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On the removal of natural organic matter from superficial water by using UF and MF membranes

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On the removal of natural organic matter from superficial water by using UF and MF membranes

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ABSTRACT

Excessive chlorine usage in surface water disinfection may constitute a problem, since residual chlorine reacts with natural organic matter (NOM), already present in surface water, giving birth to disinfection by-products (DBPs). Aim of the work was to experimentally test the feasibility of a disinfection process by combining chlorination and ultrafiltration (UF), thus minimizing chlorine dosage. At the scope, in this paper, the removal of humic acid (aldrich humic acid, AHA), representative of NOM, has been studied by using tubular UF and microfiltration (MF) membranes (50 nm, 20 nm, 0.2 μm). Results show that, regarding AHA rejection and turbidity, investigated membranes are not influenced by transmembrane pressure and AHA concentration, while UF membranes seem to be more effective in salt separation. Anyway, both UF and MF show a removal efficiency up to 90%. Moreover, a process scheme aimed at water use/reuse is proposed, in which the chlorination step is substituted by a double filtration step (UF followed by reverse osmosis), in order to obtain high quality water, usable also for industrial purposes in which a high purity water is needed.

Keywords: NOM; Water; Reuse; Membrane; Ultrafiltration; Disinfection; Chlorine

1. Introduction

One of the major issues of mankind is the availability of safe drinking water. Since advances in analytical techniques continuously reveal new and emerging contaminants in water sources that are harmful to human health, strongly emerges the need of advanced treatments to remove or reduce these compounds. To this end, membrane processes are an effective and reliable treatment for removing targeted contaminants, regardless of the quality of the water source [1].

Membrane processes play an important role on the production of drinking water, free of health hazards, offering several advantages over conventional treatments such as compact module, lower energy consumption, environmental friendliness and high quality product. As a matter of fact, microfiltration (MF) and ultrafiltration (UF) processes have been increasingly used in potable water production as an alternative technology to conventional treatments eliminating

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coagulation, flocculation and sedimentation step, in order to remove particles, turbidity, microorganisms and natural organic matter (NOM) [2,3].

NOM is a complex organic matrix consisting of a wide range of organic substances with different molecular weights and chemical properties, whose major fraction is the one relative to the humic substances [3–5]. It first came to prominence in agriculture, because of its positive influence on the structure, water retention properties and nutrient status of soils, but it poses problems to the water supply industry. Removal of NOM is extremely important and it has become a challenging research in current development of water purification technologies, not only because it affects the odour, colour and taste of water, but mainly because it is considered as a precursor of disinfection by-products (DBPs). The most dangerous DBPs, recognized to be human carcinogens, are halogenated organic compound such as trihalomethanes (THMs) and haloacetic acids (HAAs), derived from reaction of organic material with free chlorine, used as a disinfectant in conventional treatment. DBPs control can be done by minimizing NOM (precursors) in the raw water, by reducing disinfectant doses, by removing DBPs after they form or by using alternative disinfectants. In addition, NOM compounds also cause fouling of the heat exchanger surfaces in industrial applications [6], as well as on membrane surfaces themselves. Although many studies on the DBPs formation have been carried out, the reduction of the DBPs concentration in the distribution system is still an important issue; in addition, the extent of removal of NOM is highly dependent on the characteristics of the NOM including organic carbon content, ultraviolet absorbance, biodegradable carbon content, polarity and molecular weight [7]. Kim and Yu [8] found that the formation potential of THMs is highly influenced by the hydrophobic fraction, whereas HAAs formation potential (HAAFP) depended more on the hydrophilic fraction. Their study showed that while the hydrophobic fraction was quite efficiently removed through conventional water treatment, the residual hydrophilic NOM after conventional treatment needs to be removed to reduce HAAFP. Katsoufidou et al. [9] studied humic acid rejection on polyethersulfone hollow fibre membrane (whose molecular weight cut-off —MWCO—was 150 kDa) obtaining rejections close to 20% in the absence of calcium and up to 75% with 2 mM Ca^{2+}. Mozia and Tomaszewska [10] studied a hybrid process (powdered activated carbon PAC/UF) with membranes prepared from polyacrylonitrile (MWCO 110 kDa); filtration test on natural waters showed that a PAC dosage of 100 mg/l enhanced organics removal from 44 to 64% and reduced membrane fouling by preventing organics from adsorption onto the membrane surface. Other researchers [11] found that the use of PAC either as a pretreatment agent or as an additive in the integrated PAC-UF system exhibits an increased membrane fouling. Aoustin et al. [12] studied the influence of calcium concentration on humic acid rejection through membrane of MWCO 10 and 100 kDa. They obtained rejections of 84–92% at $CaCl_2$ concentration in the range of 0.5–4 mM. Domany et al. [13] tested four polyethersulfon membrane of MWCO 5, 6, 15, 100 kDa obtaining a satisfactory removal efficiency of 85–90% for model solution and 62–69% for natural well-water. Chen et al. [14] published an interesting paper in which different treatment trains in pilot plants are presented, with the calculation of the total efficiency of different combinations of processes for NOM removal, both conventional and advanced such as granular activated carbon adsorption and ozonization with biological activated carbon, which performed fairly well in removing organic matter. This research group [15] has previously studied the removal of NOM from surface water by using flat sheet polymeric UF and MF membranes, which allowed to obtain satisfying results in UF configuration, while MF membrane resulted ineffective in aldrich humic acid (AHA) removal.

