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Effect of oxidation with coagulation and ceramic microfiltration pre-treatment on reverse osmosis for desalination of recycled wastewater

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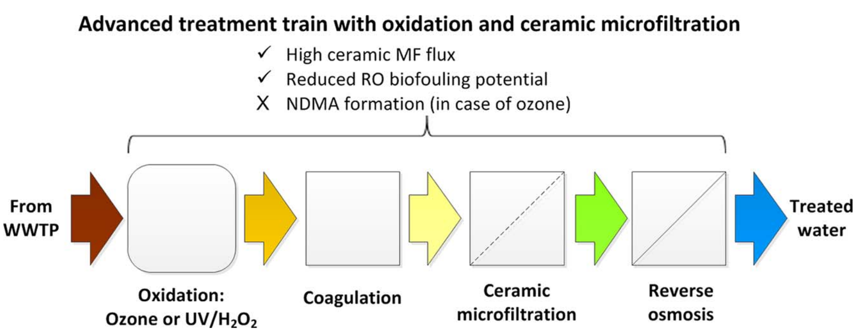
GRAPHICAL ABSTRACT



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ABSTRACT

Oxidation and coagulation before ceramic microfiltration (CMF) greatly increases membrane flux, but is unconventional for reverse osmosis (RO) pre-treatment. Impacts to RO and the wastewater recycling scheme operating CMF at high flux conditions is little understood. In this work, wastewater was treated with ozone or ultraviolet/hydrogen peroxide (UVH) oxidation, coagulation, then CMF, to explore RO membrane performance at bench scale. Sustainable high CMF fluxes were confirmed using coagulation with either ozone or UVH. Uniquely for ozone, dosing 13 mg-O_3/L for 15 min greatly increased toxic by-product *N*-nitrosodimethylamine (NDMA) to 33 ng/L. Dosing chloramine (common for RO biofouling control) added only up to 7 ng/L NDMA. RO tests on all pre-treated waters showed little variation to flux but oxidation significantly altered texture of RO fouling material from smooth and dense to porous and granular. Biofouling studies with model bacteria strain RO 22 (*Pseudoalteromonas* spp) showed higher organic biodegradability but biofilm analysis revealed ozone-coagulant-CMF greatly limited extension of bacteria communities from the membrane surface suggesting oxidation reduces RO biofouling. The novel findings of reduction of RO biofouling risk with oxidation and

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coagulation for high flux CMF pre-treatment identified in this work need to be demonstrated on different wastewater types over longer term.

1. Introduction

Ceramic membranes are an alternative technology to polymeric membranes for water treatment offering superior physical integrity, chemical resistance, higher flux, and longer life [1]. However their application as a pre-treatment for reverse osmosis (RO) desalination of wastewater is unconventional. In considering ceramic membranes, high flux is important to offset their higher material cost but must be operated in a specific way to achieve this, which would impact the downstream RO plant operation. For example Dow and co-workers demonstrated that the sustainable ceramic microfiltration (CMF) membrane fluxes for treating clarified wastewater increased 2–3 fold in response to dosing with the common coagulant polyaluminium chloride (PACl) [2,3]. Coagulation used prior to polymer membranes is already known to reduce fouling as well as to remove organic matter, particularly the large molecular weight (MW) components, being biopolymers and humic substances [4–6]. Fan et al. [7] concluded that coagulation treatment reduced organic fouling by removal of these larger-sized materials. Further, ozone used in conjunction with coagulation and ceramic membranes was observed to work together to provide > 4-fold sustainable flux increases for ceramic membranes [2]. Oxidation processes such as ozonation, and ultraviolet irradiation (UV), are commonly practised as the tertiary treatments to meet appropriate water quality in reclaimed water from secondary wastewater treatment plant (WWTP) effluents for disinfection purposes, odour treatment as well as the removal of colour caused by humic substances. With their wider use in water treatment, researchers have more recently considered their specific impact on water organic fractions [6,8] and membrane fouling [9], which is particularly useful for explaining why such high ceramic membrane fluxes can be achieved.

Studies conducted using ozone-resistant polyvinylidene fluoride (PVDF) and polysulfone (PS) membrane materials showed that using ozonation upstream of the membrane did enhance the permeate flux and reduce membrane fouling by the degradation of high molecular weight natural organic matter [10–13]. More recently, a study on polymer ultrafiltration (UF) membranes found that the mechanisms are more complex, where ozone reactions with bovine serum albumin (BSA) led to increased fouling, while reactions with alginate led to reduced fouling [9]. On top of altered organics chemistry, theories around the role of ozone regarding its ability to greatly enhance flux have focused on the role of highly reactive hydroxyl radicals ($OH^{\bullet}$) formed by the catalytic breakdown of ozone on the ceramic membrane surface [14].

