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Synthesis, application and evaluation of non-sintered zeolite porous filter (ZPF) as novel filter media in biological aerated filters (BAFs)

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ABSTRACT

A zeolite porous filter (ZPF) was prepared using mixed raw zeolite, cement, and aluminum powder through steam curing and used as a novel filter medium in biological aerated filter (BAF). The performances of ZPF and commercially available ceramsite (CAC) in two laboratory scale upflow BAFs were compared. Results showed that the interconnected porous structure of ZPF was conducive to microbial biofilm growth. ZPF featured a total porosity of 29.55%, a compressive strength of 41–47 N, and a specific surface area of 59.53 m^2/g. BAF containing ZPF showed more effectively the removal of organic carbon, ammonia nitrogen, nitrogen, and phosphorus compared to BAF containing CAC. The hydraulic retention time (HRT) was 7 h at an air/water (A/W) ratio of 3:1. The amounts of total nitrogen removed were 59.89% with ZPF and 35.96% with CAC. Moreover, the amount of phosphorus removed was 83.80% with ZPF, whereas that of CAC was only 31.50%. ZPF was more suitable for the attached growth of heterobacteria and nitrobacteria to attain simultaneous nitrification and denitrification performance in the BAF. Therefore, ZPF is a novel suitable filter medium for simultaneous removal of nitrogen and phosphorus in BAFs.

1. Introduction

Nitrogen and phosphorus compounds are nutrients essential to all life forms. The presence of these elements in substantial quantities in receiving waters, such as lakes and rivers, can cause eutrophication, such as in the Chinese four lakes; this phenomenon results in excessive growth of algae and other microorganisms and enhanced dissolved oxygen depletion and fish toxicity. As the eutrophication of waters becomes more frequent, the removal of the two kinds of nutrients has gained increased importance in wastewater treatment [1–5].

Previous studies on phosphorus and nitrogen co-contaminated wastewater primarily focused on the removal of either phosphorus [5] or nitrogen [1–5]. A few studies have examined the removal of nitrogen and phosphorus simultaneously [1]. For example, the classical activated sludge process is implemented to treat sewage and industrial wastewater by using air and biological flocs containing bacteria and protozoa. The classical activated sludge process is also used for the treatment of high concentration wastewater [3]. The biological aerated filter (BAF) is a well-known bioreactor for wastewater treatment that is extensively applied worldwide [6]. BAF can remove pollutants through carrier filtration and biodegradation [6]. BAF, a wastewater treatment technology that is based on the principle of biofiltration, possesses the advantages of high biomass retention, tolerance to toxicity, excellent removal efficiency, and slurry separation. By means of the filter medium, BAFs can simultaneously realize biological conversion of chemical oxygen demand and ammonia and physical filtration of phosphorus compounds in filter [7–11]. As the core component of BAF systems, the filter medium is used for the immobilization of microorganisms; the filter medium is significant for the treatment of wastewater. BAF systems are applied for two reasons. First, microorganisms are difficult to attach to many types

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of filler. Second, clogging and stoppage of the biofilter easily occur [6]. BAF was originally utilized in tertiary or advanced wastewater treatment and subsequently applied in secondary wastewater treatment as the technology matured [6]. Compared with classical activated sludge, BAF is more suitable for advanced nitrogen and phosphorus removal [3,6]. Therefore, BAF is an effective and economical method for the treatment of municipal and industrial wastewater.

In China, commercially available ceramsite (CAC) is a lightweight aggregate used as a filter medium in the BAFs of municipal sewage plants for the last several years [12]. Generally, the raw materials of CAC consist of mineral clay, which is mostly obtained from farmlands and mining. The development of CAC occurs during calcination at high temperatures above 1200 °C. This phenomenon will destroy the main raw mineral-clay crystal structure and decrease the number of adsorption sites. Thus, bioactivity and contaminant removal efficiency decreases in BAF. The common CAC achieves poor results for nitrogen and phosphorus removal [13,14]. Therefore, it is important to find an appropriate material as substitutes for CAC, which achieves excellent results for the simultaneous removal of both nutrients.

Zeolites are hydrated aluminosilicates with symmetrically stacked alumina and silica tetrahedra, which produces an open and stable three-dimensional honeycomb structure with a high cation exchange capacity, cation selectivity, high void volume, and great affinity for NH_4^+ [15–17]. The use of zeolite powder for removal of NH_4^+ is considered as an effective treatment method because of its low cost and relative simplicity of application and operation [15–17]. For this reason, zeolite composite ceramsite has been widely prepared and used in the wastewater treatment in recent years. However, most zeolite composite ceramsite are obtained by a high sintering process, which is complicated and features high energy consumption, and high cost [15–17]. A high calcination temperature (≥600 °C) is inevitable and will destroy the main crystal structure of the raw material, namely, zeolite [18]. Therefore, a novel non-sintered zeolite porous filter (ZPF) must be developed because of its simple process, low energy consumption, and low cost.

