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Hybrid ferrihydrite-MF/UF membrane filtration for the simultaneous removal of dissolved organic matter and phosphate

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ABSTRACT

Dissolved organic matter (DOM) and phosphorus promote microbial regrowth in water distribution networks. Ferrihydrite (Fh) has a high adsorption affinity with DOM and phosphate. Hence, a lab-scale unit of the hybrid Fh-MF/UF membrane filtration system was used to evaluate membrane fouling and the removal efficiency of DOM and phosphate. Suwannee River natural organic matter (SRNOM) was used as a surrogate for DOM in natural water. The Fh-membrane system demonstrated removal rates of dissolved organic carbon (DOC), UV_{254} and phosphate up to 50%, 80% and 90%, respectively, at the Fh dose of 17.5 mg/L. The effect of phosphate on the removal of DOM was different without or with the addition of Fh; namely, phosphate increased the DOM removal without Fh by interacting with the UF membrane made of regenerated cellulose (RC), but phosphate decreased the DOM removal by Fh due to the strong affinity of phosphate with Fh. Size exclusion chromatography revealed that phosphate mainly competed against smaller DOM molecules for Fh adsorption sites. Although the addition of Fh caused only a moderate flux decline with the RC membranes, the deposition of positively charged Fh on the surface of a negatively charged high-flux membrane, i.e., polyethersulfone (PES), caused a rapid decline of the permeation flux.

1. Introduction

Controlling microbial regrowth in water supply networks is a common problem in many water utilities around the world. In the literature, dissolved organic matter (DOM) is considered to be the limiting nutrient for bacterial regrowth (Hijnen, 2009). Phosphorus is an essential element for the growth, activity and regulation of microorganisms, including bacteria. No biofouling was observed at low phosphate concentrations in a reverse osmosis (RO) process, even at a high DOM concentration (Vrouwenvelder et al., 2010). Hence, phosphorus has also been reported to be the rate-limiting factor of microbial growth in water supply systems (Lehtola et al., 2002). Both DOM and phosphorus are present in most drinking water sources. Therefore, minimizing both inorganic nutrients

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(phosphate) and DOM is an effective strategy for controlling microbial growth in water supply networks.

Chemical coagulation is often applied prior to sand filtration or membrane filtration to enhance DOM removal in drinking water treatment. Activated carbon adsorption is also adapted to further remove DOM. However, DOM removal rates by these methods remain moderate due to the heterogeneity of DOM. The removal capacity of activated carbon for DOM remains low to moderate due to the larger molecular sizes of DOM compared to the pore sizes of activated carbon (Lin et al., 1999). Ferrihydrite (Fh) is an amorphous iron oxide particle (IOP) that has a large surface area and a high adsorption capacity for the phosphate, arsenate and organic matter present in soils, sediments and aquatic systems (Antelo et al., 2010). DOM can adsorb onto both crystallized, e.g., hematite and goethite, and amorphous IOP surfaces by ligand exchange between the acidic carboxylic and the phenolic groups of DOM and the hydroxyl groups of IOP to form inner-sphere complexes or by physisorption to form outer-sphere complexes (Gu et al., 1994; Ha et al., 2008). Numerous organic contaminants, including pesticides, herbicides and organic nutrients, have been reported to be efficiently removed by Fh from natural soil and waters (Jambor et al., 1998). On the other hand, the phosphate ions may form bidentate complexes with adjacent sites on the Fh (Gimsing and Borggaard, 2007). Therefore, Fh is expected to remove both phosphate and DOM simultaneously to control microbial regrowth in water supply networks. However, Fh must be separated from water after adsorption of DOM and phosphate.

