Mini Review

BIOMEDICAL Journal of Scientific & Technical Research

ISSN: 2574 -1241

DOI: 10.26717/BJSTR.2024.58.009143

Research Progress on the Electrocatalytic Degradation of Organic Pollutants by PbO_2-Based Electrode

Yue Yin, Wenlong Liu* and Chong Zhang

College of Chemistry and Chemical Engineering, Henan Polytechnic University, Jiaozuo 454003, China

***Corresponding author:** Wenlong Liu, College of Chemistry and Chemical Engineering, Henan Polytechnic University, Jiaozuo 454003, China

ARTICLE INFO

Received: August 12, 2024

Published: August 23, 2024

Citation: Yue Yin, Wenlong Liu and Chong Zhang. Research Progress on the Electrocatalytic Degradation of Organic Pollutants by PbO_2-Based Electrode. Biomed J Sci & Tech Res 58(3)-2024. BJSTR. MS.ID.009143.

ABSTRACT

As an inactive electrode, PbO_2-based electrode can produce abundant hydroxyl radicals which are beneficial to the degradation of organic pollutants. In recent years, as a widely used material in electrochemical oxidation technology, more and more PbO_2-based electrodes have attracted much attention due to their high oxygen evolution potential, high catalytic activity and good stability. Presently, most of the articles are devoted to the modification of the electrode. To better understand electrochemical degradation of pollutants from waste water, this review summarizes the research progress of wastewater treatment in recent years.

Keywords: PbO_2 Electrode; Organic Pollutants; Electrochemical Degradation

Abbreviations: MMO: Mixed Metal Oxide; BDD: Boron Doped Diamond; COD: Chemical Oxygen Demand

Introduction

With the acceleration of population growth, industrialization and urbanization, the problem of global water shortage has become increasingly prominent, while water pollution has become increasingly serious, and water quality has declined seriously, which threat human life and health [1-5]. The goal of ecological civilization construction and carbon neutrality have appeal human beings to protect the environment [6]. Under the goal of double carbon, water treatment has received widespread attention, which requires the synergy of pollution reduction and carbon reduction in the field of water treatment [7-10]. For water resources management, many effects have been devoted to improve the current situation [11-13]. In addition, in order to cope with water shortage and pollution problems, some researchers have been working to develop new water treatment technologies including nanofiltration, ultrafiltration and reverse osmosis based on membrane separation technology, photocatalytic water treatment, electrocatalytic water treatment and water treatment technology based on solar energy, which have the advantages of less energy consumption, less pollution and higher purification rate, [5, 14]. Among them, electrocatalytic technology has been welcomed due to its efficient energy conversion, diversity of catalyst design, and environmental friendliness.

Recently, various electrocatalytic oxidation electrode materials have been used for wastewater treatment. Electrocatalytic electrodes are divided into two categories: active electrodes (Pt, IrO_2, RuO_2) and inactive electrodes (SnO_2, PbO_2, BDD), which include noble metals (such as Pt or Mn), carbon and graphite, metal oxide (MO) and mixed metal oxide (MMO), boron doped diamond (BDD) and other composite electrodes [15-17]. Among them, the most commonly used electrodes are BDD, PbO_2 and SnO_2, because they have high oxygen evolution overpotential (OER) and superior electrocatalytic performance during the oxidation of organic pollutants. Previous studies have shown that BDD has high chemical stability, strong anti-pollution ability, and high oxygen evolution overpotential [18]. However, its production cost is relatively high and complex, and its market supply is limited. SnO_2 is also an efficient anode material, but it is usually prepared by thermal deposition method, which has poor stability at high temperature and short service life [19, 20]. In contrast, PbO_2 is considered to be the excellent anode material because of its easy preparation, low cost, high corrosion resistance and hydroxyl radical (•OH) generation ability [21, 22]. To further realize the treatment of wastewater, this paper first discusses the research progress of PbO_2-based electrode on the degradation of different kinds of pollutants. Then, the electrocatalytic oxidation was studied. Finally, some future prospects were put on electrocatalytic water treatment.

