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Fate and distribution of phosphorus in laboratory-scale membrane bioreactors

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a b s t r a c t

In this study, the removal efficiencies of phosphorus from synthetic wastewater using laboratory-scale membrane bioreactors (MBRs) with the addition of different iron salts were investigated. The distributions of phosphorus in the effluent, suspension, and sludge of the MBR systems after the addition of iron salts were analyzed. The removal efficiency of phosphorus in actual domestic sewage via the combination of MBR and Fe(II) was also investigated. The results indicated that after the MBR system effluent stabilized, the added Fe(II) was more efficient than Fe(III) was in removing phosphorus. Among the suspensions present in different zones of the MBR systems, the phosphorus concentrations varied significantly. Namely, the concentration of phosphorus in the first anoxic zone was the highest, for which the concentration of phosphorus in the MBR with added Fe(II) was higher than that in the MBR with added Fe(III). In addition, the percentage of the dissociable phosphorus in the sludge was relatively low. For the treatment of actual domestic sewage using the combination of MBR and Fe(II), a specific concentration of Fe(II) resulted in greater than 99% phosphorus removal efficiency as well as a stable effluent concentration. Furthermore, microbes present in the sludge exhibited better tolerance to Fe(II).

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1. Introduction

Membrane bioreactor (MBR) systems integrate traditional biological treatment and membrane separation technology, replacing the secondary sedimentation and sand filter tank of traditional biochemical treatment systems with membrane filtration. In MBRs, because of membrane separation, microbes are completely contained in the bioreactor, thereby realizing the separation of the hydraulic retention time (HRT) from the sludge retention time (SRT). As such, MBRs exhibit good treatment efficiency for pollutants and low sludge yield [\(Drews](#page--1-0) [and](#page--1-0) [Kraume,](#page--1-0) [2005;](#page--1-0) [Guo](#page--1-0) et [al.,](#page--1-0) [2012;](#page--1-0) [Hwang](#page--1-0) et [al.,](#page--1-0) [2008;](#page--1-0) [Le-Clech](#page--1-0) et [al.,](#page--1-0) [2006\).](#page--1-0) Although MBRs are efficientin removing organic compounds and nitrogen as compared to traditional technologies, they do not exhibit obvious advantages for the removal of phosphorus, because the longer SRT of MBRs can cause congestion via dead and inactive microorganisms ([Han](#page--1-0) et [al.,](#page--1-0) [2005;](#page--1-0) [Yang](#page--1-0) et [al.,](#page--1-0) [2011\),](#page--1-0) which reduces the sludge activity, and negatively affects phosphorus removal [\(El-Fadel](#page--1-0) et [al.,](#page--1-0) [2017\).](#page--1-0) To achieve effluent requirements, the addition of an Fe coagulant is a better alternative [\(Conidi](#page--1-0) [and](#page--1-0) [Parker,](#page--1-0) [2015;](#page--1-0) [Ferreira](#page--1-0) et [al.,](#page--1-0) [2008\).](#page--1-0)

By adding an iron-based coagulant, phosphorus removal was achieved by both biological and chemical methods using an MBR system. [Gutierrez](#page--1-0) et [al.](#page--1-0) [\(2010\)](#page--1-0) reported that the addition of polymeric iron significantly enhances the chemical removal of phosphorus in an MBR system through two routes. First, Fe^{3+} reacts with phosphate salts to generate insoluble salts. Second, iron undergoes hydrolysis after dosing. During this hydrolysis, various polymerization reactions occur, thereby generating multi-nuclear hydroxyl complexes with a long linear structure, which can remove phosphorus via neutralization, bridging, and flocculation. However, [Zhang](#page--1-0) et [al.](#page--1-0) [\(2008\)](#page--1-0) have reported that the removal effect of total phosphorous (TP) is not obvi-

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Fig. 1 - The schematic of the MBR systems with 1. anoxic zone (AN1, 5L), 2. aerobic zone (AO, 15L), 3. aerobic and facultatively anaerobic zone (AN-AO, 2L), 4. anoxic zone (AN2, 3L), and 5. membrane filtration zone (MF, 5L).

ous, attributed to two reasons: (i) the molar ratio of Fe to TP is 1:11, far from the optimum Fe/TP ratio of 1:1, and (ii) the one-batch addition of Fe is not beneficial for TP removal. [Yang](#page--1-0) et [al.](#page--1-0) [\(2011\)](#page--1-0) reported that the addition of polyferric chloride (PFC) to MBRs results in an increase in TP removal efficiency by 30%. Although iron-based coagulants can decrease the concentration of TP in the MBR effluent of a system with co-existing organic compounds, soluble microbial products, microbes, and sludge, studies about the fate of inorganic and organic phosphorus after the addition of iron-based coagulants have not been sufficiently investigated.

