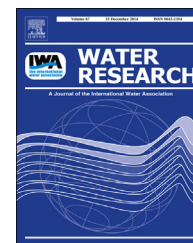


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Effect of ferric and ferrous iron addition on phosphorus removal and fouling in submerged membrane bioreactors

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ABSTRACT

The effect of continuously dosing membrane bioreactors (MBRs) with ferric chloride (Fe(III)) and ferrous sulphate (Fe(II)) on phosphorus (P) removal and membrane fouling is investigated here. Influent phosphorus concentrations of 10 mg/L were consistently reduced to effluent concentrations of less than 0.02 mg/L and 0.03–0.04 mg/L when an Fe(III)/P molar ratio of 4.0 and Fe/P molar ratio (for both Fe(II) and Fe(III)) of 2.0 were used, respectively. In comparison, effluent concentrations did not decrease below 1.35 mg/L in a control reactor to which iron was not added. The concentrations of supernatant organic compounds, particularly polysaccharides, were reduced significantly by iron addition. The sub-critical fouling time (t_{crit}) after which fouling becomes much more severe was substantially shorter with Fe(III) dosing (672 h) than with Fe(II) dosing (1200–1260 h) at Fe/P molar ratios of 2.0 while the control reactor (no iron dosing) exhibited a t_{crit} of 960 h. Not surprisingly, membrane fouling was substantially more severe at Fe/P ratios of 4. Fe(II) doses yielding Fe/P molar ratios of 2 or less with dosing to the aerobic chamber were found to be optimal in terms of P removal and fouling mitigation performance. In long term operation, however, the use of iron for maintaining appropriately low effluent P concentrations results in more severe irreversible fouling necessitating the application of an effective membrane cleaning regime.

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1. Introduction

Municipal wastewater usually exhibits total phosphorus (TP) concentrations in the range 4–12 mg/L (Rössle and Pretorius, 2001). While the activated sludge process is normally capable of achieving effluent P concentrations of 2–4 mg/L, or

0.5–1 mg/L if an enhanced biological phosphorus removal process such as Bardenpho is used, many countries are targeting effluent P concentrations of 0.01–0.3 mg/L in order to prevent eutrophication (Piekema, 2004). These low effluent concentrations are typically achieved by addition of inorganic salts such as ferric chloride or aluminium sulphate which, on addition to aqueous solutions in the circumneutral pH range,

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readily form high surface area particulate oxyhydroxides which have a very high P scavenging ability (Sedlak, 1991; Waite, 2002; Kim et al., 2008; Caravelli et al., 2010; Mao et al., 2012).

Membrane bioreactor (MBR) technology is widely viewed as being state of the art for municipal wastewater treatment due to the ability to achieve good contaminant removal whilst possessing small footprint and relatively low capital and operating costs (Judd and Judd, 2011). However, barriers to wider adoption of MBR technology still exist including a tendency for the membranes to irreversibly foul. This issue has been extensively investigated with results demonstrating that soluble microbial products (SMP) (which are considered to be dispersed in the supernatant) and extracellular polymeric substances (EPS) (which are considered to be attached to cells) are the most significant contributors to membrane fouling in MBRs (Lesjean et al., 2005; Rosenberger et al., 2005; Le-Clech et al., 2006; Drews et al., 2007; Metzger et al., 2007; Wang and Waite, 2008; Wu et al., 2008). SMP and EPS are the products of metabolic activity of microorganisms and consist of proteins, polysaccharides, lipopolysaccharides and lipoproteins and, in general, can be envisaged as complex biopolymer mixtures with a variety of functional groups (including carboxyl, amino and phosphate groups) (Le-Clech et al., 2006; Dizge et al., 2011a; Geluwe et al., 2011). Biomass flocs usually form a cake on the membrane surface since they are generally much larger than the membrane pore size. In the case of SMP and EPS, they may attach to and then accumulate on the membrane surface (Tansel et al., 2006; Metzger et al., 2007; Wang and Waite, 2008; Dizge et al., 2011a). Pore blocking may also occur since the sizes of SMP and EPS are typically comparable to or even smaller than that of the membrane pore size (Jiang et al., 2003; Wu et al., 2008).

