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Synthesis and performance of iron oxide-based porous ceramsite in a biological aerated filter for the simultaneous removal of nitrogen and phosphorus from domestic wastewater

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ABSTRACT

A novel iron oxide-based porous ceramsite (IPC) was synthesized and applied as a microbial biofilm carrier in a biological aerated filter (BAF) to treat domestic wastewater and compared to commercially available ceramsite (CAC). The results indicate that IPC has a higher porosity in comparison to CAC. The uniformity and interconnectivity of pores, as well as the rough surface of the IPC are suitable for microbial biofilm growth. Biofilm growth occurs in the internal pores of the media and promotes nitrogen and phosphorus removal. The effect of air-water ratio (A/W) on the removal of total organic carbon, ammonia nitrogen, total nitrogen, and phosphorus were investigated. The results show that the performance of IPC BAF is much better than CAC BAF. For instance, at an A/W ratio of 3:1, the total nitrogen removal was 46.26% with IPC and 15.64% with CAC, and the PO_4^{3-} removal was 72.25% with IPC compared with only 33.97% with CAC. An analysis of the microbial community in the IPC BAFs by polymerase chain reaction denaturing gradient gel electrophoresis identified *Dechloromonas* sp., *Sphaerotilus* sp., and *Nitrospira* sp. microbes. The diversity on microbial population, along with the attached growth benefit from the morphological properties of IPC, allows enhancement in the simultaneous nitrification and denitrification performance in IPC BAF. Hence, IPC can be considered a very effective novel media material in BAF for the simultaneous removal of nitrogen and phosphorus.

1. Introduction

Eutrophication can occur when excessive nitrogen and phosphorus reach water bodies through runoff or discharge of wastewater [1]. Eutrophication can lead to the deterioration in water quality damaging aquatic ecosystems. An outbreak of harmful cyanophytes, commonly known as cyanobacteria, will eventually result in increased mortality of aquatic organisms. Hence, the discharge of treated wastewater must meet effluent quality standards for phosphorus and nitrogen. Novel wastewater treatments are required for the simultaneous removal of phosphorus and nitrogen (SPN) since conventional treatments have commonly been designed to focus on removal of one nutrient. Traditionally, technologies such as biological and chemical treatment for phosphate or nitrate have been widely used [2,3]. Chemical treatment employs a metal salt to coagulate and remove phosphate by sedimentation. The excess amount of sludge produced is a major limitation to this process [2]. Adsorption is the most widely used and suitable method for phosphorus removal. Of all the adsorbents, iron oxides and untreated clays are considered the most effective sorbent materials [4,5]. Previous studies have shown that raw goethite, nano zero-valent iron (Fe^0), and hematite are very effective materials adsorbing phosphorus [6,7]. Simultaneous nitrification and denitrification (SND) for the removal of nitrogen can be achieved through biological processes. SPN removal from wastewater can be achieved by the combination of different biological and/or chemical technologies (i.e., sequential treatment). A biological aerated filter (BAF) is a fixed-film biosystem with a small footprint that employs filter media with a high specific surface area and porosity. The filter can promote in-growth biofilm(s) for wastewater treatment. BAFs can reduce chemical oxygen demand

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(COD), suspended solids and ammonia [8,9]. Microbial biofilm supported filter media, which are made of raw clay, wastewater sludge, construction waste and raw zeolite, have been evaluated in recent years and have achieved good results for nitrogen removal [2,10,11]. Dissolved oxygen (DO) plays a key role during the BAF process. Denitrifiers are active in areas of very low DO concentration whereas nitrifiers are active in areas of high DO concentration [12]. SND has been proven to be the most effective way to remove total nitrogen (TN) which occurs naturally inside the microbial biofilms of filter media [13]. Porous filter media involving a mixed population and upholding oxygen diffusion limits create both anoxic and aerobic micro-environments. As such, the filter media allows the sequential use of electron acceptors, such as oxygen and nitrate. Such mass transfer leads to the stratification of microbial species in the microbial biofilm of filter media [15,16]. BAF technology is applied to wastewater treatment; hence, the selection of filter media plays a significant role in maintaining a variety of microbial populations and a high amount of active biomass. Recently, some waste materials, such as waste ceramics and polyethylene plastic, have been utilized as filter media for biological aerobic filters, providing a new approach for disposing the waste sawdust [2]. This strategy serves as a “win–win” solution by treating wastes with wastes. Furthermore, several new filter media for BAF have been recently investigated to enhance biodegradation capability. For example, combined up-flow anaerobic biofilters and up-flow BAF that employ filter media prepared by sludge, coal cinder, and straw have been used to treat synthetic wastewater [17]. These filter media achieved favorable results for nitrogen removal. However, only a few studies investigated in detail SPN with BAF.

