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Research Progress of Chromophoric Dissolved Organic Matter (CDOM) in Landfill Leachate by Electrochemical Method

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Abstract

Over the years, industry, agriculture and other human activities have discharged large amounts of organic pollutants into landfills. Among them, color-resistant dissolved organic pollutants have attracted great attention. In order to achieve the safe discharge of wastewater, a variety of treatment technologies have been used to effectively eliminate CDOM in water. In this paper, the most promising electrochemical oxidation (EO) techniques are reviewed, their basic principles are introduced, and the research progress and application status of electrode materials and electrochemical reactors are introduced. At the same time, the substance composition and detection method of CDOM are discussed. Finally, the performance of EO technology to remove CDOM is summarized. However, we should note that electrode optimization has always been the core of EO technology improvement, and the combination of updated electrode material iteration and treatment process has broad prospects for the wide application of EO technology.

Keywords

Electrochemical Oxidation; Anode Material; Landfill Leachate; CDOM.

1. Introduction

In recent years, with the acceleration of urbanization, a large amount of electronic waste and solid waste have also been gathered in cities with population gathering. Due to the complexity of this part of waste, the disposal requirements have been constantly raised, and how to properly handle urban waste has attracted more and more attention. As a solid waste treatment method, landfill is not only low in cost, but also simple in operation, and is widely used in the disposal of municipal solid waste. However, due to its own water content, natural precipitation and surface runoff, municipal refuse will produce 10%-30% of landfill leachate. This part of leachate is usually dark brown, smelly, and has the characteristics of complex composition, high organic concentration, and high toxicity, thus posing a major threat to human health and the natural environment[1].

At present, there are three main treatment methods for landfill leachate in China: physical and chemical methods, that is, traditional methods such as physical or chemical methods are used to reduce the overall content of landfill leachate; Biochemical method, that is, by cultivating specific microorganisms, using microbial decomposition ability to degrade organic pollutants in landfill leachate; Transfer treatment, including recharge, soil infiltration, incineration and other reduction treatment[2]. The biochemical method is favored by many landfill leachate treatment workers for its advantages of high cost performance, suitable for different status of landfill leachate and stable treatment effect. However, biochemical treatment of landfill leachate also has certain problems and limitations. For example, in the treatment of aging landfill leachate with low biodegradability, due to the stubborn nature of organic carbon in the leachate, the biological process may not achieve good results, so it is generally combined with physical and chemical methods, especially when the effluent quality is strictly controlled,

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membrane filtration methods are generally combined to form a combination process of "pretreatment + biochemical treatment + membrane treatment"[3].

Membrane filtration treatment of landfill leachate generally includes nanofiltration, reverse osmosis, microfiltration, ultrafiltration and other methods. However, nanofiltration membranes with medium separation performance and free retention of macromolecular organic matter through water molecules and inorganic salts are the most widely used. It can effectively separate pollutants from water, and has a high removal rate for humic acids with large molecular weight organic matter. Water molecules or substances similar in size to water molecules can effectively pass through, while those macromolecular substances that are difficult to be degraded will be trapped[4]. In order to meet the subsequent effluent standards, nanofiltration membranes generally retain 10%-30% of concentrated liquid. Compared with the raw liquid, concentrated liquid contains higher concentrations of CODM, ammonia nitrogen and inorganic salts, has lower biodegradability, and is highly toxic. If not treated effectively, it will cause serious damage to the environment and endanger human health[5].

The membrane concentrate formed from biodegraded landfill leachate through nanofiltration has the following characteristics:

(1) Large color, toxicity. The landfill leachate is generally brown with a chroma value greater than 500, and the concentrated liquid after nanofiltration is brown and black with a chroma value generally higher than 1500. With the accumulation of toxic and harmful substances in various parts of the landfill process, the concentration is further increased, resulting in increased toxicity. If the membrane concentrate enters the soil directly, it may cause soil salinization because it contains more inorganic salts.