In contrast to chemical phosphorous removal in conventional activated sludge (CAS) systems where the inorganic salts are typically added downstream of the activated sludge process but upstream of tertiary filters, in MBR processes iron or aluminium salts are typically added to the mixed liquor. Consequently, the logistics and control of chemical addition along with the mechanics and efficiency of chemical phosphorous removal are more complex in the MBR process compared to the CAS process. It has been reported that coagulant dosing into MBRs assists greatly in reducing the extent of transmembrane pressure (TMP) (Holbrook et al., 2004; Park et al., 2006; Wu et al., 2006; Fan et al., 2007; Koseoglu et al., 2008; Wu and Huang, 2008) with the reasons attributed to the reduction of organics in supernatant or the formation of larger flocs which may limit pore blockage. However, the amount of iron dosed in these studies was not intended for phosphorous removal and, given the low concentrations used, the results of these studies may not be particularly helpful in understanding the effects of the high concentrations of inorganic salt used when almost complete removal of phosphorus is the goal. The addition of inorganic salts such as ferric chloride for phosphorus removal is almost certain to influence the nature and extent of membrane fouling though the precise manner in which fouling will be modified is unclear. Fundamental studies on the effect of coagulants on membrane filtration performance indicate a link between floc formation, floc structure and cake permeability (Park et al., 2006; Mishima and Nakajima,

2009) with studies by Waite et al. (1999) suggesting that aggregates of large size and low fractal dimension resulted in formation of more permeable cakes than cakes formed from small, compact flocs. Wang and Waite (2010) observed that most of the iron added to MBRs is retained in the sludge and is presumably readily removed from the membrane surface by the ongoing bubbling and intermittent filtration that is used to prevent accumulation of material on the membranes. Indeed, the added iron salts may well act to coagulate the bacterial cells and, in so doing, render the assemblages more prone to removal by the hydrodynamic effects of bubbling. Alternatively, cationic Fe(III) species may act to bridge soluble microbial products in a similar manner to that observed for calcium cations (Wang and Waite, 2009) thereby aiding retention of SMP but, in so doing, creating a gelatinous matrix that causes severe membrane fouling (and which necessitates an increase in trans-membrane pressure in order to maintain an acceptable permeate flux). Particular dissolved organic molecules may form monomeric complexes with Fe(III) (depicted as Fe(III)SMP by Wang and Waite (2010)) while others may stabilise amorphous ferric oxide (AFO) in colloidal form with this material presumably able to penetrate but ultimately clog membrane pores.

In this study we investigated the impact of ferric chloride (Fe(III)) and ferrous sulphate (Fe(II)) addition on phosphorus removal and membrane fouling in MBRs to which molar ratios of Fe to P typical of those used for P removal in both conventional and membrane-based wastewater treatment (Metcalf and Eddy, 2003; Ganigue et al., 2011; Manzouri and Shon, 2011). Particular insight into the impact of i) use of either Fe(II) or Fe(III) salts, ii) dosing location and amount and iii) pH control on both phosphorus removal and membrane fouling performance is provided through these studies.

2. Materials and methods

2.1. Membrane bioreactors

Three bench-scale MBRs with a working volume of 30 L each were operated at 22 ± 1 °C, identical conditions for five months before iron coagulants were continuously added to two of the reactors (the third was maintained as a “no iron addition” control). Each MBR contains an anoxic zone (6 L), an aerobic zone (18 L) and a membrane compartment (6 L) separated by baffles (Fig. 1). Two identical modules of reinforced polyvinylidene fluoride (PVDF) hollow fibre membranes with nominal pore size 0.2 μm and total surface area 0.2 m^2 (Beijing Origin Water, China) were immersed vertically into the membrane zone with coarse bubble aeration used to limit membrane fouling. The effluent was withdrawn from the membrane modules using a suction pump (Masterflex, Cole-Parmer) in intermittent mode with on/off ratio of 9 min: 1 min. The flux was 16.7 $\text{L}/\text{m}^2 \text{ h}$ (LMH) corresponding to a hydraulic retention time (HRT) of 10 h. Transmembrane pressure (TMP) was continuously monitored by pressure transducers and the data recorded using Labview® (National instruments, USA). Filtration was interrupted when the normalised TMP (against baseline) reached 40 kPa because it was difficult to maintain the set flux at TMPs above this value. The membrane module was

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