



Coagulation and oxidation for controlling ultrafiltration membrane fouling in drinking water treatment: Application of ozone at low dose in submerged membrane tank



Wenzheng Yu^{*}, Nigel J.D. Graham^{**}, Geoffrey D. Fowler

Department of Civil and Environmental Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, UK

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ABSTRACT

Coagulation prior to ultrafiltration (UF) is widely applied for treating contaminated surface water sources for potable supply. While beneficial, coagulation alone is unable to control membrane fouling effectively in many cases, and there is continuing interest in the use of additional, complementary methods such as oxidation in the pre-treatment of raw water prior to UF. In this study, *the application of ozone at low dose in the membrane tank* immediately following coagulation has been evaluated at laboratory-scale employing model raw water. In parallel tests with and without the application of ozone, the impact of applied ozone doses of 0.5 mg L⁻¹ and 1.5 mg L⁻¹ (approximately 0.18 mg L⁻¹ and 0.54 mg L⁻¹ consumed ozone, respectively) on the increase of *trans*-membrane pressure (TMP) was evaluated and correlated with the quantity and nature of membrane deposits, both as a cake layer and within membrane pores. The results showed that a dose of 0.5 mgO₃ L⁻¹ gave a membrane fouling rate that was substantially lower than without ozone addition, while a dose of 1.5 mgO₃ L⁻¹ was able to prevent fouling effects significantly (no increase in TMP). Ozone was found to decrease the concentration of bacteria (*especially the concentration of bacteria per suspended solid*) in the membrane tank, and to alter the nature of dissolved organic matter by increasing the proportion of hydrophilic substances. Ozone decreased the concentration of extracellular polymeric substances (EPS), such as polysaccharides and proteins, in the membrane cake layer; the reduced EPS and bacterial concentrations resulted in a much thinner cake layer, although the suspended solids concentration was much higher in the ozone added membrane tank. Ozone also decreased the accumulation and hydrophobicity of organic matter within the membrane pores, leading to minimal irreversible fouling. Therefore, the application of low-dose ozone within the UF membrane tank is a potentially important approach for fully mitigating membrane fouling.

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1. Introduction

Ultrafiltration (UF) membrane systems are becoming an increasingly important alternative for the treatment of surface and sub-surface waters since they can achieve a consistently high quality of drinking water economically. However, natural organic matter (NOM) is often a major foulant during the ultrafiltration of surface water (Yuan and Zydny, 2000), and the extent of the fouling is dependent on the nature of the NOM, such as the relative presence of humic substances (Combe et al., 1999) and biopolymers

(polysaccharides and proteins) (Lin et al., 2000; Laabs et al., 2006; Huang et al., 2007), commonly referred to as extracellular polymeric substances (EPS). EPS are associated with microorganisms and may be produced extensively by suspended cells and biofilms. These macro-compounds tend to attach strongly to membrane surfaces and may cause pore blockage, particularly when separated from their bacterial cells (Pacton et al., 2011).

The importance of EPS in membrane bioreactor (MBR) fouling was reported by Cho and Fane who showed that fouling occurred in two stages with firstly, a gradual deposition of particles, and then subsequently a rapid stage of biomass growth that required membrane cleaning (Cho and Fane, 2002). Amy and Cho identified polysaccharides as dominant foulants in UF and nanofiltration (NF) treatment of surface water, even though polysaccharide concentrations in surface waters were comparatively low (Amy and Cho,

^{*} Corresponding author.

^{**} Corresponding author.

E-mail addresses: w.yu@imperial.ac.uk (W. Yu), n.graham@imperial.ac.uk (N.J.D. Graham), g.fowler@imperial.ac.uk (G.D. Fowler).

1999). According to the results of Kuhl and Jorgensen, glyocalyx (EPS) may function as a sponge and adsorb nutrients present in very low concentrations in the aquatic phase, thereby enabling microbial biofilms to grow in very low nutrient environments such as ultrapure water systems (Kuhl and Jorgensen, 1992). Therefore, it is likely that EPS exists in the membrane system for treating surface waters and contributes to fouling.

Pre-treatment involving chemical coagulation or coagulation–hydraulic flocculation has been shown to be an effective and low-cost approach for improving overall water quality, and for reducing membrane fouling (Peiris et al., 2013; Kimura et al., 2014; Wray et al., 2014); this has also been reported for pre-treatment using electrocoagulation (EC) (Wert et al., 2011). Although the application of coagulation before membrane filtration can reduce bacterial and EPS concentrations prior to the flow reaching the membrane, such concentrations within the membrane tank invariably increase with operating time together with residual flocs, leading to the accumulation of bacteria and EPS on the membrane. This is a general phenomenon and has been shown to occur with different membrane materials (Vanderkooij et al., 1995). In order to control the accumulation of bacterial and EPS concentrations, the application of chemical oxidation is a potentially effective approach.

A range of specific chemicals, namely ozone, chlorine, chlorine dioxide, hydrogen peroxide and potassium permanganate, are powerful and effective oxidants that have been widely applied and studied in water treatment (Chen et al., 2003; Yu et al., 2014). However, most of these have undesirable secondary effects in their use; for example chlorine is associated with the production of halogenated by-products, and permanganate increases sludge production and the risk of elevated, residual Mn concentrations. In contrast, ozone has relatively fewer side-effects and its application to membrane processes has been considered by several researchers previously (Park et al., 2012).