As for membrane fouling due to surface water constituents (particularly NOM), fouling has long been identified as one of the most significant points for membrane applications in water and wastewater treatment [1]. Advances in this field are individuated in the development of new membrane materials with different application fields, such as photo-catalytic membrane reactors, nanoparticle and carbon nanotubes composite membranes, and stimulus-responsive membranes [1].

From the reading of the above-mentioned papers, it appears that the solely disinfectant addition could oxidize organic matter, produce DBP and increase the assimilable organic carbon concentration; therefore, much attention must be paid to the removal of organic matter before disinfection, in order to limit DBP formation and preserve the biostability, as well as to the reduction of chlorine, which plays a major role in avoiding or at still reducing the DBPs formation.

In this study, the removal of humic substances from model solution through UF (50 and 20 nm) and MF membranes (2.0 μm) in tubular configuration has been investigated. Permeate and retentate samples were analysed by UV 254 nm absorbance to evaluate humic acid removal, while permeability and flux decay test were performed to estimate biofouling caused by organic material. Based on the obtained experimental results, a process scheme is proposed, in which a two-step membrane filtration (UF followed by reverse osmosis) is provided, in order to reduce or eliminate the need of chlorine and to produce water of high quality to be used for several aims.

2. Methodology

2.1. Experimental apparatus

Experimental studies have been carried out in a tangential flow laboratory pilot plant Membralox® XLAB 3 (EXEKIA, Bazet–France) with a single tube ceramic UF membrane Membralox® Tl-70 by Pall Corporation. Fig. 1 shows the pilot plant used for this study. The plant is equipped with a tank with a capacity of 3 l, pressurized to a maximum pressure of 3 bar with air or gas (high purity nitrogen). A recirculation pump with a nominal flow rate of 1 m^3/h ensures the water in the membrane a tangential speed of 7 m/s. Temperature is controlled by the tank jacket, which is connected to a thermostat CRIOTERM 10–80. Three tubular membranes were used with a MWCO of 50 nm, 20 nm and 0.2 μm.

2.2. Feed water characteristics

A model solution was created to supply constant quality of water sample by simulating real surface water: it contained $CaCl_2$ as a representative of naturally occurring multivalent cations, $NaHCO_3$ as a natural buffer system, kaolinite and α-alumina as natural turbidity and humic acid as NOM. A commercial humic acid (Aldrich) was employed in the present experiments. Humic acid stock solution was prepared by dissolving 100 mg AHA in 1 l distilled water, the pH of the solution was set to 10 by using NaOH to assure complete dissolving of humic acid, since the latter is insoluble at low pH. Solution was then filtered through 0.2 μm membrane filter. Background solution was prepared by dissolving 2.5 mg/l α-alumina and 2.5 mg/l kaolinite in deionized water (Milli-Q) in ultrasound bath for 20 min. Solution was then supplemented with $NaHCO_3$ and $CaCl_2$, and the desired humic acid concentration was adjusted using humic acid stock solution (100 mg/l AHA). After all the components were added, the solution was rapidly mixed for an hour with magnetic stirrer.

