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Simultaneous removal of phosphorus and foulants in a hybrid coagulation/membrane filtration system

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ABSTRACT

At many wastewater treatment plants, chemical addition is required to meet the requirements for discharge of phosphorus. Membrane filtration is an attractive option for removing the chemically precipitated phosphorus, but that process can be impeded by fouling of the membrane by effluent organic matter (EfOM). In the current study, membrane fouling and *ortho*-P removal were explored in hybrid adsorbent/membrane systems dosed with alum and/or a new aluminum-based adsorbent (heated aluminum oxide particles, HAOPs). HAOPs prevented fouling more effectively than alum did, even though the two adsorbents removed similar amounts of EfOM. The HAOPs were particularly effective when they were pre-deposited on the membrane (as opposed to being mixed into the feed upstream). On the other hand, alum removed *ortho*-P more efficiently than HAOPs did. Combining both adsorbents in a single membrane system takes advantage of the most attractive features of each, accomplishing good *ortho*-P removal while simultaneously minimizing fouling. In addition to being potentially applicable directly, the results demonstrate the potential benefits of using multiple coagulants and/or adsorbents in a single hybrid membrane system.

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1. Introduction

Due to concerns about the enrichment of receiving waters with nutrients and the resulting excessive growth of phytoplankton, phosphorus concentrations in wastewater discharges are tightly regulated. Phosphorus can be removed from wastewater either biologically or chemically. Conventional activated sludge treatment typically reduces effluent total phosphorus (TP) concentrations to 1–2 mg/L, and enhanced biological phosphorus removal processes can lower that value to 0.1–0.2 mg/L under ideal conditions (Blackall et al., 2002). Nevertheless, in practice, supplemental additions of chemicals such as Al or Fe(III) salts, poly-aluminum chloride (PACl), and/or lime are often required to maintain acceptable effluent TP concentrations.

When Al^{3+} or Fe^{3+} salts are added to P-containing wastewater, they form stable precipitates such as $\text{AlPO}_4(\text{s})$ (variscite) and $\text{FePO}_4(\text{s})$ (strengite) at pH between 5 and 7 (Jiang and Graham, 1998). At higher pHs, Ca^{2+} also precipitates phosphate efficiently in the form of $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2(\text{s})$, but this option is less attractive because of the large lime requirement for optimal TP removal (Marani et al., 1997).

Because Al^{3+} and Fe^{3+} react rapidly not only with phosphate, but also with hydroxyl ions, the molar ratio of added Al^{3+} or Fe^{3+} to TP must generally be greater than 1:1 to achieve efficient TP removal; for example, stoichiometries of $\text{Al}_{1.4}\text{PO}_4(\text{OH})_{1.2}$ and $\text{Fe}_{2.5}\text{PO}_4(\text{OH})_{4.5}$ have been found for the solids precipitated in such processes (Fytianos et al., 1998). It is also possible that a portion of the metal coagulants is first precipitated as hydroxides, and that phosphate ions then

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adsorb onto those solids (Galarneau and Gehr, 1997; Lijklema, 1980).

Membrane filtration can achieve excellent solid/liquid separation after biological treatment and/or coagulation processes (Gander et al., 2000), but fouling limits the water flux through the membrane, sometimes to the point that the process is economically unattractive. Such fouling can be induced by particles and colloidal material that accumulate at the membrane surface and block the membrane pores, or by effluent organic matter (EfOM), which can form a gel layer on the membrane surface either by itself or together with the rejected particles (Jarusutthirak et al., 2002). EfOM includes natural organic matter (NOM), soluble microbial products, and synthetic organic compounds.

We have recently shown that heated aluminum oxide particles (HAOPs), a novel adsorbent synthesized in our laboratories, can adsorb NOM effectively from drinking water sources (Kim et al., 2007a). Although the HAOPs could remove somewhat less NOM than powdered activated carbon (PAC) did, HAOPs treatment was more effective than PAC treatment at removing the fraction of the NOM that was primarily responsible for membrane fouling; we had previously reported similar results using heated iron oxide particles (HIOPs) as the adsorbent (Chang and Benjamin, 1996; Zhang et al., 2003). The current study compared HAOPs and alum addition as options for simultaneously removing phosphorus and membrane foulants from secondary-treated wastewater.