Conventional processes for phosphorus removal involve physical, chemical, and biological processes [19]. The chemical process of flocculation is an attractive phosphorous removal method because of its simple implementation. However, its efficiency in nutrient removal remains controversial [20]. Fly ash, slag, ordinary Portland cement, and related blends have been successfully used to remove phosphate from wastewater [20–22]. Adsorption is an attractive alternative in removing nutrients (such as phosphorus) from the effluent by using porous materials, such as cement and zeolite-like substances, because cement is enriched with oxides of calcium. These oxides can strongly adsorb or precipitate phosphates. Therefore, cement can be a candidate material for phosphate removal [23].

Porous structures are important factors for microbial biofilm growth [12]. Scanning electron microscopy (SEM) and mercury intrusion porosimetry (MIP) are commonly used to demonstrate pore architecture and morphology. SEM provides high resolution and highly detailed views of the three-dimensional (3D) surface topology. However, the inherent weakness of SEM is that it is limited to two-dimensional (2D) measurements on relatively small fields of view. Consequently, the structure of interconnections may be difficult to envisage or obtain in perspective. MIP provides information on the porosity, pore size distribution, and internal surface area of the entire specimen. However, this method cannot detect isolated pores in scaffolds and has potential difficulties for analysis of deep pores connected by narrow channels in complex porous structures. Moreover, both methods necessarily result in sample destruction. As an alternative, X-ray micro computed tomography (micro-CT), uses X-rays to create cross-sections of a physical object that can be used to recreate a virtual model (3D model) without destroying the original object [24,25]. Micro-CT primarily applied in medical imaging for bone structure analysis, but it finds more and more usage in industrial computed tomography [26–29]. Although this method is highly efficient in analysis of pore formation, few researchers use this method for porous filter media.

In this study, zeolite porous filter-ZPF is manufactured by steam curing of raw materials consisting of cement, zeolite, pore-generating aluminum powder, and water. The BAF loaded with this kind of ZPF has high processing capability but low investment and operation costs. The formation of interior porosity improves microbial biofilm growth. This study may reveal a new promising method for preparation of porous filter media. Therefore, ZPF is considered a novel filter media for simultaneous removal of nitrogen and phosphorus (SNP).

2. Materials and methods

2.1. Materials

Zeolite was obtained from Xuan Cheng, Anhui Province, China. Cement was obtained from Anhui Conch Cement Co., Ltd., Wuhu City, China. Aluminum powder was purchased from Hefei Chemical Reagent Corporation (China). CAC was obtained from the city of Ma'anshan, Anhui Province, China.

2.2. Preparation of ZPF

Compressive strength is an important index for preparation of ZPF [13,14]. In this study, zeolite was the main component of the ZPF tested. Cement and aluminum powder were used as additives and were mixed with the zeolite to make a new type of non-sintered ZPF. Three control factors were optimized to investigate the effects of ZPF production; these factors include amount of cement, aluminum powder, and water. Moreover, an $L_9 3^3$ orthogonal experimental was performed to determine three control factors through the compressive strength of ZPF. For each factor, three levels were chosen to cover the wide variational regions based on the results obtained from our above mentioned investigations. These factors and their levels are shown in Table S1. The $L_9 3^3$ orthogonal experiment consists of nine experiments. The final analysis shows that the first factor is the aluminum powder ratio, and the second factor is water content. The third factor is the cement ratio ($R_C > R_D > R_B$). The effects of each factor on the compressive strength of ZPF were investigated, and the results are shown in Table S1. According to this principle, Table S2 shows that the optimal parameters for ZPF production are as follows: aluminum powder, 0.12 wt%; cement, 32 wt%; water content, 52 wt% materials. The optimal steam temperature and time for ZPF are 123 °C and 480 min, respectively. The photographs of ZPF are presented in Fig. S2.

The preparation of ZPF includes the following steps (Fig. 1): ZPF is made from zeolite, cement, and aluminium powder which are mixed with amounts of water and molded to produce a cellular green body by H_2 gas generation at atmospheric pressure, and then autoclaved under saturated steam pressure at 123 °C for 480 min. The aluminium reacts with hydroxide of calcium or alkali, which liberates hydrogen gas and forms bubbles, as shown in Eq. (1). The speed at which the air bubbles form is critical to the success of the final ZPF [30].

$$2Al + 3Ca(OH)_2 + 6H_2O \rightarrow 3CaO \cdot Al_2O_3 \cdot 6H_2O + 3H_2 \quad (1)$$