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Investigating the relative contribution of colloidal and soluble fractions of secondary effluent organic matter to the irreversible fouling of MF and UF hollow fibre membranes

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ABSTRACT

Effluent organic matter (EfOM) matrices were modified by applying physical chemical treatments on biologically treated wastewater effluents in order to identify the EfOM fraction(s) responsible for irreversible fouling of low-pressure membranes.

Anion exchange resin (AER) adsorption preferentially removed humic-like structures, while oxidation processes, such as ozonation and H_2O_2/UV resulted in the breakdown of high molecular weight (MW) structures into lower MW compounds.

After re-concentration to the original organic carbon content, the fouling potential of the modified EfOM matrix was investigated using multi-cycle filtration tests performed with commercially available microfiltration (MF) and ultrafiltration (UF) membranes.

The pre-oxidized effluents showed low fouling potential regardless of the membrane used. The results confirmed the major role of high MW biopolymers on MF and UF total fouling. Results from MF experiments highlighted that AER treated effluents exhibited similar fouling properties as the untreated effluent indicating that humic-like substances do not significantly affect MF fouling and confirm again that biopolymers is the fraction responsible for fouling. The same tests performed with UF membrane showed a lower irreversible fouling with AER treated effluent suggesting the strong contribution of humic fractions to irreversible fouling. These findings were supported by the results obtained with two different secondary effluent matrices, a conventional activated sludge treated effluent and a membrane bioreactor supernatant.

1. Introduction

Wastewater reclamation plants mainly use biological processes as secondary treatment for the removal of suspended solids, organics and nutrients followed by membrane filtration as tertiary treatment in order to produce high quality water suitable for reuse and/or reclamation purpose. Suspended solids form cake layers that are likely reversible fouling and can be removed through backwash cleaning, while biofouling can be controlled using chlorine or monochloramine. Then the major issue of the tertiary treatment is irreversible membrane fouling. Effluent organic matter (EfOM), including colloidal and soluble compounds, is ubiquitously present in biologically treated wastewater effluent and exerts strong fouling properties towards microfiltration (MF) and ultrafiltration (UF) membranes [1,2]. Reversible and hydraulically or chemically irreversible fouling can occur [3]. As maintenance and replacement of membranes are costly, it is important to identify the EfOM fraction(s) responsible for reversible and irreversible fouling in order to improve membrane performance.

Biopolymers, such as polysaccharides and proteins were identified as major foulants in drinking water and wastewater membrane applications [4,5]. Some studies pointed out the major role of proteins in promoting stronger adsorption phenomena [6,7]. There is also speculation about the importance of humic substances with respect to their fouling potential. Whereas studies

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often reported that humic substances play a minor role in fouling issues observed in drinking water membrane units [5,8,9], some studies conducted with wastewater effluent or model substances emphasized a synergic effect between biopolymers and humic substances [5,10,11]. The contradictory results can be explained by the different nature of terrestrial humic substances and those produced in biological reactors [7,12].

In a previous publication, Filloux et al. applied physical-chemical treatments to a biologically treated wastewater effluent with the objective to modify the composition of the organic matrix and better understand the role of the different EfOM fractions on low-pressure membrane fouling [13]. Anion exchange resin (AER) sorption removed humic-like structures, while ozonation induced a significant degradation of the biopolymers due to the breakdown of high molecular weight (MW) structures into lower MW compounds. The impact of the EfOM characteristics on low-pressure membrane fouling was assessed based on a single filtration test at constant flux, using tailor-made hollow fibre modules. Results indicated that biopolymers play a major role in low pressure membrane fouling; intermediate and low MW compounds have minor impact. Taking into account the importance of membrane properties, the use of manufactured hollow fibre membranes operated at constant flux with real wastewater matrices was shown to be a valid methodology to identify the foulant fraction of EfOM. However, this previous work only assessed total fouling. The reversible/irreversible character of the fouling was not studied.

Studies conducted to understand fouling mechanism at lab-scale are mainly performed with flat-sheet membrane coupons under constant pressure mode and using single-cycle filtration tests [5,14]. This approach is very different from full-scale operation conditions (i.e., constant-flux mode with regular backwash sequences) and presents major limitations when investigating the reversibility of the fouling. Crittenden et al. proposed a model referring to the resistance in-series for the interpretation of flat-sheet membrane experimental data [15]. Recently, lab-scale experiments were reported using hollow fibre membranes to improve cleaning strategies [16], study the impact of pre-treatment [17], understand fouling mechanisms [18] and optimize filtration operating conditions [19]. Experimental set-up allowing backwash operations were used and new fouling indices were proposed to take into account the reversibility and non-reversibility of fouling processes [3,20,21].

