needs improvement; the presence of excessive impurities in the wastewater can easily lead to electrode failure, necessitating the frequent cleaning and replacement of the electrodes. Fourth, safety issues; due to the inherent properties of the materials, traditional titanium-based metal oxide electrodes display a phenomenon of surface metal element dissolution during usage. This phenomenon persists throughout the electrode's lifecycle, causing varying degrees of heavy metal secondary pollution risks. Intermediate products are compounds formed during the degradation process before complete mineralization (i.e., the conversion to CO_2 and H_2O) occurs. Depending on the nature of the pollutants being treated and the specific conditions of the electrochemical process, these intermediates can sometimes be more toxic or harmful than the original pollutants, which may pose safety concerns.

In view of the shortcomings of electrocatalytic oxidation technology in organic wastewater treatment, the following solutions may be adopted in the future. The development of new power generation technology is anticipated: electrocatalytic oxidation technology consumes a large amount of electric energy and has high costs, and it is possible to develop cheaper power generation technology in the future, which will directly reduce the use cost of electrocatalytic oxidation technology. Additionally, we should consider the integration of various wastewater treatment technologies; electrocatalytic oxidation technology can be combined with other wastewater treatment technologies, such as biodegradation, membrane separation, etc., to form a more efficient and comprehensive wastewater treatment program. Furthermore, the electrode materials and structure can be optimized. The electrodes used in the existing electrocatalytic oxidation technology are often prone to deactivation. In the future, new materials or structures may be used to manufacture electrodes to improve the stability and efficiency of the electrocatalytic oxidation reaction. In order to mitigate or prevent the formation of harmful intermediates in the electrochemical catalytic oxidation process of wastewater treatment, the following strategies can be adopted: the optimization of process conditions, the real-time monitoring of toxic substances, new post-treatment steps for toxic by-products, the development and use of more advanced electrode materials, etc.

Metal oxide electrodes have been widely used as active electrodes in the electrocatalytic oxidation treatment of organic wastewater due to their excellent properties. However,

these materials also have some drawbacks, including high preparation costs; the cost of preparing metal oxide electrodes is high, resulting in a relatively high application cost. They are also susceptible to surface contamination: metal oxide electrodes often need to have full contact with sewage/organic molecules to exhibit their electrocatalytic oxidation capabilities, but the surfaces of these electrodes easily absorb pollutants, which may affect the electrocatalytic oxidation process and result in reduced efficiency for organic wastewater treatment. Moreover, the electrochemical active layer is prone to detachment and damage: external forces or chemical actions, among other factors, can cause the electrode's surface to peel, become damaged, or fail. Such detachment or damage directly affects the performance and service life of the electrode.

To address the shortcomings of metal oxide electrodes, the following methods can be adopted: reducing the cost of electrodes by developing low-cost and easy-to-prepare metal oxide composite electrode materials to replace existing high-value metal oxides. Additionally, the cost of preparation can be reduced by improving the preparation technology and optimizing the production workflow. Electrode maintenance technology can also be optimized. Chemical or physical methods can be used to treat the electrode surface, such as surface oxidation treatment and high-temperature calcination, to reduce or eliminate surface contamination and improve electrochemical catalytic efficiency. Contaminants deposited on the electrode surface can also be removed by reversing the electrode (switching the polarity of the two electrodes). Additionally, electrode lifespans can be improved; the stability and durability of the electrode can be enhanced by improving the electrode structure and strengthening the bond between the electrochemically active layer and the electrode base (such as by introducing an intermediate layer). In conclusion, effectively overcoming the shortcomings of metal oxide electrodes and improving the performance and application value of electrocatalytic oxidation for organic wastewater treatment can be achieved by optimizing the material preparation processes, surface treatment methods, and electrode structure design.

5.2.2. The Possibility of Undertaking Electrocatalytic Oxidation Using Renewable Energy

Carbon peak and carbon neutrality are both important strategic goals in addressing global climate change. Electrocatalytic oxidation technology requires electrical energy to operate. If this electricity comes from coal-fired or other fossil-fuel-fired power plants, significant carbon emissions are generated in the process of treating the wastewater. However, if this electrical energy comes from renewable sources, such as solar or wind power, then electrocatalytic oxidation technology will have significantly lower carbon emissions during operation. Therefore, by changing the power source, electrocatalytic oxidation technology can have an impact on carbon peaking and carbon neutralization.

Electrocatalytic oxidation technology can effectively treat organic wastewater and convert organic matter into harmless substances such as water and carbon dioxide. Although the process produces carbon dioxide, electrocatalytic oxidation produces far less carbon dioxide than other treatment methods, such as incineration. In addition, if high-efficiency electrocatalytic oxidation technology is adopted, when the same amount of organic wastewater is treated, the amounts of electrical energy required and carbon dioxide generated will also be reduced, which is conducive to carbon peaking and carbon neutrality.

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