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Pollutant removal mechanisms in a bio-diatomite dynamic membrane reactor for micro-polluted surface water purification

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ABSTRACT

The bio-diatomite dynamic membrane (BDDM) reactor is an emerging micro-polluted surface water treatment technology that combines diatomite (the microorganism carrier) and a stainless steel mesh (the dynamic membrane support module). A constant water head of 20 cm was designed to drive the BDDM filtration. The BDDM with sintered diatomite had good water penetration capacity, a filtration flux as high as 92 L/m^2 h after a filtration time of 15,780 min, and an effluent turbidity in the range of 0.15 NTU–0.20 NTU. The BDDM reactor effectively removed organic matter and ammonium nitrogen. The diatomite adsorption and the BDDM interception did not have high pollutant removal efficiencies. The dehydrogenase activity (DHA) of the bio-diatomite was in the range of 2.27–3.20 (mg TF)/(gVSS) h, indicating good microorganism activity for organic matter removal. The PCR-DGGE analysis showed that the microbial community was very abundant. Bacteroidetes, Firmicutes, Proteobacteria (e.g. α-, β-, γ-proteobacteria), Verrucomicrobia, and Nitrospirae were dominant in the bio-diatomite mixed liquor and removed organic matter and ammonium nitrogen. The microbial degradation of pollutants by the bio-diatomite mixed liquor was primarily responsible for the pollutant removal in the BDDM reactor.

1. Introduction

The micro-pollution of surface water is a common challenge faced by China and many other countries. Unfortunately, conventional water treatment systems, i.e., screening, coagulation, flocculation, sedimentation, rapid sand filtration and disinfection [1,2], have difficulty removing dissolved organic matters from source water, with only approximately 30% removed at most [3,4]. The application of ultrafiltration/microfiltration membranes to treat drinking water separates particles, colloids, and bacteria from source water [5]. However, neither effectively removes natural organic matter [6], synthetic organic compounds [7], or ammonium nitrogen. Biological processes have been used extensively to eliminate biodegradable organic matter and ammonium from polluted raw water for potable water production [8,9]. Different materials, such as polyethylene particles, granular activated carbon, sand, anthracite, and zeolite, have been used as microorganism carriers [10].

Diatomite, which consists primarily of amorphous SiO_2, has high porosity, good hydrophilicity, and high chemical stability [11]; it is negatively charged in natural surface water [12]. Based on low cost, environmental friendly nature and above characters, diatomite has been widely used as filter aid, adsorbent and catalytic support [13–16]. Recently, to produce high quality treated water, diatomite has been used as a microorganism carrier to form bio-diatomite for surface water and municipal wastewater treatment [17,18]. The suspended diatomite continuously moves in the aeration tank, while the active biomass grows as a biofilm on the surface of the carriers.

In addition, the dynamic membrane technology has been adopted for bio-diatomite mixed-liquor separation. This dynamic membrane is dynamically created on the underlying stainless steel support mesh with big pore size when filtering the bio-diatomite particles; thus, it is also called a formed-in-place membrane [19] or a secondary membrane [20]. The combination of the bio-diatomite and dynamic membrane technologies to form a bio-diatomite dynamic membrane (BDDM) reactor is a promising technology for micro-polluted surface water treatment [18]. In an integrated BDDM reactor system, pollutants can be removed effectively in a single reactor. Liu et al. [21] divided the formation process of dynamic membrane into four stages based on the flux behaviors under constant filtration pressure, i.e., substrate formation, separation layer formation, fouling layer formation and filtration cake formation; while the formation of dynamic membrane can also be divided into three stages reported by Wang et al. [22], i.e., the formation of separation layer, the stable growth stage, and the fouling stage. When the dynamic membrane was formed, the support membrane itself may be no longer necessary, since solid rejection will be accomplished by the cake layer [22,23]. Once the

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membrane is severely fouled, the dynamic layer can be removed and replaced by a new deposited layer.

The mechanisms contributing to the reduction of pollutants in the BDDM reactor may include diatomite adsorption, dynamic membrane interception, and microbial degradation. It is important to sufficiently clarify which mechanism is primarily responsible for pollutant removal to enable the development of this new technology. Therefore, this paper investigates the pollutant removal mechanisms for micro-polluted surface water purification in a BDDM reactor using a gravity filtration mode to drive the BDDM filtration.

2. Materials and methods

2.1. Diatomite characteristics

Sintered diatomite of the filter aid type was used in this study to facilitate the filtration process. Diatomite has a honeycomb structure and a mean particle size of 5.80 μm. The BET specific surface area of sintered diatomite is a relatively low average of 0.2628 m^2/g. Chemical analyses of the sintered diatomite composition showed 88.2% SiO_2, Al_2O_3 3.5%, Fe_2O_3 1.9%, and CaO 1.1%.

2.2. BDDM reactor processes

The method of bio-diatomite cultivation can be referred from the previous research [18]. The BDDM reactor with a total effective volume of 0.035 m^3 consisted of three parts: two aerobic tanks and a dynamic membrane filter (DMF) in sequence (Fig. 1). The concentrations of mixed liquor suspended solid (MLSS) and mixed liquor volatile suspended solid (MLVSS) in the reactor were maintained at about 21,436–24,064 mg/L and 612–682 mg/L, respectively. The average sludge age was 50 d with 700 mL of sludge discharged daily to renovate the microorganisms' community. The dosing rate of diatomite was properly adjusted according to the discharged sludge quantity and the real-time MLSS and MLVSS concentrations. During the experiment, the dissolved oxygen in the reactor was maintained at 3–4 mg/L. The mixed liquor in the DMF was recycled to the first aerobic tank at a flow rate of the influent flow.