In this study, we apply our previously published methodology modifying EfOM properties [13] prior to multi-cycle filtration tests in order to identify the major organic fraction responsible for irreversible fouling. The impact of physical chemical treatments (i.e., AER, ozone and H_2O_2/UV as a source of hydroxyl radicals) on the reversibility of membrane fouling was assessed for secondary effluents of different origin and EfOM composition. Details on EfOM character changes, specific flux and backwash efficiency (i.e., reversible versus irreversible fouling) are discussed. Because the main objective was to significantly modify the character/structure of the organic matrix, the experimental conditions used in this investigation could be different from industrial applications.

2. Materials and methods

2.1. Wastewater effluents

Three biologically treated wastewater effluents from different locations subjected to different process conditions were used in this study:

- SJA-SE effluent, collected from the secondary clarifier of a municipal wastewater treatment plant.
- Bu-SE effluent, collected from the final effluent of a municipal wastewater treatment plant.
- Ban-MBR-S supernatant, collected from the supernatant of a membrane bioreactor (MBR) at a municipal wastewater treatment plant.

Table 1
Main characteristics of the wastewater effluents (n = 4–20).

Name	SJA-SE	Bu-SE	Ban-MBR-S
DOC (mgC/L)	6.8 ± 0.9	9.3 ± 0.4	7.0 ± 0.3
UV_{254nm} (1/cm)	0.21 ± 0.01	0.25 ± 0.06	0.14 ± 0.02
SUVA (L/mgC m)	3.0 ± 0.4	2.7 ± 0. 7	2.1 ± 0.2
TP (mg/L)	1.9 ± 0.8	0.6 ± 0.6	0.2 ± 0.1
NH_4^+ (mg/L)	0.9 ± 0.2	0.3 ± 0.6	3.5 ± 0.7
NO_3^- (mg/L)	1.5 ± 1.9	1.6 ± 0.3	0.6 ± 0.3
Conductivity (μS/cm)	1163 ± 40	884 ± 59	741 ± 35
pH	8.0 ± 0.2	7.4 ± 0.6	6.9 ± 0.2

Ban-MBR-S supernatant refers to the MBR supernatant (i.e., the non-settled fraction of the MBR biomass) that was subjected to 4000*g* centrifugation for 10 min (Beckman Coulter ALLEGRA X12, USA). The characteristics of the water samples are summarized in Table 1. All water samples were pre-filtered using glass fibre cartridge filters with 10 μm pore size (Millipore, USA) before MF and UF experiments in order to remove the large particles and focus the study on the soluble and colloidal fractions.

2.2. Physical chemical treatments applied for EfOM structural modification

The wastewater effluents were subjected to three different physical chemical treatments, i.e., AER sorption, ozonation and advanced oxidation process using H_2O_2/UV. Sorption treatments were performed using a strong base AER (i.e., polyacrylate-based AER with a total capacity of 0.86 eq/L). AER treatments were performed in batch mode under magnetic stirring using 5 mL/L of resin. Ozonation was applied at high dose of 5.5 mgO_3/mgC on the SJA-SE effluent to modify the hydrophobic/hydrophilic distribution of the TOC. Additional characteristics of the AER and ozonation treatment protocols are given in Filloux et al. [13].

UV oxidation experiments performed with and without H_2O_2 addition were conducted in a photo-reactor (LC-20 model, UVTA Pty. Ltd., Australia) with a 1.6 L/min recirculate flow on Bu-SE effluent and Ban-MBR-S supernatant. The description of the experimental device can be found elsewhere [22,23]. During the irradiation, the solution (1.5 L) was continuously stirred using a magnetic stirrer (250 rpm) and re-circulated through the photo-reactor to achieve a uniform UV dose distribution. UV doses from 0 to 3000 mJ/cm^2 were applied by using a low pressure mercury lamp emitting at 253.7 nm (UV-C, 60 W electrical power, 50 Hz, 240 V). Hydrogen peroxide concentrations between 0 and 100 mg/L were obtained from the dilution of a 30% w/w H_2O_2 solution. The H_2O_2 concentration was determined according to the metavanadate method as described in Nogueira et al. [24]. As for ozonation, the objective of performing advanced oxidation was to modify the molecular weight distribution of EfOM with minimal TOC reduction. A hydrogen peroxide dose of 25 mg/L with a UV dose of 1500 mJ/cm^2 was chosen for the filtration experiment. The results of the respective preliminary experiments and the impact of H_2O_2/UV treatment on EfOM characteristics are shown in supplementary information (SI), part A.

All dosages were selected based on preliminary experiments (not shown) where the objective was to significantly modify the EfOM characteristics. These treatment conditions should be viewed