Degradation of Different Pollutants by PbO_2-Based Electrode

Azo Dye

The dyes used in the printing and dyeing process would be discharged within wastewater, seriously causing water pollution. Dye wastewater has the characteristics of high chroma, high alkalinity, high content of organic pollutants and large changes in water quality. Moreover, the dyes are chemically stable and difficult to be biodegraded, posing a threat to the environment and human health (Figure 1&2). In the published papers, most are composite electrocatalytic materials used for dye degradation. Sanaa El Aggadi, et al. [23] used iron (III)-doped PbO_2 to decompose phthalocyanine dyes, achieving a degradation effect of 92.7 % under the optimal experimental conditions. Besides, Sanaa El Aggadi, et al. [24] prepared Fe/C doped lead dioxide modified anode for electrocatalytic degradation of reactive yellow 14 dye. Some azo dyes can not only produce aromatic amines during decomposition, but also produce carcinogenic aromatic amines after contact with the skin. Therefore, Yuan Zeng Jin, et al. [25] designed Ti/SnO_2-Sb/α -PbO_2/Fe-β-PbO2-PTFE electrode to degrade methyl orange. In addition, Ce modified Ti-PbO_2 electrode was also used for the degradation of reactive brilliant blue KN-R [26]. Basic dyes of rhodamine B and fuchsin were respectively degraded by MXene ($Ti_3C_2T_x$) modified Ti/PbO_2 as well as cerium and sodium dodecyl benzene sulfonate co-modified Ti/PbO_2 electrode [27, 28]. Moreover, the positive dye methylene blue was also degraded by rare earth element doped PbO_2 [29].

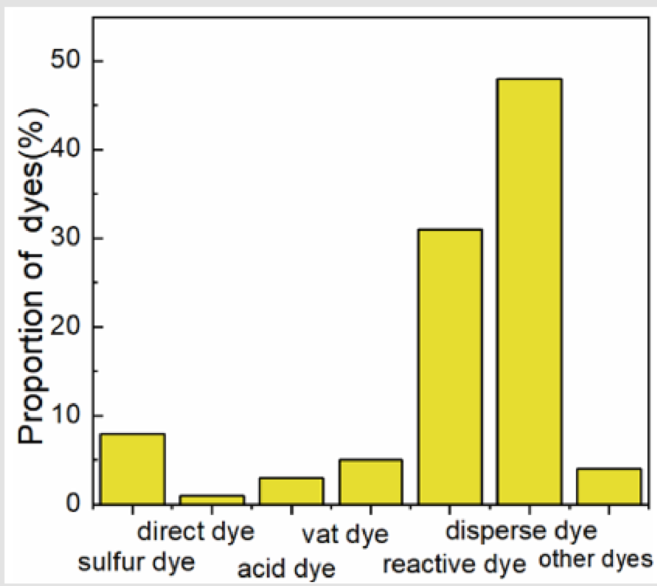

Figure 1: The approximate proportion of different types of dyes.

Phenolic Wastewater

Phenol-containing wastewater mainly comes from coking, refining, petrochemical, gas power plants, plastics, resins, insulation materials, wood preservation, pesticides, chemical industry, papermaking, synthetic fiber and other industries. The corresponding wastewater contains phenols, chlorophenols, phenoxy acids, ammonia, sulfides, cyanide, tar and other substances, showing high chemical oxygen demand (COD), wide pollution range and great harm, which has brought serious harm to human body, water body, fish and crops. It is mainly manifested in toxic effects on humans, harming to water bodies and aquatic organisms, as well as crops [30-35]. Nitrophenol as a toxic and refractory organic matter largely affects the environment, which is easy to be adsorbed and accumulated in soil and water. Importantly, only a very small amount of nitrophenol exists in the atmosphere, mainly through wet deposition into water and soil. C. Borrás et al. studied the oxidation of p-chlorophenol and p-nitrophenol by Bi-PbO_2 [36]. In addition, 3DN-PbO_2 [37] and Ti/Ti_4O_7-PbO_2-Ce [38] were respectively prepared to degrade p-nitrophenol. Besides, trinitrophenol can be efficiently degraded by PbO_2-ZrO_2 anode solid. It has been reported that ZnO/PEG-Co(II)-PbO_2 composite electrode was prepared by anodic electrodeposition [39], and used for electrocatalytic degradation of phenol. Additionally, PbO_2-ILs/Ti electrode has

 DOI: 10.26717/BJSTR.2024.58.009143

been attempted to degrade Bisphenol A [40]. A composite electrode of Ti/SnO_2-Sb/PPy/PbO_2 was also designed and prepared to degrade m-cresol [41]. Polyethylene glycol assisted synthesis of praseodymium doped PbO_2 electrode shows a high catalytic performance for chlorophenol degradation [42].

Elemental Compound Pollution

The treatment of wastewater containing mental elements is an important task for water purification. If poured into water body, wastewater containing mental elements would become serious threat to the environment and human health [43, 44]. Some elements such as arsenic, copper, chromium, cadmium, nickel, zinc, lead, mercury, and manganese have been identified with toxic and non-biodegradable. The mentioned elements easily accumulate in groundwater, surface water, soil, and crops, thereby endangering the health of humans, animals, and plants. Organic compounds containing one or more elements such as fluorine (F), chlorine (Cl), bromine (Br) and iodine (I) can also cause serious environmental pollution [45]. In order to remove these dissolved elements within wastewater, many effects have been devoted. Zhijie Chen, et al. [45] summarized the electrocatalytic degradation of halogenated organic pollutants by PbO2-based anode. The degradation efficiency of perfluorooctanoic acid (PFOA) on Ti/SnO_2-Sb/PbO_2 anode reached 91.1%, while, Ti/SnO_2-Sb_2O_5/PbO_2-PVDF anode showed a higher degradation efficiency for PFOA. The multilayer TNAs/SnO_2/PPy/β-PbO_2 anode has an excellent ability for the electrocatalytic degradation of As (II) [46].

