REVIEW ARTICLE

Research progress in the treatment of high salt and high concentration organic industrial wastewater

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Due to the high content of dissolved organic matter in high-salinity organic wastewater, the physicochemical treatment method is costly and difficult to achieve the desired purification effect. The biological treatment method has the advantages of economy, high efficiency, and harmlessness. Therefore, the biological method is currently the mainstream organic wastewater treatment technology. However, it is difficult to achieve high-salt organic wastewater treatment standards with traditional biological and physical-chemical treatment technologies. This article reviewed the research status of high salt and high concentration organic industrial wastewater treatments and suggested that research could be conducted on the optimization selection, biological carrier enhancement, reactor process optimization, and other aspects of high-efficiency anaerobic reactors in combination with current research hotspots to better solve and improve the problems existing in different wastewater processes.

Keywords: biofortification; anaerobic reactor; high salt; high concentration; organic industrial wastewater.

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Introduction

As the pillar industry of China, the chemical and pharmaceutical industry plays a pivotal role in the national economic development. However, it also brings enormous pressure on environmental pollution. The wastewater generated during these production processes has complex chemical composition and significantly varying pH values and are easy to form impact loads. They contain not only high concentration of organic matter (containing organic substances) such as glycerin, medium and low carbon chains, but also large amount of salt contents such as inorganic ions including Cl⁻, SO₄²⁻, Na⁺, Ca²⁺, etc. These types of wastewaters can also be produced in many other industrial production activities such as paper, oil and gas production, mining and mineral processing, pickling, meat processing, seafood processing, *etc*. The amount of highsalinity organic wastewater produced in China counts for 5% of the total wastewater. In addition, the amount of high-salinity organic wastewater is still growing at a rate of 2% every year [1]. Not only does it cause pollution to the environment, but also it poses a threat to water quality, aquatic flora and fauna, and human health. Therefore, the treatment technology research and application for these wastewaters has significant practical significance.

Processing methods

The general conventional wastewater treatment processes mainly include physical, chemical, and biological methods.

1. Physical methods

The commonly used physical treatment methods for wastewater include gravity sedimentation, filtration, and air flotation [2]. The filtration method refers to filtering impurities in wastewater through the porous particle layer of the material to reduce suspended solids in the water. The gravity precipitation method refers to the use of the settleability of particles suspended in water to settle under the action of gravity, thereby achieving solid-liquid separation. Air flotation refers to the method of wrapping the generated microbubbles around the surface of suspended particles and bringing them out of the water surface. These three physical methods not only have simple processes but also are easy to manage. However, they are not suitable for removing soluble components from wastewater and have significant limitations.

2. Chemical methods

Some substances soluble in wastewater can be removed through certain chemical reactions. The main chemical methods include oxidation, electrochemistry, and coagulation. The chemical coagulation method mainly acts on relatively small, suspended solids and colloids in water and precipitates these substances by placing flocculants in wastewater to achieve solid-liquid separation [3]. Chemical oxidation method typically introduces oxidants into wastewater to oxidize some organic compounds, achieving the goal of purifying wastewater [4].

3. Physical and chemical methods

Common physical and chemical methods include ion exchange, membrane separation, and extraction [2]. The ion exchange method is the exchange of ions with water to exchange and eliminate ions from harmful substances, ultimately achieving the goal of purifying water. The principle of extraction method is to achieve the final extraction effect through the difference in solubility of substances in the substance. Usually, it uses substances that cannot be dissolved in water to extract pollutants that are dissolved in water, fully contacts the extractor with wastewater, and uses the different solubility of the extractor in pollutants and water to separate and purify pollutants from water, thereby purifying water. Membrane separation technology utilizes semi permeable membranes to filter molecules for wastewater treatment. Membrane separation technology can also be called reverse osmosis, mainly utilizing the characteristics of semi permeable membranes to achieve the separation of toxic substances from wastewater. In this semi permeable membrane, only water can pass through, and toxic substances in the water will be blocked. Therefore, it can block some dissolved organic compounds and colloidal states in the water, achieving the separation of substances from water and purifying water.

4. Biological methods

By utilizing the biological methods of microorganisms, the organic matter in wastewater is transformed into non-toxic and harmless substances through microbial metabolism, thereby achieving the goal of purifying water quality [5]. Biological methods are mainly divided into two categories including aerobic treatment and anaerobic treatment. The aerobic treatment can be further divided into activated sludge method and biofilm method. The biofilm method refers to the combination of biofilm and wastewater to absorb organic matter in the wastewater through the adsorption of biofilm. The anaerobic wastewater treatment refers to the process of decomposing organic matter in wastewater through the metabolism of anaerobic microorganisms under anaerobic conditions, thereby converting organic matter into methane and carbon dioxide, and achieving the goal of purifying water quality. The advantages of biotechnology in the control of sewage are mainly manifested in the following aspects. First, secondary pollution will not be produced. This advantage is mainly compared to the chemical governance methods of traditional water pollution treatment that is easy to cause secondary pollution in water bodies when using chemical governance technology. It is mainly due to the addition of chemical reagents, and the pharmaceutical agent will bring structural changes to the unpolluted water bodies. Second, biotechnology methods have high efficiency of the treatment. It can quickly remove pollutants such as organic matter, nitrogen, phosphorus, *etc*. in sewage. Third, it has low -cost nature. Due to the strong reproduction ability of microorganisms, it can be taken to the spot, which saves transportation and processing costs. In addition, biotechnology processing sewage can make full use of the organic substances in the sludge by converting it into useful resources such as fertilizer, biological gas, *etc*. to achieve realization resource use.