2. Materials and methods

2.1. Source of P-containing wastewater and clean water

Water for use in the tests was collected as a single grab sample of the permeate from a pilot-scale membrane bioreactor (MBR) treating municipal wastewater in an activated sludge process at the King County South Treatment Plant in Renton, WA. The MBR module (GE-Zenon ZeeWeed-500) contained membranes with a nominal pore size of 0.04 μm . The sample was stored at 4 °C and was brought to room temperature immediately prior to use in tests. The wastewater was at pH 7.45 and had an *ortho*-P concentration of 2.02 mg/L, a DOC concentration of 6.55 mg/L, and UV absorbance at 254 nm (UV_{254}) of 0.160 cm^{-1} . This water was used in all tests except one batch adsorption experiment in which a synthetic solution (described below) was used. In most experiments, the MBR permeate was used without any alteration, but in some tests, it was dosed with alum, and the resulting suspension was fed to the laboratory membrane system either directly or after 30 min of quiescent settling.

Deionized (DI) water (Millipore Milli-Q) was used to prepare standard solutions of *ortho*-P (from potassium dihydrogen phosphate, J.T. Baker) and DOC (from potassium acid phthalate, Integra Chemical Co.). All chemicals used in this study were of reagent quality.

2.2. Preparation for heated aluminum oxide particles (HAOPs)

HAOPs were synthesized by neutralizing 3M $\text{Al}_2(\text{SO}_4)_3$ to pH 7.0 and heating the resulting suspension in closed containers

at 110 °C for 24 h. This procedure, which is slightly modified from that used to prepare HIOPs in earlier investigations (Chang and Benjamin, 1996), partially dehydrates the solids while allowing them to retain a substantial fraction of their adsorption capacity. The mean diameter of the HAOPs was 10.4 μm based on particle size analysis (Saturn DigiSizer Model 5200, Micromeritics); their pzc was at pH 7.7 based on analysis of their electrophoretic mobility (ZetaPlus, Brookhaven Instrument Corporation); and their BET surface area was 116 m^2/g (FLOWSORB II 2300 System, Micromeritics). The mass fraction of aluminum (f_{Al}) in HAOPs that had been rinsed with DI water and then freeze-dried was 0.26. For comparison, the theoretical value of f_{Al} in Al_2O_3 is 0.53, and that in $\text{Al}(\text{OH})_3$ is 0.35; a solid with a chemical formula of $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ would have an Al fraction of 0.28.

2.3. Batch isotherm tests

Adsorption isotherms were developed by adding alum or HAOPs to 100 mL of the test water at doses up to 1 and 1.5 mM as Al, respectively, followed by pH adjustment to 7.5. The suspensions were mixed for 2 h at room temperature (~ 21 °C) and settled for 30 min, and the supernatants were filtered using 0.45- μm syringe filters (Millipore). *ortho*-P, DOC, and UV absorbance were then analyzed in the filtrate. An isotherm was also developed by applying the same procedures to a synthetic solution containing the same concentration of *ortho*-P as the MBR permeate, but no DOC. The synthetic solution was prepared using equimolar additions of Na_2HPO_4 and NaH_2PO_4 to DI water and then adjusting to pH 7.5 with NaOH.

2.4. Membrane filtration

The membranes used in the research were 47-mm-diameter disks, made of mixed cellulose esters (model VSWP04700, Millipore), with a nominal pore size of 0.025 μm and an effective area of 9.62 cm^2 . The feed to the membrane systems was the permeate from the MBR pilot unit, either unaltered or after treatment with alum. In the latter case, separate tests were conducted using the whole suspension and the supernatant after 30 min of settling. In some tests, HAOPs were pre-deposited on the membrane (i.e., before any feed was applied) at a mass/area ratio of 3 mg/cm^2 as HAOPs (29 $\mu\text{mol Al}/\text{cm}^2$). In all tests, the feed solution was continuously mixed with a magnetic stir bar. The permeate flux was maintained at 100 $\text{L}/\text{m}^2\text{h}$ using peristaltic pumps until the transmembrane pressure (TMP) increased to >200 kPa (~ 30 psi). TMP was measured online using a transducer (Omega, PX302-100GV).

2.5. Phosphorus, UV absorbance, and DOC analyses, and SEM images

ortho-P concentrations were measured using an AutoAnalyzer 3 (Bran+Luebbe, SEAL Analytical Ltd., UK), following the manufacturer's method US-696B-82, which closely approximates Standard Method 4500-P. UV absorbance was measured at 254 nm using a dual-beam spectrophotometer (Lambda-18, Perkin-Elmer GmbH., Überlingen, Germany).

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