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Review Article

Hydrogen-based membrane biofilm reactors for nitrate removal from water and wastewater

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ABSTRACT

H_2-based membrane biofilm reactor (MBfR), a kind of autotrophic denitrification system, is a novel and special membrane bioreactor using hydrogen as inorganic electron donor to reduce nitrate and nitrite in water and wastewater. In this paper, the state of the art of recent research on denitrification through H_2-based MBfRs is reviewed, including theoretical fundamentals, key influencing factors, possible problems and applications. Hydrogen/nitrate counter-diffusion has been described as Dual substrates limitations. The denitrifying bacteria in H_2-based MBfR were summarized. The key factors affecting the performance of H_2-based MBfR were listed in terms of substrate concentrations, membrane materials, reactor types, biofilm management and operation conditions. The pH value, salinity, dissolved oxygen, HRT (hydraulic retention time) and carbon source have been identified as main operational conditions affecting H_2-MBfR performance. Furthermore, membrane fouling in H_2-based MBfRs was emphasized. H_2-MBfR was proved excellent in denitrification based on its high performance for groundwater, IX brine and aquaculture wastewater treatment. Several aspects may be considered in future works were proposed.

Contents

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Introduction

Hydrogen is the most plentiful element in the universe. Hydrogen has extensive applications, for example, hydrogen is an important raw material in chemical industries, and it is also used as a reducing agent, energy carrier and fuel [1]. Various technologies have been used for the production of hydrogen. From the perspective of energy source, hydrogen can be produced from biomass (agricultural wastes, municipal wastes, algae etc.), renewable energy (solar, wind etc.), fossil fuel (coal, oil, natural etc.) and nuclear energy [2–5]. Theoretically, all the oxidized pollutants can be reduced by H_2-MBfRs. Hydrogen can be used for the bioreduction of oxidized pollutants in water and wastewater, including NO_3^-, ClO_4^-, BrO_3^-, Cr(VI), SeO_4^{2-}, $H_2AsO_4^-$ and U(VI) and the like. Among them, nitrate is the most important pollutant, which can cause the eutrophication of water body, how to remove nitrate has been received increasing attention in recent year, and various methods have been studied for the removal of nitrate from water and wastewater. In this review paper, we focused on the bioreduction of nitrate by H_2-based MBfR.

Nitrate contamination of surface and groundwater has become an increasingly serious issue throughout the world due to the intensive fertilizer application and industrial wastewater discharge [6]. Nitrate contamination has been shown to cause adverse health effects, including methemoglobinemia (blue-baby syndrome) in infants, non-Hodgkin's lymphoma, gastric cancer, mutagenesis and teratogenesis, miscarriage in pregnant women, coronary cardiac diseases, cancer of the ovaries and growth of hypertrophy of the thyroid [7,8]. The World Health Organization sets a nitrate nitrogen limit of 11.3 mgL^{-1} in drinking water supplies and the US Environmental Protection Agency sets the maximum admissible concentration limit of 10 mgL^{-1} for nitrate nitrogen [9].

Among the various methods available for nitrate removal involving ion-exchange (IX), reverse osmosis (RO), electro dialysis (ED), chemical methods, etc., heterotrophic denitrification is the most economical [6,10–19]. However, external organic carbon sources such as methanol, ethanol, acetate, and corn syrup, are required to be added in the process to overcome lack of an organic electron donor, which contributes significantly to the overall operating cost and result in by-product and residual organics in the effluent [10]. Moreover, it is not easy to obtain a persistent low effluent organic concentration by controlling the external organic carbon source dosage due to the influent organic concentration fluctuation [6,11]. Autotrophic denitrification, where inorganic carbons source is used such as carbon dioxide, could avoid the presence of residual organic substrates or excess biomass in the process effluent [20–23]. In addition, the growth rate of autotrophic denitrifying bacteria ensures low biomass build-up and limited operating problems [10]. Hydrogen, as the electron donor, is clean and harmless to humans, and even cheaper than the organic electron donor [8]. It has therefore been considered as an outstanding electron donor for autotrophic denitrification, so that many attentions have been paid on hydrogenotrophic denitrification [24]. However, disadvantages of hydrogenotrophic denitrification are the low solubility of hydrogen and its explosive risk. A number of authors have overcome this problem using a special membrane bioreactor, that is, hydrogen based membrane biofilm reactor (H_2-based MBfR) where membrane is used to transfer hydrogen with huge area and avoid hydrogen leakage [20,25–27].

Membrane bioreactor (MBR), containing membrane and biological units, is a promising technology for the effective and economical removal of many kinds of contaminants from water. Various types of MBRs have been reported as pressure-driven, gas transfer, extractive, biocatalytic and electrochemical MBR and so on [28–36]. H_2-based MBfR is a kind of gas transfer MBR. Hydrogen diffuses through the gas transfer membrane, where attached the biofilm by whom the oxidized contaminants were bio-reduced. H_2-based MBfR is so far applied for denitrification, bio-reduction of oxidized anionic contaminants and dehalogenation of organics in waters and

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