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Production of zeolite composite filters using waste paper pulp as slow release carbon source and performance investigation in a biological aerated filter

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ABSTRACT

The filter medium plays an important role in modulating the performance of biological aerated filters (BAFs). In this study, a zeolite composite filter (ZCF) was fabricated using a mixture of natural zeolite powder as the main material, cement as the binder, and waste paper pulp as the pore-forming agent (slow-release carbon source). Scanning electron microscopy and porosimetry were then conducted to evaluate the properties of the ZCF. Results showed that the uniform and interconnected pores of the ZCF are suitable for microbial growth. ZCF and commercially available ceramsite (CAC) used in an upflow laboratory-scale BAF were investigated to determine their suitability for advanced wastewater treatment. The results indicated that the ZCF–BAF is superior to the CAC–BAF in terms of total organic carbon, total nitrogen, ammonia nitrogen, and phosphorus removal. The carbon source from the waste paper pulp was released slowly, which decisively influenced simultaneous nitrification and denitrification. Therefore, ZCF is suitable for use as a novel filter medium for the simultaneous removal of nitrogen and phosphorus in BAFs for advanced wastewater treatment.

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1. Introduction

In recent years, water pollution and shortage have become serious problems all over the world. Eutrophication of wastewater sources is a pervasive global pollution issue, and phosphorus and nitrogen represent essential nutrients for the growth of algae [1]. Recent research investigations have shown that 54% of lakes in Asia are eutrophic; 53% in Europe; 48% in North America; 41% in South America; and 28% in Africa [2]. Domestic sewage discharge has totaled about 462.7 million tons over the last few years [3]. In 2015, the Chinese Academy for Environmental Planning drafted policies to address problems regarding water pollution through the Water Pollution Control Action Plan. One of the major actions suggested to solve these problems is domestic sewage reuse. Nitrogen and phosphorus removal and recovery are expected to be increasingly applied within the next several decades [4].

Biological aerated filters (BAFs) are fixed-film systems that offer potential use in various stages of wastewater treatment. BAFs contain filter media with higher specific surface areas per unit volume for microbial biofilm development [5]. The selection of filter medium is crucial in maintaining large amounts of active biofilm biomass and a variety of microbial populations. Synthetic materials have also been used as filter media, including polystyrene and polyethylene; the production of these chemicals, however, usually entails high costs [6,7]. Natural mineral materials found in zeolite and palygorskite clay have been frequently used to fabricate BAFs. Zeolite exhibits a higher storage capacity, lower cost, and stronger adsorption ability than other materials. Improved microorganism growth and reproduction, as well as ammonia nitrogen shock resistance, are among the advantages of a clinoptilolite BAF [8–11]. Previous research has shown that the number of nitrobacteria growing on the surface of clinoptilolites is 3.5 times greater than those growing on sandy filtering media, thereby resulting in 70–90% removal of NH_3–N and NO_3–N [12]. Feng et al.

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demonstrated the use of haydite and sorption functional media as filter media for BAF in the treatment of synthetic wastewater [8]. Chang et al. applied natural zeolite particles (NZP) and modified NZP to the BAF process to treat textile wastewater [12]. These studies indicate that zeolite exhibits good affinity toward ammonium ions and is suitable for use in their removal. However, although these materials achieved desirable results for nitrogen removal, few researchers investigated their capacity for phosphorus removal [11]. Hence, investigation of the simultaneous removal of both nutrients from wastewater is warranted.

In China, due to the rapid industrial development, the management of solid waste has become a heavy economic burden to industry; thus, the use of solid waste as resource has become an important strategy [13]. Waste paper pulp refers to the various slurry preparations used in the manufacture of paper products. Paper products are generally obtained from wood or cotton fibers. The carbon source from the waste paper pulp is released slowly. Microorganisms present in the wastewater release carbon from the porous structure of a BAF. Wang et al. reported that rape straw and waste paper pulp can be used as slow-release carbon sources for long-term anaerobic wastewater treatment [14]. Tang et al. reported that polylactic acid and starch to develop a slow-release carbon source and filter media by blending and fusing techniques for removing nitrate contamination from groundwater [15]. Cement is a binder, which pertains to a substance that sets, hardens, and fastens materials together. The word “cement” traces back to the Romans, who used the term *caementicium* to describe masonry resembling modern concrete that was made from crushed rock and burnt lime, which served as a binder. Cement contains ettringite, portlandite, and calcium silicate hydrate (CSH) and/or monosulfate as major hydration products. Amorphous CSH, composing about 60% of cement, has a high sorption potential with a high specific surface area and variable composition of Ca/Si [16]. Cement has been used successfully for the removal of phosphate from municipal wastewater treatment [17]. Li et al. demonstrated that the development and application of non-sintered fly-ash/cement composite ceramsite in a BAF achieves high efficiency during pollutant removal [18]. Because certain solid waste materials, such as waste ceramics, have been successfully used as filter media for BAFs, a novel approach for waste paper pulp disposal, has become available for the effective treatment of wastewater [19,20]. Commercially available ceramsite (CAC) is used as a large-scale filter medium in city wastewater treatment plants in China. However, researchers indicate that the bioactivity and contaminant removal efficiency of CAC in BAFs are low, probably because of the poor affinity of CAC to the biofilm biomass, which is caused by suboptimal surface hydrophilicity and biocompatibility [10]. Therefore, determination of an appropriate substitute for CAC must be performed.