Because it is difficult to meet the economic and technical requirements of purification treatment using the traditional treatment method to treat high-salt and high concentration organic wastewater, the research of this type of wastewater treatment technology has become one of the hot research projects in environmental protection fields domestically and internationally. Compared with the material chemical method, the biochemical method has the advantages of reliable operation, low operating costs, and convenient operation management, so that the biochemical law can be widely used. Therefore, this article specifically introduced the current status of high-salt and high-concentration organic wastewater biological treatment technology.

Research advance

According to previous studies, the research of biotechnology is still a hot spot. In China, there are a lot of studies in this area. About 40-50% of the research in this field was reported by Chinese authors, which may be related to the confusion in the treatment of high-salt organic production wastewater brought by China's rapid industrial development in recent years. The past high-salt organic wastewater studies can be divided into the following categories including pharmaceutical wastewater [6], phenol wastewater [7], tannery wastewater [8], polycarbonate plant wastewater [9], urban wastewater [10], petrochemical wastewater [11], aquaculture wastewater [12], printing and dyeing wastewater [13], chemical superfine fiber alkali heavy wastewater [14], synthetic olefin plant wastewater [15], sulfonic acid plant wastewater [16], pickle wastewater [17], *etc*. The current research trends on this type of wastewater treatments mainly include highefficiency reactors [18], bio-enhancement [19], and research on other processes [20].

1. Research on high efficiency reactor

Anaerobic digestion can be used for high concentration wastewater treatment, and its main product methane can be used as energy for power generation or heating. With the in-depth study of anaerobic digestion theory, various efficient anaerobic bioreactors have been developed such as anaerobic filters, upflow anaerobic sludge bed (UASB), anaerobic attached membrane expanded bed (AAFEB), and internal circulation (IC) reactors. These new highefficiency anaerobic reactors can ensure high biomass and longer solid retention time (SRT) in the reactor from a structural or operational perspective, greatly increasing the organic load of the reactor volume and shortening the hydraulic retention time (HRT), opening a new field for wastewater treatment. Wei *et al*. used IC anaerobic bioreactor to treat the actual pharmaceutical wastewater [18]. The anaerobic reactor was started by medium temperature fermentation at 35℃ and continuous water intake, and the chemical oxygen demand (COD) removal rate was stable at about 65%. In the

salinity range of 0 to 1.5%, the increase of influent salinity would cause the removal rates of COD and sulfate to decrease to varying degrees, but the final removal effect was still stable, indicating that the IC anaerobic system had a certain tolerance to external salinity. Foglia *et al*. studied the efficiency of the anaerobic membrane bioreactor (AnMBR) in the treatment of high-salt low-load wastewater in the joint sewer [10]. They determined the changes of biotransformation and system stability under different organic loading rate (OLR) and high salinity conditions. In addition, fermented cellulose sludge was added in the process. When the COD concentration of sewage was lower than 100 mg $O₂/L$, the COD removal rate of AnMBR was 83 \pm 1%. The addition of fermented sludge could increase the biogas production, but it would affect the membrane operation. The high saline-alkali condition of 1,500 mg Cl/L adversely affected biogas production without deteriorating membrane operation. Wei *et al*. experimentally studied the robustness and microbial community structure of the UASB reactor in the treatment of hydrochloric phenol wastewater and treated phenol wastewater under salt and non-salt conditions [21]. The reactor was operating stably, and the phenol concentration increased from 100 mg/L to 500 mg/L at 10 g Na⁺/L. The stability of the reactor was impaired at 1,000 mg phenol/L and 10 g Na⁺ /L. Lujan-Facundo *et al*. evaluated the performance of an osmotic membrane bioreactor (OMBR) for the treatment of tannery wastewater [8], while Xu *et al*. reviewed the MBR technology for brine wastewater treatment from the perspective of biology [22].

2. Biofortification

Biofortification technology refers to the introduction of specific microorganisms in traditional biological treatment, increasing the effective concentration and enhancing the degradation ability, to improve the removal rate of organic matter. From the type point of view, biofortification technology belongs to the category of new biotechnology, and the main application scope is water pollution control. The specific application principle is that through the configuration of bacteria, the bacteria play their own special ability such as stronger metabolic function, *etc*., put it into the polluted water source, decompose the pollutants in the sewage with the help of microorganisms and bacteria, realize the transformation of polluted organic matter to non-polluting inorganic matter, and finally achieve the purpose of sewage purification and treatment. Biofortification technology can be applied in high concentration and high salt sewage water and can maintain strong activity. Bio-enhancement includes application research of new biological carriers [23], research and application of salt-tolerant bacteria [24], and the application of microalgae [25], *etc*.

