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## Enhanced synergistic denitrification and chemical precipitation in a modified BAF process by using Fe<sup>2+</sup>



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#### HIGHLIGHTS

- Synergistic denitrification and chemical precipitation of P and TSS in BAF process.
- Fe<sup>2+</sup> is suitable for dosing in the pre-denitrification stage of a BAF process.
- ullet Dosing  $Fe^{2+}$  increased the denitrification loading rate in BAF pre-denitrification.
- A more efficient controlling of residual P was exhibited by dosing Fe<sup>2+</sup> than Fe<sup>3+</sup>.
- Dosing Fe<sup>2+</sup> greatly improved the settleability of BAF pre-denitrification sludge.

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#### ABSTRACT

A series of laboratory-scale experiments for examining the feasibility and suitability of using Fe<sup>2+</sup> as the precipitant dosed in the pre-denitrification stage of a modified BAF process employing simultaneous chemical precipitation of TSS and phosphorus were carried out. The effects of dosing Fe<sup>2+</sup> on effluent quality and sludge characteristics of the pre-denitrification stage were assessed with comparing to the cases of no additional chemical dosing and dosing Fe<sup>3+</sup>. Results obtained demonstrated a sound performance of synergistic denitrification and chemical precipitation in pre-denitrification of the modified BAF process when dosing Fe salts, which showed enhanced by using Fe<sup>2+</sup> as the dosed precipitant in increasing the denitrification loading rate, exhibiting a better controlling of the residual phosphorus in pre-denitrification effluent, and improving sludge settleability. Dosing Fe salt showed no adverse impact in removing COD, but resulted in a relatively higher SS content in the pre-denitrification effluent.

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#### 1. Introduction

Biological aerated filter (BAF) process, which relies on attached growth of biomass on inert granular medium within compact units, is an alternative to the traditional activated sludge process (Mendoza-Espinosa and Stephenson, 1999). Since it possesses a number of advantages, including achieving solids, organic matter, and nutrient removal in a same reactor, higher pollutants loading, and hence reduced space requirement, researches, and applications of BAF have grown with great popularity in recent years (Gilbert et al., 2008; Zhao et al., 2009; González-Martínez et al., 2010; Bai et al., 2011; Abu Hasan et al., 2012; Li et al., 2013). Because solids removal in BAF is achieved mainly through filtration, total suspended solids (TSS) in BAF influent is usually below

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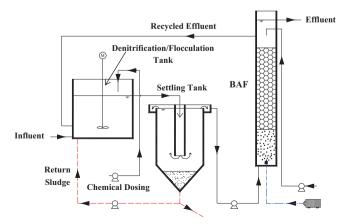
100 mg/L (Canler and Perret, 1994) to avoid filter bed clogging and associated frequent backwashing. To deal with the high suspended solids content in influent streams, chemical coagulation/ precipitation is an often applied pre-treatment step for removing TSS (Farabegoli et al., 2009). Additionally, due to the lack of alternate anaerobic and aerobic conditions, BAF is incapable of enhanced biological phosphorus removal (Lee et al., 2005), and supplementary physio-chemical precipitation of phosphorus is often used in BAF plants for meeting stringent effluent standards (Rogalla et al., 1990; Clark et al., 1997). Though an enhanced chemical pre-treatment step can reduce both TSS and total phosphorus in the influent stream fed into BAF, removal of considerable amount of organic matter can also be resulted at the same time, which reportedly could reach up to over 50% COD removal (Rogalla et al., 1990). The simultaneous organic matter removal in the chemical pre-treatment step might lead to the shortage of carbon sources in the post-denitrification process for BAF nitrified effluent, particularly when treating weak sewage with relatively low C/N

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ratio. In order to meet the total nitrogen discharge requirement in the effluent, an external carbon source has to be added in the denitrification process, and hence operating cost is increased.

In this study, a modified BAF process employing simultaneous chemical precipitation in pre-denitrification is proposed. A schematic diagram of the proposed process is shown in Fig. 1. When dosing chemicals in the pre-denitrification tank, synergistic denitrification, and removal of phosphorus and TSS through chemical precipitation is anticipated. Simultaneous chemical precipitation in biological processes can utilize the advantages of biological and chemical methods for removing nutrients (Philips et al., 2003; Caravelli et al., 2010), and offer benefits in ease of operation, flexibility to changing conditions, and relatively small additional solids production (De Haas et al., 2000). Furthermore, parts of organic matter content in the influent can be utilized by the denitrifiers in the denitrification/flocculation tank, avoiding the problem of carbon source shortage occurring in biological processes implemented with a normal chemical pre-treatment step. To obtain sound treatment effectiveness and control operating costs, the selection of suitable dosed precipitants is essential for this proposed synergistic denitrification and chemical precipitation

