

EXPERIMENTAL STUDY ON THE MINIMIZATION OF FOULING BY NOM IN MF/UF USING SUBMERGED "SINGLE" CAPILLARY MEMBRANES

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ABSTRACT

This paper looks into the problem of fouling of UF/MF membranes due to NOM present in surface waters. NOM can be divided into particulate, colloidal and dissolved fractions. The idea here is to fractionate the surface water into the above-mentioned fractions, and to test the fouling characteristics of each fraction on a MF membrane, with and without pre-treatment by coagulation. Coagulation was performed at a pH of 7.5, using poly aluminium chloride (PAC) (2.5mg/L Al^{3+}), and the flocculated solution was slowly transferred into the feed tank of the filtration unit. A lab-scale single membrane capillary (SMC) test unit was used for the filtration test runs. The bulk surface water was fractionated using $0.45\mu\text{m}$ (MF), 5kD (UF) and/or 3.5kD dialysis bags. Membrane performance was measured using permeability decline and its recovery after each backwash. Feed and permeate samples were analysed using UVA_{254} (m^{-1}) to estimate the organics removal through filtration. It could be shown that (1) particulates (organic/inorganic) helped minimise irreversible fouling, (2) the colloidal organics are the most irreversible fouling causing fraction, and (3) coagulation pre-treatment plays a significant role in improving and stabilising membrane performance.

KEYWORDS: NOM-fouling, fractionation, hybrid processes, microfiltration (MF), ultrafiltration (UF), coagulation

I. INTRODUCTION

Membrane operations in water treatment systems for the production of potable water are increasingly gaining attention. The technical limitation of this promising technology lies often in the so-called phenomenon of membrane fouling. In drinking water treatment, organic substances naturally present in surface waters, grouped together and known as Natural Organic Matter (NOM), are believed to be the major cause for irreversible fouling of UF/MF membranes. NOM in natural waters originates from aqueous, terrestrial, vegetative waste/products/by-products, and other algal cellular matter.

II. BACKGROUND

Membrane fouling depends largely on the following interactions between the substances (organic or inorganic) present in the feed water: (a) interactions between the different organics/inorganics present, and (b) the individual and combined interactions between the organics/inorganics and the membrane surface/pores. There have been a lot of studies reported on the different NOM fractions and their fouling tendencies [1], [2]. Fractionation procedures for NOM present in natural waters are being done for quite some time now [3], [4], [5], [6]. The idea to fractionate NOM in natural waters and to characterise the fouling tendencies of the different fractions is quite a recent step forward in the direction to minimise UF/MF fouling. Fan et al. (2001) [7] identified potential foulants in the order of hydrophilic neutrals > hydrophobic acids > transphilic acids. Howe et al. (2002) [8] indicated that a relatively small size range of inorganic

and organic colloids (3 ~ 20 nm in size range) represent an important foulant fraction in membrane filtration. Previous researchers have found that NOM adsorbs both inside pores and on the membrane surface (Combe et al., 1999 [9], Jones et al., 2000 [10]), or forms a gel layer (Yuan and Zydney, 1999) [11]. Lin et al. (2000) [12] and Carroll et al. (2000) [13] reported that the rate of fouling was reduced after coagulation pre-treatment. The primary objective of this work is to determine the NOM fraction(s) most responsible for irreversible fouling of submerged and backwashed UF/MF membranes, and the influence of coagulation pre-treatment on their fouling behaviour. Moreover, this work is part of a broader effort to setup a standardised procedure to characterise different feed waters with respect to their fouling characteristics.

III. MATERIALS AND METHODS

A. Surface water source

The surface water used as the source for fractionation was from a reservoir near Roetgen, a small town close to Aachen, in the extreme west of Germany. This reservoir supplies feed water to a water treatment system for the production of potable water. A medium scale pilot plant with submerged MF membranes is being run here. This water has a TOC content of about 3 – 5 mg/L, a high turbidity and is quite yellowish in colour.

B. NOM fractionation units

B.2a. $0.45\mu\text{m}$ flat sheet membrane cell (cross-flow unit)

This is a commonly used cross-flow cell, using flat sheet membranes, with a feed pump, valve controlled feed

inlet, permeate and retentate outlet, with the retentate circulated back into the feed tank. Permeate is collected on a physical balance for flux measurement. Two manometers are mounted at the inlet and outlet to the cross-flow cell. A flow-meter in the retentate line reads the volume flow rate (L/h). The 0.45 μ m cellulose nitrate membrane was from the company Sartorius AG. Filtration below critical flux is necessary to achieve a sharp fractionation (which is possible only when fouling is at minimum), i.e., to be certain that only the particulate fraction of the bulk NOM is getting retained by the 0.45 μ m membrane.

B.2b. 5 kD flat sheet membrane cell (cross-flow unit)

The fractionation with the 0.45 μ m membrane produces a filtrate containing the so called DOC, which contains colloidal and genuinely dissolved NOM. For further fractionation of these substances the same flat sheet membrane cell as above was used. The membrane sheet used was from the company Nadir NF, made of PES, with a nominal MWCO of 5 kD. Again, the cross-flow unit has to filter at minimum fouling conditions and thus below critical flux.

According to Leenher *et al.*, (2000) [14], with nominal 3.5 kD dialysis, the colloidal fractions are retained and the genuinely dissolved get through. The idea to use a cross-flow UF unit instead was primarily due to the drawbacks of the dialysis process, where it takes very long to fractionate the large volumes of water needed for the fouling experiments. Moreover, in dialysis, the DOC fraction ends up highly diluted and has to be re-concentrated before using the water for fouling experiments. This concentration step usually is performed by resin adsorption and desorption under rather harsh chemical conditions. It is arguable to what extent the characteristic properties of the dissolved NOM constituents are altered during this procedure.