(2) Poor biodegradability. The biodegradability of landfill leachate itself is not ideal, and some of the landfill leachate that can be treated with biochemical treatment (B/C>0.5) further reduces the B/C of the landfill leachate after biochemical treatment, and the biodegradability becomes worse. Nanofiltration concentrate contains polycyclic aromatic hydrocarbons, various humus organic matter and heavy metal ions, which will affect the biochemical activity of microorganisms.

(3) High concentration of CDOM. The landfill leachate from the landfill plant is subjected to biochemical treatment in the front stage, coupled with the interception effect of nanofiltration membrane, and the COD concentration is very high, generally about 5000mg/L, and some old nanofiltration concentrate can even reach more than 10000mg/L [16]. Due to the previous biochemical treatment, the easily degradable organic matter in the nanofiltration concentrated solution was basically eliminated, and a large number of humic acid and fulvic acid CDOM that were difficult to be treated by biochemical treatment remained.

In recent years, electrochemical oxidation (EO) has become one of the most widely studied water treatment technologies. EO has been observed to be a very powerful technology that oxidizes pollutants by generating in situ electricity to produce strong oxidants, especially hydroxyl radicals (•OH)[6]. Since electrons are the only reagents formed during the treatment process, EO offers an environmentally friendly alternative to some other conventional treatment techniques[7]. In the process of electrochemical catalytic oxidation of organic pollutants, electrode materials are crucial. Different electrode materials will have different electrocatalytic activity, stability, conductivity, corrosion resistance, etc., which will have a direct impact on the removal of pollutants in polluted water. It is crucial to select an electrode material with good performance, good electrolytic activity, high stability, corrosion resistance and electrical conductivity for the degradation of CDOM and the removal of inorganic salt ions such as Na^+, Cl^- and SO_4^{2-} in the waste concentrate[8].

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2. Overview of EO Research

2.1. Basic Principles of EO

The basic principle of electrochemical oxidation (EO) is to apply an electric current to the wastewater to be treated to oxidize and reduce the persistent organic pollutants at the anode and cathode respectively by electrolysis into low-toxic and low-molecular weight substances that are easily biodegradable[9,10]. In some cases, complete degradation of organic matter can be achieved through electrolysis, so that pops can be completely converted into harmless substances, such as CO_2 and H_2O, for wastewater treatment purposes[11]. EO has proven to be a powerful and versatile technology for degrading wastewater pollutants, as they can fully treat both biodegradable and non-biodegradable organic matter, and also have good treatment effect on some inorganic matter. The process is characterized by high versatility, low chemical requirements and high environmental compatibility. This technology can replace traditional processes or be combined with another treatment technology. However, the main disadvantages of EO are high concentration of suspended solids and long treatment time, making it difficult to achieve complete treatment of pollutants[12]. In addition, process shortcomings such as electrode cost and electrode scaling limit the performance of EO in water treatment[13].