Iron and aluminum salts are currently the most used precipitants in sewage treatment. Al salts are relatively costly, and residual Al in effluent would arouse concerns on the risk of neurologic diseases potentially associated with long-term exposure to Al in environments (Willhite et al., 2012). Ferric chloride and ferrous sulfate are the most used Fe salts. When dosed in oxic reactors, one principal disadvantage of using Fe2+ salt is the consumption of dissolved oxygen due to oxidation of Fe<sup>2+</sup> to Fe<sup>3+</sup>. Fe<sup>3+</sup> salt is however much more expensive than Fe<sup>2+</sup> salt. Additionally, the pH value of the mixed liquor in denitrification process is around pH 7.5, and the optimum pH ranges for phosphorus precipitation with Fe<sup>3+</sup> and Fe<sup>2+</sup> are between pH 4 and 5 and close to pH 8, respectively. Hence, if dosing Fe salts in the proposed synergistic denitrification and chemical precipitation process, pH condition would be favorable for Fe<sup>2+</sup> rather than Fe<sup>3+</sup> to precipitate phosphorus. Despite Fe<sup>2+</sup> is considered potentially a suitable precipitant being used in the proposed process due to its relatively low cost and high phosphorus removal efficiency under the typical pH condition of denitrification, an uncertainty still lies in the possible smaller size of Fe<sup>2+</sup> dosed sludge flocs compared to Fe<sup>3+</sup> dosed ones (Oikonomidis et al., 2010), which might adversely affect liquid-solid separation performance in the settling tank. To date, report on applying Fe<sup>2+</sup> salt in the pre-denitrification stage of BAF process to achieve simultaneous chemical precipitation of TSS and phosphorus remains few. To provide basic information for modifying a



**Fig. 1.** The schematic diagram of a modified BAF process with applying simultaneous chemical precipitation in pre-denitrification.

BAF process with employing simultaneous pre-nitrification and chemical precipitation, this study aims to examine the feasibility and suitability of using Fe<sup>2+</sup> salt as the precipitant dosed in the pre-denitrification stage of a BAF process. The effects of dosing Fe<sup>2+</sup> salt on effluent quality and sludge characteristics were monitored, and comparisons with the cases of no additional chemical dosing and dosing Fe<sup>3+</sup> salt were conducted to investigate any adverse effect introduced by dosing Fe<sup>2+</sup> salt.

#### 2. Methods

#### 2.1. Experimental setup

A series of laboratory scale experiments were conducted in 25 L rectors to examine the performance of dosing chemicals in the denitrification/flocculation tank shown in Fig. 1. Three sets of experiments were carried out (see Table 1). The reference experiment (R1) was performed to examine the performance of pre-denitrification without dosing any additional chemical. Another two sets of experiments were dosed with Fe<sup>2+</sup> salt (R2) and Fe<sup>3+</sup> salt (R3), respectively, to investigate the effects of applying chemical precipitation in the pre-denitrification process.

A synthetic domestic wastewater was used as the experimental influent. It was prepared by diluting methanol, KNO<sub>3</sub>, and KH<sub>2</sub>PO<sub>4</sub> stock solutions with deoxygenated tap water to simulate the mixed liquor of sewage influent and recycled BAF nitrified effluent, whose chemical oxygen demand (COD), NO<sub>x</sub>-N(NO<sub>3</sub>- $N + NO_2^- - N$ ), and total phosphorus (TP) concentrations, and pH value were within their typical ranges. Initial activated sludge used in the reactor was collected in a municipal wastewater treatment plant (Luofang Wastewater Treatment Plant, Shenzhen, China). The experiments were continuously carried out in cycles for more than 70 days. In each cycle, 18 L of the simulating solution was firstly fed into and mixed with the sludge already inside the reactor. For the experiments dosed with Fe<sup>2+</sup> or Fe<sup>3+</sup> salt, certain amount of FeSO<sub>4</sub> or FeCl<sub>3</sub> stock solution was added, respectively, to achieve a final Fe concentration of 24 mg/L. In a preliminary experiment (data not shown), this Fe concentration was justified sufficient to control the residual phosphorus concentration in the effluents of Fe dosed reactors (R2 and R3) to below 1 mg/L. The mixed liquor in the reactor was mechanically stirred for 9 h, and then settled down for 0.5 h for liquid-solid separation. Supernatant after liquid-solid separation was discharged. Prior to starting next cycle, the sludge remained in the reactor was rinsed by deoxygenated tap water for 5 times to minimize residual nutrient accumulation. The whole operation process for each cycle took about 12 h. About 1 L of the mixed liquor in the reactors, or the supernatant after liquid-solid separation were regularly sampled for monitoring effluent quality and sludge characteristics, respectively. Initial sludge concentrations of the experiments were about 1000 mg/L. Within the time span of the experiment, no excess sludge was discharged.

#### 2.2. Materials

Chemical stock solutions were prepared by dissolving reagent grade chemicals, methanol, KNO<sub>3</sub>, KH<sub>2</sub>PO<sub>4</sub>, FeCl<sub>3</sub>·6H<sub>2</sub>O, and FeSO<sub>4</sub>·7H<sub>2</sub>O, into ultrapure water (>18.1 M $\Omega$  cm) (deoxygenated for preparing Fe<sup>2+</sup> stock solution). All stock solutions were stored in 4 °C cold room. Fe<sup>2+</sup> stock solution was acidified by sulfuric acid to pH 3 prior to storage.

#### 2.3. Analytical methods

Water quality parameters are analyzed following APHA Standard Method (APHA, 1998). COD and alkalinity were analyzed by

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