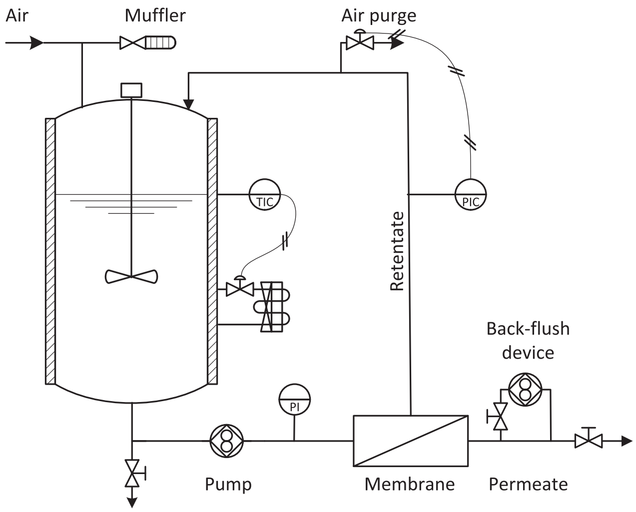

Fig. 1. Sketch of the experimental apparatus.

The average characteristics of synthetic water are presented in Table 1.

2.3. Testing procedure

The effect of membrane cut-off and AHA concentration on membrane performance was studied. Permeability, flux decay and concentration tests were executed for different AHA concentrations for each membrane. During the flux decay tests, the transmembrane pressure (TMP) was adjusted to 1 bar and temperature was controlled by the water jacket and kept constant to the value $T = 20^\circ C$. Permeate flux was manually measured for 3 h and the samples were collected from retentate and permeate and analysed for the parameters such as humic acid concentration, pH, turbidity.

AHA concentrations were determined by UV absorbance measurements in a spectrophotometer (Lambda 2S PerkinElmer) at the wavelength 254 nm; AHA concentration was linearly correlated with UV 254 nm ($R^2 = 0.9994$). After each test, the equipment and membrane were washed with an alkaline solution (NaOH) and rinsed with distilled water, until pH returned to the value of about 7.

Table 1
Synthetic water characteristics

Chemicals	Quantity	CAS number
NOM		
Aldrich humic acid	2.5–5.0 to 7.5–10 mg/l	1415-93-6
Inorganic particles		
Kaolinite	2.5 mg/l	1332-58-7
α-Alumina	2.5 mg/l	1344-28-1
Salts		
$NaHCO_3$	0.5 mM	144–55-8
$CaCl_2$	0.5 mM	10043-52-4

3. Results and discussion

In this study, UF and MF filtration experiments were carried out with three tubular membranes, to evaluate the effects of membrane cut-off (0.2 μm, 50 and 20 nm) and feed composition on membrane performance regarding AHA rejections and fouling.

3.1. Permeability and flux decay tests

Permeability tests were performed to evaluate membrane resistance to mass transfer at different AHA concentration; they were carried out by measuring the permeate flowrates with varying TMP. In each run, the operating parameters (temperature, cross flow velocity and feed concentrations) were constant (steady-state conditions). Figs. 2–4 show the influence of TMP on permeate flowrates at different AHA concentrations, for tubular of 0.2 μm, 50 and 20 nm MWCO, respectively.

As expected permeate flowrates decreased moving from MF to UF process: this behaviour was due to higher selective capabilities of membrane, as their pore dimension decreases from 0.2 μm to 20 nm.

Anyway, for each tested membrane, higher TMPs always assured better permeate flowrates, according to Hagen–Poiseuille model [16]. This behaviour seemed to suggest that no concentration polarization was formed on membrane surface, under different TMPs investigated in the present work, even if AHA concentration increased until 10 mg/l.