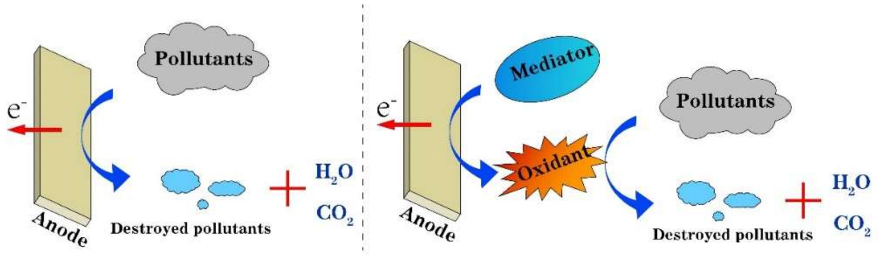

Fig. 1 Basic principles of electrochemistry

Direct oxidation (also known as anodizing) is a simple advanced oxidation process (AOP). This occurs at the electrode surface or through direct electron transfer to the anode. In this process, the persistent organic pollutants are decomposed by oxidation, the molecular chain is broken, and the molecular weight is rapidly decreased. It is mainly due to in situ current generation of powerful oxidants such as hydroxyl radicals. The process takes place in two steps. In the first step, the contaminant is diffused and transported from the bulk solution to the anode surface. In the second step, the contaminants are oxidized on the anode surface, as shown in Fig. 1. The generation of free radicals can be divided into physical adsorption and chemical adsorption because of the different ways of interaction with the electrode. Both mechanisms are involved in a competitive manner, which depends largely on the properties of the anode. The two methods affect the main steps of pollutant oxidation are also different. When the electrode is inactive, such as boron-doped diamond and lead dioxide electrode (PbO_2), physical adsorption is the main mechanism, and OH is physically adsorbed on the electrode surface without forming chemical bonds.

Indirect oxidation refers to the generation of intermediates with strong oxidation on the electrode surface or the interface of solution environment through anodic oxidation reaction under the action of external current, such as hydroxyl radical (·OH), ozone (O_3), hydrogen peroxide (H_2O_2) and other oxidizing particles in wastewater[14], such as $S_2O_8^{2-}$. Cl_2, HClO and $P_2O_8^{4-}$. These oxidized particles react with organic pollutants through electron transfer, double bond addition, ring opening, group removal and other processes to achieve efficient degradation of organic pollutants. Although the existence of these oxidizing particles in solution

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for a long time allows them to oxidize organic pollutants on the electrode surface, while diffusing into the electrolyte solution and reacting with organic pollutants, due to their weaker reactivity than ·OH, they cannot completely degrade persistent organic pollutants, but can only convert them into less toxic intermediate products. Therefore, the presence of a large amount of Cl^- in wastewater will not only produce chlorine active substances such as Cl_2, HCl, Cl^-, but also produce some highly toxic organochlorine by-products such as chlorate and perchlorate[15].

2.2. Factors Affecting EO

EO has many advantages over many traditional technologies for dealing with pollutants. Electrochemical oxidation processes are known for their high environmental performance and minimal waste generation during treatment. Although EO has been hailed as the technology of the future, its use in the industry has been limited due to its environmental compatibility and certain conflicting features . The main problem with electrochemical oxidation technology is that the catalytic activity decreases over time due to anode layer poisoning. Deactivation depends on the nature of the anode and the concentration of organic compounds present in the electrolyte. The use of inert surface materials with weak adsorption properties, such as "boron-doped diamond (BDD)" electrodes, can reduce electrode poisoning[16]. The use of nanomaterial-based electrocatalysts can compensate for these limitations, as they provide a large surface area for enhancing reaction dynamics. Other disadvantages are its high electrode cost and operating costs, the production of unwanted chlorinated compounds, and the need for sufficient conductive effluents. Researchers generally find that removal is most efficient in acidic conditions[17]. The pH value affects the concentration of OH ion formation, oxygen overpotential and the lifetime of the anode, which also determines the EO efficiency. One study found that more OH and higher oxygen overpotential were produced under optimal acidic pH conditions. At this time, the oxygen production reaction is inhibited, thus improving the degradation efficiency. If oxygen is produced at a higher rate in an alkaline solution, it will result in a decrease in OH production. On the other hand, the electrode material may also affect the optimal pH value. For example, at pH 1 and 2, the OEP value of the Ti/Pt anode is about 3.5 V (relative to Ag/AgCl), while for pH 0 and 9, it is about 7.9.

3. Overview of CDOM Research

3.1. CDOM Definition

Dissolved organic matter (DOM) in water is a complex mixture of molecular fragments containing carbon, nitrogen, phosphorus, and sulfur, with molecular weights typically ranging from a few hundred to 100,000Da. Chromophoric DOM(CDOM), also known as yellow substance, colored dissolved organic matter or gelbstoff, is mainly composed of aromatic amino acids, lignin phenols, undefined humus and other substances, with a complex source, composition and molecular structure[18]. There are also significant differences in the composition and characteristics of CDOM under different environmental conditions, as well as significant differences in its bioavailability. CDOM is an important link in the nutrient availability and regeneration process supporting the growth of bacteria and phytoplankton in aquatic ecosystems. The source, composition, degradation and mineralization of CDOM participate in the global carbon cycle and carbon budget, which may lead to greenhouse gas emissions and influence global climate change[19].