B.2c. Dialysis bags of cut-off 3.5 kD

This unit was used as an optional/alternative fractionation step for the 5 kD flat sheet membrane setup. The dialysis bags used were Spectra/Pore 3.1, with a MWCO of 3.5 kD, supplied by the company Spectrapore. The dialysis bags with closures at both ends, along with a reasonably large container for the buffer, placed on a magnetic stirrer, together comprise the dialysis unit. A buffer volume of about 100 times the volume of the bag contents was recommended by Spectrapore, for effective dialysis.

B.3. Coagulation pre-treatment

Coagulation prior to membrane filtration is used to transform dispersed particles and other inorganic and/or

organic water contaminants into a preferable form for retention, aimed at minimizing particulate and NOM fouling. Coagulation was performed in a 2 l glass beaker (which was later used as the feed tank of the SMC test unit), and in a conventional two stage procedure. (1) The destabilisation stage comprises drastic mixing with an ultra-mixer (at speeds of 9000-12000rpm), where the coagulant is added and uniformly distributed throughout the volume. Here the equilibrium existing between finely dispersed particles and colloids gets disturbed through the neutralisation of their negative surface charge. This phase usually takes about 30sec. (2) The second phase is the flocculation phase, where the solution is slowly stirred (at about 50-100rpm) for about 30min. The dissolved colloids precipitate (sweep coagulation), and the particulates and colloidal particles form micro flocs, which gradually grow in size with time, forming macro flocs (flocculation). The coagulation conditions are described in Table 1.

Table 1. Coagulation conditions

Coagulant	Type	Conc. (mgAl ³⁺ /L)	pH	Temperature
Sachtoklar®	PACl	2.5	7.5	20°C

B.4. Membrane used for fouling experiments

The membranes used for the fouling experiments were from Puron AG, Aachen, Germany, and their properties are given in Table 2.

Table 2. Properties of the membrane used for fouling experiments

Producer	Puron AG
Type	Hollow fibre submerged membranes
Material	Standard PES
Nominal Pore Size	~ 0.1 μ m
Pure water Permeability (L/m ² .h.bar)	~ 900 – 1200

B.5. Filtration unit for fouling experiments (SMC test unit)

The unit (shown in Fig. 1) was designed and built by Puron AG (one of the project partners of this work) for filtration experiments using a single hollow fibre submerged membrane driven in “out-in” mode. Fully automated operation including backwashing as well as a CIP unit (Cleaning in Place) are the key features of this unit.

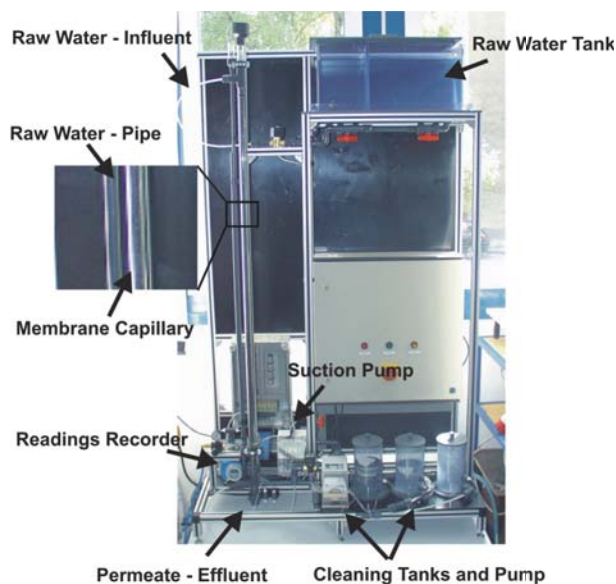


Fig. 1 Front view of the single membrane capillary (SMC) test unit

V. EXPERIMENTAL PROTOCOL

A.1. NOM fractionation

Roetgen reservoir water was used as the mother water containing the bulk NOM (particulate, colloidal, and dissolved organic carbon, or shortly P+C+D) that was fractionated step by step as described in Fig. 2. No fractionation was done beyond the dissolved organic carbon. As mentioned earlier, the 0.45 μm filtration was done in cross-flow mode to produce (C+D) filtrate. The UF membrane was operated in conventional cross-flow and diafiltration mode to produce the fractions (P+C) and (D), respectively. (C) was isolated using a dialysis unit (3.5 kD dialysis bags). This procedure rendered fractionation products containing the natural concentration of different NOM constituents. To summarise, the different fractions were produced as follows:

1. (P+C+D) – bulk NOM water itself
2. (C+D) – filtrate from the 0.45 μm filter operated in cross-flow with (P+C+D) as feed
3. (P+C) – retentate of diafiltration (with 5kD cross-flow UF unit), with (P+C+D) as feed
4. (D) – permeate of 5kD cross-flow ultrafiltration, with (P+C+D) as feed
5. (C) – bag contents after 24h of dialysis (3.5kD), starting with (C+D)

Fig. 2 Scheme of the fractionation steps

Fig. 3 Detailed flow diagram of the fractionation procedure

A more detailed description of the production of (P+C) is shown in Fig. 3. The diafiltration procedure was used to obtain (P+C) in its purest form, where cross-flow filtration below the critical flux was carried out with regular addition of pure water (Carbon-free) to the feed for every unit permeate volume produced. As only (D) can permeate through the 5kD membrane, more and more (D) were isolated from the (P+C+D) feed. At one point of time, the feed (now retentate) tank contained pure (P+C), and the permeate was a very dilute (D), due to all the pure water added to the feed during the course of filtration.