Furthermore, in case of fixed TMP, permeate flux decreased while increasing AHA concentration, due to higher flux viscosity.

Relevant decreases of flux with time were observed with all tested membranes. Fig. 5 shows permeate flux of 50 nm MWCO membrane as a function of time for all AHA concentrations studied: after three hours of filtration, a flux decay of about 40% was detected, thus confirming that AHA is a very fouling substance. This serious problem is enhanced by molecular structures in NOM, which adheres to the sides of membrane pores, affecting its permeability [5].

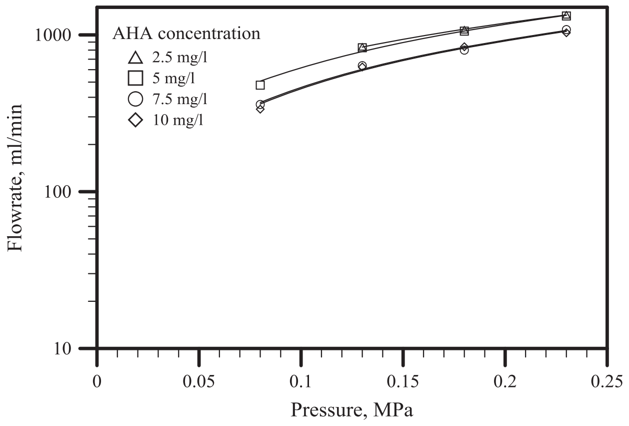

Fig. 2. Influence of TMP and AHA concentration on permeate flowrates for 0.2 μm membrane.

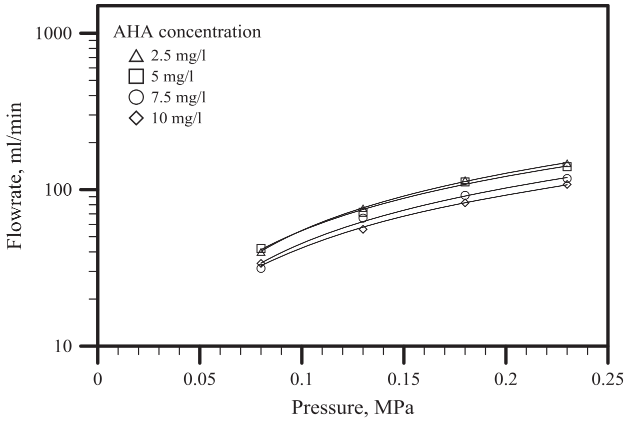

Fig. 3. Influence of TMP and HA concentration on permeate flowrates for 50 nm membrane.

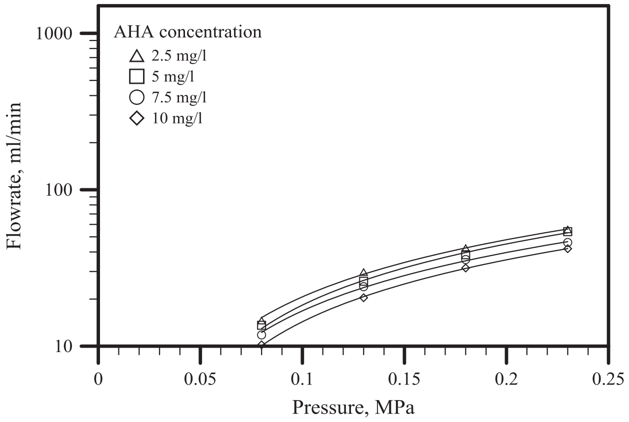

Fig. 4. Influence of TMP and HA concentration on permeate flowrates for 20 nm membrane.

3.2. Rejection tests

Figs. 6–8 show the results of rejection experiments towards AHA, representative of organic material present in surface water, for the membrane with MWCO = 0.2 μm, 50 and 20 nm, respectively.

Experimental results demonstrated that no remarkable differences can be detected in rejection values for each membrane, when varying TMP and AHA concentration values.

