

## Effect of coagulation conditions on ultrafiltration for wastewater effluent

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**Abstract.** Low-pressure membrane filtration is increasingly used for tertiary treatment of wastewater effluent organic matter (EfOM), mainly comprising organic base/neutral compounds. In-line coagulation with underdosing, charge neutralization, and sweep floc conditions prior to ultrafiltration (UF) was studied to determine removals of the EfOM components and consequent reduction of fouling using polyethersulfone (PES) membranes. Coagulation and UF substantially reduced fouling for all coagulation conditions while removing from 7 to 38% of EfOM organic acids. From 7 to 16% of EfOM organic base/neutrals were removed at neutral pH but there was no significant removal for slightly acid coagulation conditions even though fouling was substantially reduced. Sweep floc produced the lowest resistance to filtration but may be inappropriate for in-line use due to the large added volume of solids. Charge-neutralization resulted in poor recovery of the initial flux with hydraulic cleaning. Under-dosing paralleled sweep floc in reducing hydraulic resistance to filtration (for sub-critical flux) and the initial flux was also easily recovered with hydraulic cleaning. HPO/HPI base/neutrals were identified on the fouled membranes but as previously reported the extent of fouling was not correlated with accumulation of organic base/neutrals.

**Keywords:** Effluent organic matter; in-line coagulation; ultrafiltration; fouling; membrane autopsy

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### 1. Introduction

Ultrafiltration (UF) or microfiltration (MF) is increasingly employed for treatment of wastewater, especially for separation of mixed liquor suspended solids in membrane bioreactors (MBR) or for pre-treatment prior to reverse osmosis in the context of water reuse. Fouling is often the limiting factor in UF or MF, and is defined here as an increase in resistance to filtration due to the accumulation of particles or solutes on the surface of the membrane or within the membrane pores.

Hydrophilic (HPI) base/neutrals and colloidal materials have frequently been implicated as the dominant foulant in natural organic matter (NOM). Koh et al. (2006) demonstrated that fouling of polyethersulfone (PES) UF membranes by lake water was decreased by pre-treating with polysulfone colloids. Authors attributed improved flux to removal of >30 kDa materials from the raw water. Fourier transform infrared (FT-IR) was used to demonstrate that HPI base/neutrals were dominantly adsorbed onto the polysulfone colloids, suggesting HPI base/neutrals were most responsible for fouling. Zularisam et al. (2007) used FT-IR to find that polysaccharide-like materials were most concentrated on fouled polysulfone membranes. Cho et al. (1998) also reported that HPI neutrals were important foulants in UF. Lee et al. (2001) reported that it was more difficult to remove hydrophilic than hydrophobic NOM from membranes, using acid or base cleaning procedures.

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Note: Paper to be submitted to "Membrane Water Treatment, An International Journal" for the purpose of Special Issue.

Colloids in the base/neutral fraction from conventional fractionation procedures have often been identified as the most important foulants in NOM. Carroll et al. (2000) reported that the HPI neutral fraction that caused most fouling of MF membranes also contained the highest concentration of >30 kDa colloids. Fan et al. (2001) reported that fouling of hydrophobic polyvinylidene fluoride (PVDF) MF was in the order HPI neutrals > hydrophobic (HPO) acids > transphilic acids  $\approx$  HPI charged fraction. Review of the literature discloses that fouling by HPI neutrals is linked to a high concentration of >30 kDa organic matter and  $\text{Ca}^{2+}$  in that fraction (Kim and Dempsey, 2012). Gray et al. (2008) also recently summarized experimental evidence that colloids in the base/neutral fraction of NOM are important foulants.

HPO acids have also been implicated as the dominant foulant for various membranes. Liang and Song (2007) found that organic fouling of MBR MF membranes decreased after removal of HPO acids but was not affected by removal of particulates. Hong and Elimelech (1997) reported that higher molecular weight (MW) humic fractions were most responsible for fouling hydrophobic membranes. Crozes et al. (1993) found that a HPO acid (tannic acid) was more strongly sorbed and produced much greater fouling of UF membranes compared to a HPI neutral (T10 dextran). Cho et al. (1999) reported that flux decline in UF was directly related to the aromaticity of NOM, as described by specific ultraviolet absorbance (SUVA). SUVA in NOM is typically highest for HPO acids. Aoustin et al. (2001) reported that humic acids produced rapid and irreversible flux decline while fulvic acid and the hydrophilic fraction of NOM produced smaller and mostly reversible flux declines. Lin et al. (2000) reported that the hydrophilic fraction of humic materials resulted in greatest flux decline for hydrophobic UF membranes.

Thus there is justifiable disagreement and confusion about the components of NOM or EfOM that are most responsible for fouling in UF or MF. Conclusions about the nature of organic foulants are usually based on operational definitions, e.g., many investigators have used the NOM fractionation protocol of Aiken et al. (1992) as modified by Carroll et al. (2000), initially adjusting to pH 2, sorption onto non-ionic resin beads with subsequent alkaline extractions (strongly HPO, weakly HPO, and transphilic fractions), adjust pH to 8 and elution through anion exchange resin beads (negatively charged fraction), leaving residual NOM in the water (HPI base/neutrals). Colloidal or large MW organic substances as well as most of inorganic dissolved constituents can pass through the columns of resin beads (Kim and Dempsey, 2012). In our work, we avoid manipulation of pH. We remove particles (>1  $\mu\text{m}$ ) and then colloids (>20 nm) by filtration processes, remove HPO/HPI acids using anion exchange resin gel, and then remove HPO base/neutrals using non-ionic resin beads, leaving only HPI base/neutrals.

Coagulation has commonly been identified as a successful pre-treatment for NOM or EfOM prior to membrane filtration (Chen et al., 2007; Guigui et al., 2002; Judd and Hillis, 2001; Lee et al., 2000). Fabris et al. (2007) showed that coagulation of lake water significantly decreased fouling of hydrophilic PVDF MF. Activated carbon removed dissolved organic carbon (DOC) but did not remove colloids and did not decrease fouling. Howe et al. (2006) concluded that coagulation of NOM decreased membrane fouling by removing material between 100 kDa and 1  $\mu\text{m}$ . Bose and Reckhow (1998) used an extended fractionation strategy and studied adsorption of NOM onto aluminum hydroxide flocs. The humic substances were most strongly adsorbed; percent sorbed was positively correlated with SUVA and molecular size and removal of HPO/HPI acids was inversely related to the negative charge density. These results are consistent with many other investigations that have shown that coagulation was most successful in removing NOM with high MW and SUVA, especially humic materials. Haberkamp et al. (2007) studied coagulation of EfOM prior to UF. They reported that coagulation removed bio-colloids and humic materials better than low MW acids and low-MW neutrals were hardly removed (EfOM characterization based on HPSEC). Differences in the removal of NOM were found between charge neutralization and sweep floc conditions (Chow et al., 2004). Pikkarainen et al. (2004) showed that specific cake resistance decreased with increasing coagulant dose, associated with increased removal of