Ozone, when applied solely as a pretreatment (i.e. without coagulation), was able in some cases to reduce membrane fouling (Wu and Huang, 2010; de Velasquez et al., 2013), while others found ozonation caused a significant degradation of biopolymers that led to a minor reduction in flux for both UF and microfiltration (MF) systems (Wang et al., 2007; Zhu et al., 2010; Sun et al., 2011; Filloux et al., 2012). In further studies, ozone treatment was effective at degrading colloidal NOMs (or biogenic colloids) which are likely to be responsible for the majority of membrane fouling (Lehman and Liu, 2009; Barry et al., 2014; Zeng et al., 2014), and membrane fouling (sheet ceramic microfiltration) could be mitigated by ozonation combined with hydrogen peroxide (Karnik et al., 2005b; Szymanska et al., 2014) or using ozonated water for backwashing (Fujioka and Nghiem, 2015).

The combination of ozonation with other pre-treatments has been shown to enhance the removal of contaminants and the reduction of membrane fouling, and examples include the following: ozone with iron oxide nanoparticles (Karnik et al., 2005a), ozonation-biological activated carbon (BAC) filtration (treating activated sludge (AS) effluent) (Nguyen and Roddick, 2010), ozonation-powdered activated carbon (PAC) (removal of aromatic DOC and other organic matter in drinking water) (Treque et al., 2010) and ozonation-coagulation (where flux decline was reduced with increasing coagulant concentration and increasing ozone dosage) (Genz et al., 2011).

In order to control the concentration of bacteria in the membrane tank, the combination of ozone, as an oxidant-disinfectant, and coagulation may represent a superior pre-treatment arrangement. However, the application of ozone prior to coagulation may be non-ideal since organic matter after oxidation may be difficult to remove by coagulation, resulting in a greater adsorption of organic

matter on the membrane surface and pores, inducing a significant increase in irreversible fouling (Genz et al., 2011). To avoid these effects, ozone can be applied after the coagulation stage and immediately prior to the UF membrane (CUF-O₃), where it can have a direct influence on biofouling processes. While conventional water treatment by coagulation–sand filtration–disinfection processes can remove a large proportion of the influent impurities (e.g. bacteria, particles and organic matter), some contaminants are only poorly removed by this process, such as Giardia and Cryptosporidium, and particular viruses. However, treatment by CUF-O₃ can remove these protozoa spores and most viruses through enhanced particle separation and disinfection. This arrangement has not been reported previously, possibly because of concerns that ozone may damage the integrity of the membrane (Shanbhag et al., 1998). However, polyvinylidene fluoride (PVDF) membranes have been reported as being able to resist any significant adverse effects of ozone, which confirmed that an ozone–microfiltration system could be used for >5 years at low ozone concentrations (Mori et al., 1998). For the investigation described in this paper, PVDF hollow-fiber membranes were employed and the results, as described subsequently, have shown that the application of ozone at low dose within the UF membrane tank, after coagulation, substantially prevented external and internal membrane fouling and was highly beneficial to the overall operation of the process.

2. Materials and methods

2.1. Model raw water and coagulant

A model raw water was employed in the tests in order to simulate a surface water supply and to provide sample consistency and reproducibility for the extended period of testing (~60 days). This was prepared by adding a small quantity of domestic sewage effluent to the local (London, United Kingdom) tap water in a volumetric ratio of 1:50, and 5 mg/L Suwannee River Humic Acid (2S101H, International Humic Acid Substance Society, USA). The addition of domestic sewage effluent and humic acid represents organic matter which typically is difficult, and easy, to be removed by coagulation, respectively, and the effluent provided background levels of microorganisms that would be expected in surface waters impacted by urban runoff and effluent discharges. Prior to mixing with domestic sewage effluent and humic acid solution, the tap water was left over night to ensure the complete decay of residual chlorine. The characteristics of the model raw water are listed in Table 1, and during the course of the experimental programme the temperature of the water was maintained constant at 20 ± 2 °C.

2.2. The treatment processes of ultrafiltration systems

A schematic illustration of the experimental set-up involving the coagulation–UF processes without, and with, the addition of ozone in the membrane tank (CUF, CUF-O₃, respectively), operated in parallel, is given in Fig. S1. Model raw water was fed into a constant-level tank to maintain the water head for the membrane tanks. A constant dose of Al₂(SO₄)₃ (0.15 mM, calculated as Al) was continuously added into the rapid mixing units; the alum dose corresponded to near zero zeta potential of the resulting flocs. The rapid mix speed was 200 rpm (184 s⁻¹) in the mixing units with a hydraulic retention time (HRT) of 1 min, which then reduced sequentially to 100 rpm (65 s⁻¹), 80 rpm (46.5 s⁻¹) and 50 rpm (23 s⁻¹) in the three continuous flow flocculation tanks, each having a HRT of 5 min. After the flocculation tanks, the flow passed directly into the membrane tanks. Each tank contained a submerged PVDF hollow-fiber UF membrane module (Tianjin Motimo Membrane Technology Co., Ltd, China) with a nominal membrane

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