3.2. Characterization of CDOM based on UV-VIS Absorption Spectrum

Ultraviolet-visible absorption spectroscopy (UV-Vis) technology is used to analyze the absorption spectrum of CDOM using ultraviolet and visible spectral regions. It makes use of the characteristics of the sample to absorb ultraviolet light, and identifies and quantitatively

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analyzes the organic matter in the sample by measuring the wavelength and absorption intensity of the sample to absorb ultraviolet light[20]. The advantage of ultraviolet absorption spectroscopy is that it has the characteristics of fast, simple, sensitive and cheap, and can be used for rapid and efficient analysis of dissolved organic matter in water. In addition, the ultraviolet absorption spectrum also has a high sensitivity and can identify trace amounts of organic matter. In short, the application and development of ultraviolet absorption spectroscopy in dissolved organic matter in water is very extensive, and has become an important technical means for the analysis of dissolved organic matter in water. With the continuous development of optical technology, the application and development of ultraviolet absorption spectroscopy in the analysis of dissolved organic matter in water will continue to improve[21].

Since CDOM is a mixture of substances from different sources and complex chemical components, it is difficult to quantify CDOM in terms of mass or concentration . Therefore, the amount of CDOM is generally defined by the absorption coefficient of the reference wavelength. The absorption coefficient and DOC concentration of the characteristic wavelength (254, 280, 350, 375 or 440nm) measured by absorption spectroscopy are used to characterize the concentration of CDOM in many studies . CDOM absorption coefficient can often be used to evaluate water pollution and eutrophication. Kowalczuk et al., the absorption coefficient of 350nm represents the concentration of CDOM, and it is concluded that the absorption coefficient of CDOM is the highest at 350nm, and it is believed that 350nm is related to photodegradation.

3.3. CDOM Was Characterized based on Three-dimensional Fluorescence Spectrum

Fluorescence DOM (FDOM) is a component of CDOM that can fluoresce when excited at short wavelengths. Fluorescence spectroscopy, especially EEM technology, has been widely used to characterize the source and composition of CDOM due to its high sensitivity, high information content, low cost, and no influence on samples. Three-dimensional fluorescence spectrum combines fluorescence emission spectrum and fluorescence excitation spectrum to reflect the chemical composition and structural characteristics of CDOM. This technology breaks through the limitation of traditional two-dimensional fluorescence spectra, and can fully present excitation wavelength, emission wavelength and fluorescence intensity in a three-dimensional contour spectral map, thus providing more abundant information, so it is more widely used in the characterization of CDOM[22]. The three-dimensional fluorescence spectrum of CDOM can be obtained by this technology, in which fluorescence intensity and fluorescence peak position can characterize the content and quality of CDOM, and fluorescence peak area and fluorescence peak ratio can characterize the chemical composition and structural characteristics of CDOM[23]. Three-dimensional fluorescence spectra can identify different fluorescent components in CDOM, including humoid, humus, proteoid and amino acid. These fluorescent components are closely related to the source and composition of CDOM, so the source and chemical composition of CDOM can be analyzed in detail through EEMs[24]. Three-dimensional fluorescence spectroscopy has the characteristics of high sensitivity and high flux, so it has a broad application prospect in the analysis of dissolved organic matter in water. Three-dimensional fluorescence spectroscopy can detect dissolved organic matter in water at a trace level, which is of great significance for protecting human health and environmental safety[25].

4. Research Status of Electrodes

In EO technology, the electrode is the most important component, and the material and performance of the electrode directly determine the cost and efficiency of the oxidation process. The oxidation reaction in EO process mainly occurs in the anode, and the catalytic activity and

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stability of the anode are the key factors determining its oxidation capacity and working life[26]. Therefore, most researchers focus on preparing anode materials with high performance, low cost and suitable for practical applications by selecting suitable materials, designing favorable structures and improving preparation processes. The research on EO anodes mainly focuses on dimensional stable metal oxide electrode (DSA) and boron-doped diamond (BDD) electrode.