(1) Research on the application of new biological carriers

One of the core technologies for wastewater treatment in biofilm process is carrier. The main habitat for microorganisms is the surface of the carrier, and the physicochemical properties of the carrier can affect the composition and shape of the biofilm, water and gas distribution, oxygen utilization efficiency, and so on. By adding the biological carrier, the symbiotic microbial system of adhesion growth and suspension growth can be constructed and the microbial abundance and diversity in the system can be increased. Since the 1970s, biological carriers have become the main research objects with continuous improvement and research and development. The promotion of the biofilm method has been continuously developed. In recent years, the sewage treatment technology of increasing microbial concentration and optimizing microbial community structure by adding biological carrier has been widely used. Zhou *et al*. discussed a new carrier and their use for fixing biomass to increase the biodegradation of saline organic wastewater [23]. This new carrier was found to have a layered structure and larger surface area, supporting bacterial growth and high salinity tolerance. Compared with the removal efficiency of CODCr with PVA as the carrier only (52.8%), This new carrier could achieve a higher degradation rate of 62.8%. It had been proven to be a good choice for enhancing the

biodegradation efficiency of high salt organic wastewater through biological enhancement. Kok *et al*. compared the treatments of pharmaceutical wastewater using a novel anaerobic biofilm reactor (AnBEMR) with a control anaerobic membrane bioreactor (AnMBR) and found that AnBEMR increased COD removal rate and produced more biogas [19]. Through DNA pyrophosphate sequencing analysis, the bacterial *Elusimicrobia phylum* was only detected in AnBEMR. The abundance of the dominant archaea genus *Methanicoccus* discovered in AnBEMR was relatively high, which might play an important role in the degradation of the main organic pollutant, trimethylamine, in pharmaceutical wastewater. Wen *et al*. reviewed the application of zeolite in saline wastewater and presented research challenges in improving surface properties and reproducibility [26].

(2) Research and application of salt-tolerant bacteria

Li *et al*. combined a kind of highly efficient photosynthetic bacteria - graphene oxide /polyvinylidene fluoride (PSB-GO/PVDF) membrane photobioreactor (MPBR) with a multiphase Fenton fluidized bed and successfully applied it to the treatment of seafood wastewater with extremely high salinity and difficult treatment [27]. The removal efficiencies of COD and NH3-N from MPBR using GO/PVDF membrane were about 95% and 98%, respectively. Hülsen *et al*. used anaerobic purple phototrophic bacteria (PPB) to treat high-salinity wastewater and marine wastewater in a continuous anaerobic infrared photobioreactor [28]. The operation exceeded 372 days. The results showed that PPB could quickly adapt to the treatment of high-salinity wastewater. Golshan *et al*. developed a salt-tolerant microbial consortium in the treatment of salt-containing diphenol A (BPA) wastewater [24].

(3) Research and application of microalgae

Meng *et al*. studied the feasibility of using continuous flow reactor (CFR) to compare bacterial aerobic granular sludge (AGS-CFR) and algae bacterial granular sludge (ABGS-CFR) to treat 1-4% saline-alkali wastewater [29]. It was found that high salinity (1-3%) could promote the growth of algae in ABGS-CFR, which led to the increased removal efficiency for total nitrogen and phosphorus. ABGS-CFR maintained good particle size stability at the salinity of 1-4%, while AGS-CFR gradually decomposed at the salinity of 4%. Church *et al*. proposed that marine microalgae were effective in removing nitrogen and phosphorus from saline wastewater and verified the influence of the type and concentration of salt through experiments [30]. Shi *et al*. studied the pretreatment method for amoxicillin (AMX) - containing brine antibiotic wastewater using the green microalga *Chlorella* isolated from the marine environment [25]. *Chlorella* showed multifunctional carbon source metabolism potential under self-nutrition growth conditions. In the treatment of salt-containing antibiotic wastewater, the combined microalgaebacterial process had shown a great potential to simultaneously remove organic matter and amoxicillin residues. Phong Vo *et al*. studied the potential competition of freshwater microalgae compared with marine microalgae in the treatment of high-salinity wastewater [31].

3. Research on other processes

In addition, studies have been reported on electrochemistry [32], modified ion exchange resins [33], humid air oxidation (WAO) [9], new solar wastewater-energy recovery systems [34], carbon nanotubes (CNTs) based membranephotocatalytic processes [35], Fenton oxidation [36], membrane distillation and crystallization process [37], and nano filtration membrane [38].

Conclusion and recommendations

From the literature review, high-efficiency reactors and biofortification are research hotspots in the field of high-salt organic wastewater treatment. The research and application of biofortification are mostly focused on salt-tolerant bacteria and new biological carriers. Based on the treatment of high salt organic production wastewater, in order to

better solve and improve the problems existing in different wastewater processes, research can be conducted on the optimization selection, biological enhancement, reactor process optimization, and other aspects of high-efficiency anaerobic reactors in combination with current research hotspots. In terms of biological enhancement, research can be conducted on carrier enhancement such as studying new carriers and whether they are also suitable for enhancing reaction processes such as IC. The above research is important to verify the feasibility and reliability of its industrial application.

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