Hence, the impact of the addition of Fe(III) and Fe(II) on phosphorus removal efficiency in MBR systems during long-time testing was investigated in this study. We also examined the phosphorus distribution in three MBR systems which differ in feeding iron ions. It was expected that the findings of our study would provide an understanding of phosphorus transport in MBRs dosed with iron, which is recognized as a useful method for phosphorus control, as mentioned above.

2. Experimental methods

2.1. MBR system introduction

To investigate the changes in phosphorus removal efficiency after the addition of iron, three small-scale MBR systems were built in our laboratory. Fig. 1 is a schematic of these operating systems. The dimensions of the MBRs were 500 mm \times 150 mm, and the water level was 400mm. The MBRs were separated by baffle plates into an initial anoxic zone (AN1, 5L), an aerobic zone (AO, 15L), an aerobic and facultatively anaerobic zone (AN-AO, 2L), a second anoxic zone (AN2, 3L), and a membrane filtration zone (MF, 5L), from influent entrance to effluent exit. In the filtration zone, two identical hollow fiber membrane modules, composed of polyvinylidene fluoride, were perpendicularly immersed into the water. The bore diameter and total surface area of these fiber pipes were 0.04 μ m and 0.2 m², respectively. The bottom of the filtration zone was subjected to rapid aeration, in order to generate bubbles that were used for rinsing the fiber pipes. Under cross-flow conditions, membrane pollution was not serious, as every 10 days, the fiber pipes were cleaned using three steps: (i) purified water was used to remove the cake layer from the membrane, (ii) the membrane was soaked in dilute hydrochloric acid (10%, w/w) for 30min, and (iii) the membrane was immersed in citric acid (2%, w/w) solution for 30min ([Santos](#page--1-0) et [al.,](#page--1-0) [2015\).](#page--1-0) In the aerobic zone, a lower aeration rate (about $0.15 \,\mathrm{m^3/h}$) was applied to stir and oxygenate the wastewater. In both anoxic zones, blenders were used at 80 rpm for slow mixing. Suction pumps were

Table 1 – Operational parameters for the membrane bioreactors.

used to suck the effluent from the fiber pipes. The switching of the suction pumps was controlled via a computer to maintain an on/off ratio of 9:1 and an effluent flux of $15 \frac{\text{L}}{\text{m}^2}$ h for achieving an HRT up to 10h. The inflow water level was maintained at 400mm using an electromagnetic valve. The sludge from Jinan No. 1 Sewage Treatment Plant was sieved through a 0.1mm mesh and then used as the starting seed sludge, with a mixed liquor suspended solid (MLSS) lever of around 12 g/L. Two sludge recirculation periods were set: (i) from the aerobic and facultatively anaerobic zone to the first anoxic zone with a sludge recirculation ratio of 200%, and (ii) from the filtration zone to the aerobic zone, with a sludge recirculation ratio of 400%. At the set time every day, 1L of sludge was discharged from the filtration zone to maintain an SRT of 30 days. Table 1 summarizes the parameters of the MBR systems.

2.2. Impact of the addition of iron on the migration and transformation of phosphorus

2.2.1. Composition of the synthetic wastewater

For maintaining stability of the inflow water quality and facilitating data analysis, synthetic wastewater, prepared according to the AEESP laboratory manual [\(AEESP,](#page--1-0) [2001\),](#page--1-0) which was shown in [Table](#page--1-0) 2, was selected as the inflow. Glucose and sodium acetate were added in a molar ratio of 1:2 to maintain the pH of the wastewater at 7.0–7.5. The phosphorus content comprised 0.2mM of inorganic phosphorus $(KH_2PO_4$ and $NaH_2PO_4.2H_2O$) and 0.13 mM of organic phosphorus (yeast extract containing 1.5% of phosphorus). Trace amounts of ferrous iron (10 μ M) were added in the form of $(NH_4)_2Fe(SO_4)_2·6H_2O$ as the Fe nutrition source for microbes

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