The support layer used a stainless-steel mesh with an equivalent aperture of 48 μm. The flat support module was fixed in a submerged mode with a double-sided effective filtration area of 0.043 m^2 (19.5 cm × 11 cm). The BDDM was formed using the self-forming mode. The operation period of BDDM involved three stages, i.e., precoating, filtration and backwash [18]. A constant water head (20 cm) was applied to produce the gravity filtration of BDDM in both the precoating and the filtration stages, and the flux decreased as the filtration resistance increased. The filtration stage was stopped once the BDDM flux was at about 100 L/m^2 h, and then air backwash was started using an air pump (Maple Mini Air Compressor, China).

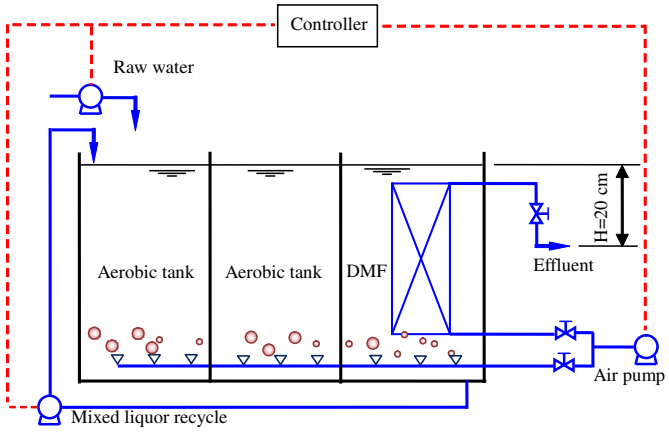

Fig. 1. Schematic diagram of the BDDM reactor process.

2.3. Static absorption test

A series of sorption experiments were carried out by transferring 1000 mL of distilled water into a 2000 mL jar and mixing 20,000 mg of diatomite measured by an analytical balance (AL204, Mettler Toledo, China). The solution was then stirred at 100 rpm and left to stand for 120 min after stirring. Subsequently, 200 mL samples were collected separately from the jar and analyzed.

2.4. Dehydrogenase activity (DHA) analysis

DHA was measured using the triphenyl tetrazolium chloride-dehydrogenase activity (TTC-DHA method, which is based on the production of triphenyl formazan (TF) from the reduction of 2,3,5-triphenyltretrazolium (TTC)). Samples of 100 mL of mixed sludge were centrifuged at 6000 rpm for 6 min. The supernatant was then discarded. Next, 2 mL of this sludge was mixed with 2 mL of 1% TTC, 2 mL of 1% glucose, and 2 mL of Tris–HCl. The samples were incubated in the dark for 24 h at 37 °C. After incubation, 2 drops of H_2SO_4 (98%) and then 5 mL of toluene were added to each tube. The samples were then shaken for 2 min and centrifuged at 4000 rpm for 5 min, after which the organic layer was removed. The absorbance of the organic layer at 485 nm was then measured using a spectrophotometer. The amount of TF was determined from the standard TF curve. TTC-DHA activities were expressed in terms of mg TF produced/g volatile suspended solid h ((mg TF)/(gVSS) h).

2.5. DNA extraction and PCR-DGGE

The bacterial DNA was extracted using a FastDNA® spin kit for soil (MP, United States) according to the manufacturer's instructions. Primers 8f (5′-AGAGTTTGATCCTGGCTCAG-3′) and 1492r (5′-GGTTACCTTGTTACGACTT-3′) were initially used to amplify the bacterial 16S rDNA. Then, primers 341f with a GC-clamp (5′-CGCCCGCCGCGCGCGGCGGGCGGGGCGGGGGCACGGGGGGCCTAGGGGAGGCAGCAG-3′) and 534r (5′-ATTACCGCGGCTGCTGG-3′) were used to amplify the V_3 region of the bacterial 16S rDNA. The PCR mixtures had a final volume of 25 μL and contained 2.5 μL 10 × PCR buffer, 1 U of Taq DNA Polymerase, 1.0 μL of dNTP (2.5 mmol/L each), 0.5 μL of each primer (10 μM) and 50 ng of DNA as the template. DGGE was conducted using the D Code™ Universal Mutation Detection System (Bio Rad, USA). The PCR products were electrophoresed directly in a 10% (v/v) polyacrylamide gel in a 1 × TAE buffer containing a linear gradient that ranged from 35% to 55% denaturant (100% denaturant was equivalent to 7 mol/L urea and 40% (v/v) deionized formamide). Electrophoresis was performed at 60 °C under a constant voltage of 120 V. After 6–7 h of electrophoresis, the gel was stained with a DNA-nucleic acid stain dye (BBI, Germany) for 30 min and then photographed.

2.6. Sequencing of DGGE band

Prominent bands were excised from the DGGE gel for 16S rDNA fragment sequencing. The fragments were then re-amplified by PCR and purified using a gel extraction system B (BioDev, China), after which they were cloned into the pMD19-T plasmid vector system (TaKaRa, Japan). The DNA sequences were then determined by a commercial service (Shanghai Invitrogen Biotechnology Co., Ltd., China). The vector sequence was cut off, and the remaining nucleotides were compared to those available in GenBank using the BLAST program to identify the most similar 16S rDNA fragments.