Further study of DSA electrodes found that doping metal and non-metal elements can further improve the performance of DSA. This is because doping of elements can create surface defects in the electrode coating, such as crystal plane steps and dislocations[27]. These defects in the electrode can increase the number of electron transfer channels, improve the conductivity and catalytic activity of the electrode, accelerate the electrochemical reaction process on the electrode surface, and improve the current efficiency[27]. At the same time, the overpotential of oxygen and chlorine evolution on the modified electrode will increase[28,29]. The doping of elements can also avoid the formation of polymer films on the electrode surface that hinder electron transfer, and ensure that the electrode maintains a high catalytic oxidation activity under long-term operation. Wang et al[30]. doped Ti/SnO2-RuO2 electrodes with different amounts of rare earth Yb elements and found that doping 1.5% Yb increased the oxygen evolution potential and electrocatalytic oxidation activity of the electrodes. The kinetic analysis shows that the degradation process of TOC in coking wastewater is consistent with the first-order kinetic equation. When the current density is 10 mA cm^{-2}, the reaction rate constant is the largest (0.1834 min^{-1}), so this current density is determined to be the optimal current condition. Under these conditions, the COD removal rate of coking wastewater reached 85.06% after 85 min electrolysis. In other studies, Liu et al. used the sol-gel route to synthesize terene oxide coatings doped with different Ru/Ir/Zr molar ratios of RuO_2-IrO_2-ZrO_2, and studied the effects of different amounts of Ru doping on Ti/IrO_2-ZrO_2 electrodes[31]. The analysis of the surface morphology and electrochemical behavior of the electrode shows that with the increase of Ru content, the porosity and cracking of the trioxide coating increase significantly, which improves the electrocatalytic activity of the electrode, but decreases the stability of the electrode. The experimental results show that when Ru content is 21 mol%, the dense coating structure with good electrocatalytic activity and longest service life is obtained.

DSA electrode has been active in the field of electrochemical oxidation of wastewater. The lower production cost and better electrochemical performance make it favored by many researchers. The researchers improved the DSA electrode from the aspects of elemental composition and preparation process, and selected the best operating conditions to improve the wastewater treatment capacity, which fully proved that DSA electrode is an electrode material with excellent electrical properties and has a high application prospect in wastewater treatment.

5. Conclusion

The presence of CDOM in landfill leachate is adversely affecting the health of organisms. Effective treatment techniques are essential to remove CDOM from wastewater to protect the environmental substrate. After years of development, EO has become the most promising wastewater treatment technology. It has shown excellent performance in both the degradation treatment of persistent organic pollutants and the detoxification treatment of highly biotoxic organic matter, and is widely regarded as the leading technology in the field of wastewater treatment. EO typically removes CDOM through direct, indirect, and mediated oxidation by producing in situ oxidizing species.

Up to now, EO has been applied in practical wastewater treatment to some extent, but most of the research is still in the laboratory stage and lacks practical application. In addition, most of the wastewater used in the experiment is single-component wastewater configured in the laboratory, but the actual wastewater is often complex and changeable, affected by a variety of

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factors. Electrode materials play a key role in EO, so current research hotspots will focus on the development of new electrode materials, the design of excellent electrode structures, and the optimization of efficient electrode preparation processes. Elucidating the mechanism of organic oxidation in EO is critical to improving the efficiency of organic degradation in electrochemical systems and will also provide a basis for the implementation of these systems for industrial use. In short, after understanding the mechanism of EO practical application, matrix interaction and the material principle of CDOM, the electrode materials and the mechanism of EO removing CDOM have been extensively studied in this paper. In order to help the actual removal of organic pollutants in the water environment, it is possible to efficiently remove CDOM substances in water.

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