In China, commercially available ceramsite (CAC) has been newly used in filter media for domestic wastewater treatment. However, such material was reported to exhibit extremely low nitrogen and phosphorus removal efficiencies in BAFs. This result is probably due to the poor affinity of CAC to microbial biofilm biomass, which is caused by biocompatibility and suboptimal surface hydrophilicity [2,11]. For this reason, developing a novel filter medium as a substitute for CAC in the simultaneous removal of nitrogen and phosphorus must be accomplished as quickly as possible.

Previous studies reported that sawdust was treated at 700 °C to produce a desirable porous material. The formation of interior pores improves microbial growth. It was reported that hematite and palygorskite calcined at 700 °C can effectively absorb phosphorus [2,5]. In this study, iron oxide-based porous ceramsite (IPC) with goethite, sawdust, and palygorskite were synthesized by calcination in an O_2 atmosphere, used as microbial biofilm support in BAF and compared to commercially available ceramsite (CAC) [14]. The properties of IPC, CAC and microbial characteristics were characterized by scanning electron microscopy (SEM), porosimetry analysis (PM) and micro-computed tomography (Micro-CT). The effect of air-water ratio (A/W) on the removal of total organic carbon (TOC), ammonia nitrogen (NH_3-N), total nitrogen (TN) and phosphorus (P) was investigated using both supports. Finally, the relationship between protozoa and metazoa as well as the characteristics of microbial biofilms were determined using polymerase chain reaction denaturing gradient gel electrophoresis (PCR-DGGE) of the microbial community of the IPC BAF.

2. Materials and methods

2.1. Characterization and analytical methods

Details of reagents and preparation of the proposed filter media-IPC are provided in the supplementary material. IPC and CAC surface morphologies were examined using a Scanning Electron Microscope (SEM, Philips XL30 ESEM-Netherlands Philips Company). The physical characteristics of IPC and CAC were measured in accordance with the sandstone pore structure method of image analysis. Rubber casting experiments were used to generate quantitative size and shape data from pores in the thin section (d = 3 cm) [2,14,18]. The porosities of IPC and CAC specimens were determined using Micro-CT (μCT 40, Scanco Medical, Brüttisellen, Switzerland). The two samples were scanned in air at 70 kV and 57 μA, with an isotropic voxel size of 12 μm. A region of interest (ROI) was defined for each gray-value image, which corresponded to the exact circumference of the sample. A binary threshold was applied to each sample and Scanco built in reconstruction algorithms were used to derive the porosity and to construct the 3D image [19–21].

Microscopic observations for protozoan and metazoan population growth were carried out using a U-RFL-T Olympus Biological Microscope (Olympus Corporation, Tokyo, Japan). The multi-point BET surface area of IPC and CAC was measured using a Quantachrome Nova 3000e automated surface area analyzer. The growth of biofilm was determined according methods available in the literature [2,13]. The microbial populations in IPC BAF were analyzed by using domain-specific F341 R907 primers and PCR amplification of 16S rRNA gene fragments for genomic DNA from bacteria. The PCR products were applied in the DGGE analysis using a BioRad Dcode system (BioRad Co. Ltd., USA). A DGGE gel of 8% polyacrylamide with a linear denaturing gradient ranging from 35% to 60% (100% denaturing gradient contains 7 M urea and 40% formamide) was used. Electrophoresis was conducted at a constant voltage of 75 V in 1 × TAE buffer at 65 °C for 14 h. The gels were then stained with 5% Gold view in 1 × TAE buffer for 40 min and imaged using UV trans-illumination. Selected DGGE bands were excised using the SK1131 kit (Sangon Biotech (Shanghai) Co., Ltd) and re-amplified by PCR with the aforementioned primers. The PCR products were sequenced at Sangon Biotech Co., Ltd (Shanghai, China). The obtained sequences were submitted and compared to the reference microorganisms in the GenBank database using the BLAST program [22,23].

2.2. Operating conditions of IPC BAF and CAC BAF

A schematic diagram of the experimental set-up is provided in the supplementary material. Biofiltration was initiated by introducing seed sludge obtained from the Hefei City wastewater treatment plan into BAFs. Each experiment was divided into 3 stages for each BAF. During each test stage, the operating conditions of both BAFs were identical (see Table 1). A stable hydraulic retention time (HRT) of 7 h was used. Three air-water (A/W) ratios of 1:1, 3:1, and 6:1, respectively, were tested. The two BAFs were monitored for 4 months after the initiation of biofiltration. Both BAFs were operated in summer at wastewater temperatures ranging from 26 °C to 30 °C. Samples were analyzed for NH_3-N and P in accordance with Chinese EPA standards [2,24]. A TOC/TN (Jena Multi N/C 2100) analyzer was used to measure TOC and TN. Dissolved oxygen (DO) concentrations were measured using a portable digital DO meter (Oxi-315, Hao-Yang Biotech Company, Shanghai, China).

3. Results and discussion

3.1. Characteristics of IPC and CAC

Table 1 shows the elemental properties of IPC and CAC. IPC possesses a higher surface area and porosity than those of CAC. Lower apparent and bulk densities were also observed in the IPC. All of these properties are associated with the chemical compositions