So in the case of upstream oxidation where membranes benefit in terms of performance, there is a clear alteration of the chemical properties of the water borne compounds that will impact other downstream processes. In the case of saline wastewater, low pressure membranes are widely applied prior to reverse osmosis (RO) as a pre-treatment. Normally oxidation would be applied in a water recycling scheme downstream of RO, however, it is generally understood that the mechanisms to increase hydrophilicity of organics in wastewater would be useful in controlling RO membrane fouling. Such benefits including minimising cleaning and membrane replacement, and reduced energy requirements due to reduced RO fouling, were explored in a dedicated study [15]. Membrane bioreactor (MBR) effluent was fed directly to a dual train pilot RO system with one train featuring an ozone stage, while the other fed directly by MBR permeate. The reduction to membrane fouling was demonstrated over 3000 h of testing, showing reduced membrane permeability deterioration suggesting longer term benefits to RO membranes in terms of longevity, reduced cleaning costs and lower energy requirement [15]. Without ozone, RO flux declined by 12% while with ozone only declined by 6%. Similar beneficial effects were reported at bench scale [16]. Recent work on application of ozone and CMF followed by biologically active filtration upstream of RO for water recycling application found uniquely that RO foulants after ozone and CMF were easily removed with water rinsing [17]. This promising finding shows that in the case when ozone is applied upstream, reduced cleaning maintenance of the RO membranes is expected. The process was subsequently adopted for a 9 month potable reuse trial [18,19]. However, these used biological processes after oxidation, may not be necessary to apply prior to RO.

Oxidation (i.e., ozone or UV) in practice is typically followed by biological filtration. Ozone breaks down larger molecular weight organic matter increasing the assimilable organic carbon proportion, favouring micro-organism growth [20]. The study by Nguyen and Roddick highlighted that the ozonation of the raw activated sludge effluent produced biodegradable dissolved organic carbon (BDOC), and biological activated carbon (BAC) filter did not completely remove those compounds [21]. Thus it is uncertain if deliberate use of BAC to prevent biofouling of downstream RO membranes would be effective. Recent work has shown that ozone and BAC application prior to ceramic membranes can have a negative impact to CMF performance compared to ozone on its own [22] suggesting that despite the BDOC removing ability of BAC, it is not useful for high CMF performance and could be avoided for pre-treatment to RO.

Disinfection by chloramines is generally practised prior to the RO process to prevent the membrane from biofouling in a conventional RO-based water recycling application [23]. Hence, despite the increase in biodegradability of organics due to ozone, the application of chloramine may assist in controlling biofouling. However, the use of chloramines can lead to the formation of disinfection by-products (DBPs), especially nitrogen-containing DBPs such as *N*-nitrosodimethylamine (NDMA) and other *N*-nitrosamine compounds [24]. On top of this, ozone is also well-known to form NDMA as a result of the oxidation of NDMA precursors [25–28]. NDMA is an important concern if the intended use of the water is limited by this compound, e.g., potable reuse. A study on ozone application upstream of RO should consider use of chloramine disinfectant and the formation of NDMA.

Therefore, it still remains unknown of the viability of using the high CMF flux arrangement (with oxidation and coagulation) as a pre-treatment to RO for saline wastewater recycling purpose, particularly in the case where no post-oxidation biological treatment stage (e.g. BAC) is used. At the same time, working towards understanding differences in RO membrane fouling (both organic and bio) of this non-traditional water recycling process compared to the more traditional approach (without oxidation prior to RO) is of more fundamental interest. Addressing these points forms the more novel feature of this work. This study therefore has the following objectives 1) to confirm reported high flux performance when ceramic membranes are coupled with coagulation, ozonation and UV/H_2O_2 (UVH) treatment and their combinations; 2) to demonstrate the impact of the pre-treatment processes on water quality including formation of a well-known wastewater disinfection by-product, NDMA; 3) to test the influence of the pre-treatment options on RO membrane performance; and 4) to determine the potential for biofouling on the downstream RO membranes. The source water collected from a full-scale water recycling plant was used for the purpose of this work.