The Principle of Electrochemical Oxidation Technology

There are many kinds of pollutants, whether it is classified according to the source of pollutants, such as herbicides, insecticides, dyes, antibiotics, etc., or the composition of pollutants. The degradation mechanisms of the wastewater are mainly divided into direct electro-oxidation and indirect electro-catalytic oxidation. To oxidizing the organic matter adsorbed on the anode surface by directly losing electrons is called direct oxidation. Due to the high oxygen evolution overpotential of the anode, H_2O and OH- would undergo oxidation reaction on the surface of the anode, which loses electrons and generates •OH [15,47,48]. There is also a process called indirect oxidation. In this process, organic matter is not directly involved in the electron transfer process to the anode. The electrolyte solution produces substances with strong oxidizing ability within the electric field. These substances serve as an intermediate medium to build a bridge for electron exchange between pollutants and electrodes, and then convert macromolecular organic pollutants into low-toxic or even non-toxic small molecules (Scheme 1). According to the composition of the electrolyte, the generated oxygen-containing active species are also different (Eqs. (1) and Eqs(2)). When no other substances are added to the electrolyte, •OH dominates the oxidative degradation of pollutants, and •OH reacts with organic pollutants on the anode surface.

$$PbO_2 + H_2O \rightarrow PbO_2\left(\cdot OH\right) + H^+ + e^- \quad (1)$$

$$PbO_2(\cdot OH) + Organics \rightarrow PbO_2 + CO_2 + H_2O \quad (2)$$

Besides, ClO^- would be generated for Cl^- containing solution. The specific formation process is presented as Eqs.(3), Eqs. (4) and Eqs. (5):

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (3)$$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (4)$$

$$HOCl \leftrightarrow H^+ + OCl^- \quad (5)$$

In addition, O_3 may also be produced in the solution, and the reaction is as follows as Eqs.(6) and Eqs.(7):

$$3H_2O \rightarrow 6H^+ + O_3\left(g\right) + 6e^- \quad (6)$$

$$O_2 + H_2O \rightarrow O_3\left(g\right) + 2H^+ + 2e^- \quad (7)$$

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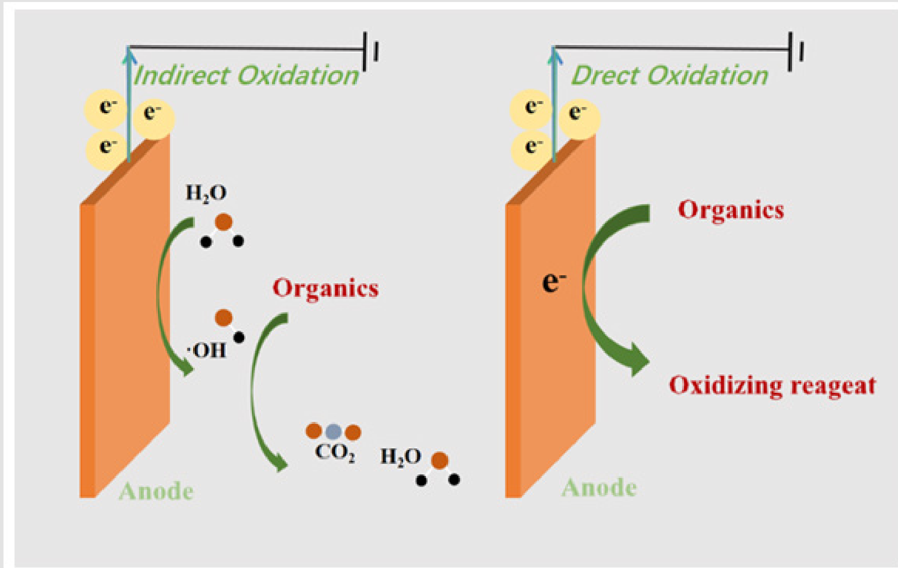

Figure 2: Schematic of direct and indirect oxidation of organics on the surface of PbO_2 anode.

Conclusion

This paper comprehensively reviews the development of PbO_2-based electrodes in recent years. The composite electrodes have the ability to degrade almost common pollutants such as azo dyes in dyes, nitrophenol wastewater in phenolic wastewater, etc. Finally, the mechanism of electrocatalytic oxidation is discussed in detail.

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ISSN: 2574-1241

DOI: 10.26717/BJSTR.2024.58.009143

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