In the present study, natural zeolite powder, waste paper pulp, and cement were used as the main materials for fabricating zeolite composite filter (ZCF), which was then be used as biofilm biomass support in a BAF. The physical properties of the ZCF and the microbial biofilm characteristics of the resulting BAF were analyzed by various characterization and analytical techniques. The effectiveness of the ZCF as microbial biofilm support in BAF was compared with that of CAC for municipal wastewater treatment.

2. Materials and methods

2.1. Material

The natural zeolite particles (NZP) were obtained from Xuan Cheng, Anhui Province, China. Natural zeolite powder size was less than 0.15–0.18 mm after extrusion, cutting and crushing. Waste paper pulp was obtained from He Fei Paper Making Company, in He Fei City, Anhui Province, China. Cement was obtained from Anhui Conch Cement Company Limited, in Wuhu City, Anhui Province, China. CAC was obtained from the city of Ma'anshan, Anhui Province, China.

Table 1
Operating conditions.

Start-up operating conditions			
PH	T (°C)	A/L	HRT (h)
6.32–6.73	20–25	3:1	7(±0.5)
Water quality indexes			
TOC	NH_3–N	TN	PO_4^{3-}
25.84	10.51	13.96	0.51

*TOC/NH_3–N/TN/PO_4^{3-}.
Values are average influent concentrations in mg/L.

2.2. Physical characterization of ZCF, CAC, NZP and natural zeolite powder

The chemical composition of CAC, natural zeolite powder, and waste cement was measured on a Shimadzu XRF-1800 with Rh radiation. X-ray diffraction (XRD) was performed using a Rigaku powder diffractometer with Cu Kα radiation [21]. The physical characteristics of ZCF, CAC and NZP were evaluated in accordance with the sandstone pore structure method of image analysis. [21–23]. For biological structure analysis, ZCF, NZP and CAC were gilt with pores and their surface morphologies were examined using a Scanning Electron Microscope (SEM, Philips XL30 ESEM). Elemental analysis of sawdust was carried out in a VARIO EL III analyzer (Elemental Analysis System Co., Ltd., Germany). The microscopic observation for protozoan and metazoan population development was carried out using a U-RFL-T Olympus Biological Microscope (Olympus Corporation, Tokyo, Japan). The growth of biomass biofilm was determined according methods in the available literature [21].

2.3. Experimental set-up

A schematic diagram of the two BAFs used in the experiments is given in Fig. S1. BAF was constructed of a PVC pipe with a diameter of 60 mm and a media depth of 1500 mm. The two BAFs were packed with ZCF and CAC, respectively. During start-up stage, operating conditions of ZCF BAF and CAC BAF were identical, which are summarized in Table 1. Wastewater samples were harvested from the outlet and inlet pipes of the two BAFs. Analytical methods for measuring ammonia nitrogen (NH_3–N), phosphorus (P), nitrite (NO_2–N), and nitrate (NO_3–N) were carried out in accordance with Chinese EPA standards [24]. Total organic carbon (TOC)/total nitrogen (TN) (Jena Multi N/C 2100) analyzer was used to measure TOC and TN [22].

3. Results and discussion

3.1. X-ray fluorescence (XRF) and X-ray diffraction (XRD)

Fig. 1 and Table S1 shows the XRD and XRF patterns of ZCF, cement and natural zeolite powder. As shown in Fig. 1(1), the peak of 2θ was intense around 9.88, 22.48, 27.78 and 30.38 and was identified as zeolite (JCPDS 89-6538) [12]. A reflection at $2\theta = 26°$ is also found and identified as quartz. It indicates the natural zeolite powder is mainly composed of zeolite, montmorillonite, and only small amounts of quartz. Fig. 1(2) shows the XRD patterns of cement. The main mineral components of cement were quartz (SiO_2), calcite ($CaCO_3$), and calcium silicate ($CaSiO_3$). ZCF is synthesized according to the parameters mentioned above and the XRD

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