Low-pressure membrane filtration processes such as microfiltration (MF) and ultrafiltration (UF), have many advantages, such as high removal efficiency of suspended solids and smaller footprints; therefore, they are widely used in drinking water treatment for the removal of pathogens and suspended particulate matter (Guo et al., 2010). However, low-pressure membrane filtration processes are not effective in removing inorganic nutrients and DOM, which can support bacterial regrowth and can form disinfection by-products (DBPs) (Chong et al., 2009; Richardson, 2003). Hence, a hybrid Fh-adsorption and MF/UF membrane filtration system is an attractive process for the simultaneous removal of nutrients, i.e., phosphate and DOM, and particulate matter, which is consistent with the recent trend of integrating pretreatment processes to low-pressure membrane systems (Huang et al., 2009). Cui and Choo (2013) used an IOP column as a pretreatment, before the use of a UF membrane, to control DOM. The integrated IOP/UF system promoted DOM removal and controlled membrane fouling. However, a hybrid system of Fh adsorption integrated into a MF/UF membrane filtration system for the simultaneous removal of DOM and phosphate has not yet been investigated.

Therefore, the aim of this study was to investigate the simultaneous removal of DOM and phosphate by a hybrid Fh-MF/UF system and to assess the effect of Fh addition on membrane fouling. Dead-end filtration experiments with and without the addition of Fh were performed to filter feed waters containing Suwannee River natural organic matter and phosphate. The removal efficiencies of SRNOM and phosphate and the permeation flux were monitored for different types of MF and UF membranes.

Table 1 – Characteristics of synthesized ferrihydrite (Fh).

Surface area (m^2/g)	Pore volume (cm^3/g)		Pore size (nm)	
	BET equation	BJH equation	BET equation	BJH equation
314.2	0.180	0.194	2.32	2.42

IUPAC classification: <2 nm:micropore, 2–50 nm: mesopore, >50 nm: macropore.

2. Materials and methods

2.1. Preparation and characterization of ferrihydrite (Fh)

The 2-line Fh was synthesized following the method reported by Patrizia et al. (2001). The concentration of Fh was measured by inductively coupled plasma atomic emission spectroscopy (ICP-AES, Optima 3000DV, PerkinElmer Inc., USA), if necessary, after dilution. Surface area and pore size distribution were characterized by the nitrogen adsorption method (Belsorp mini-II, Bel Japan Inc.), and the results are presented in Table 1. Scanning electron microscopy (SEM) (VE-8800, Keyence Corp., Osaka, Japan) revealed that the mean particle size of Fh was 8 ± 2 μm (Fig. 1).

2.2. Feed water

Suwannee River natural organic matter (SRNOM, RO isolation, 1R101N), purchased from the International Humic Substances Society (IHSS, St. Paul, MN, USA), was dissolved in Milli-Q water. The solution pH was adjusted to 7.5 by adding NaOH while being mixed using a magnetic stirrer. Afterwards, the solution was filtered through a 0.45-μm PTFE membrane (JH, Millipore, USA). The stock SRNOM solution had a dissolved organic carbon (DOC) concentration of 100 mg-C/L and was stored at 4 °C in the dark until use. In the membrane experiments, the initial concentration of DOC was kept at 2.5–2.8 mg/L based on the water quality of a river that is the source of water supply in Tokyo. A 100 mg/L phosphate stock solution was prepared by dissolving K_2HPO_4 chemical reagents and then added into the diluted SRNOM solution for the Fh-membrane filtration experiments.

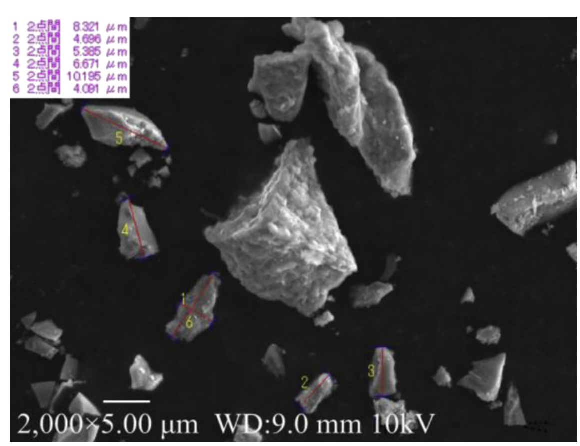

Fig. 1 – SEM image of Fh.