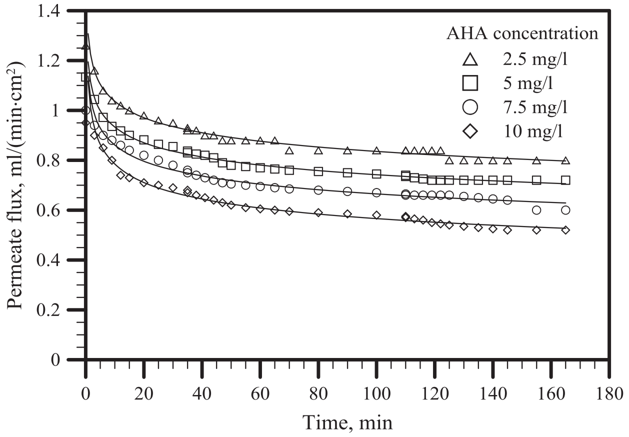

Fig. 5. Influence of TMP and AHA concentration on permeate flux for 50 nm membrane.

These results confirmed that detected membrane showed a satisfying selectivity feature in all experimental conditions, even with low TMPs. So, as for AHA retention, the examined UF membranes (50 and 20 nm, Figs. 7 and 8) were both suitable for the humic acid removal: rejection of AHA is higher than 90% up to 7.5 mg/l AHA. This was the case of MF membrane (0.2 µm, see Fig. 6), too, which showed almost similar and very high rejections, notwithstanding the higher pore size.

Fig. 9 shows the influence of TMP on conductivity rejection for MF (0.2 µm) and UF (20 nm) membrane, when AHA concentration is constant (10 mg/l). Experimental runs confirmed two important conclusions as regards membrane with higher rating (0.2 µm). Firstly, there was no great notable difference in conductivity rejection when varying TMP; secondly, its value was always low (not greater than 50%). This behaviour is mainly due to the membrane pore size: the rating of 0.2 µm is large enough, if compared to the dissolved salts present in surface water, so they pass through membrane to merge into permeate flux.

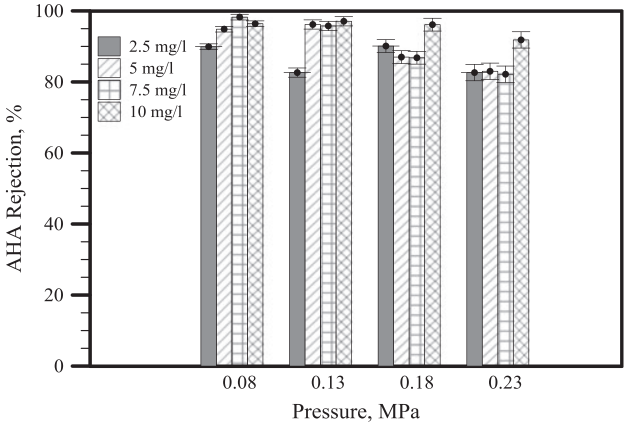

Fig. 6. Influence of TMP and AHA concentration on AHA rejection for 0.2 µm tubular membrane.

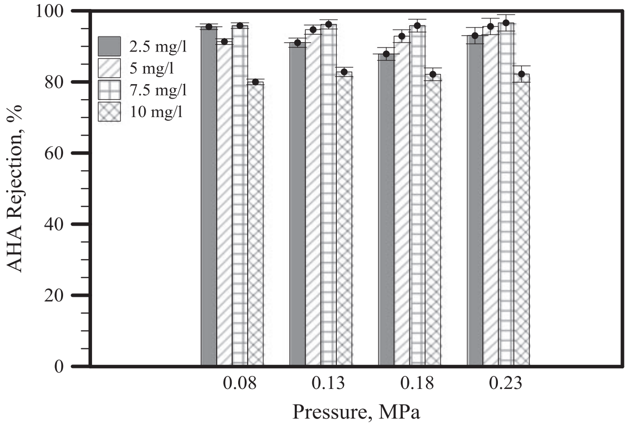

Fig. 7. Influence of TMP and AHA concentration on AHA rejection for 50 nm tubular membrane.

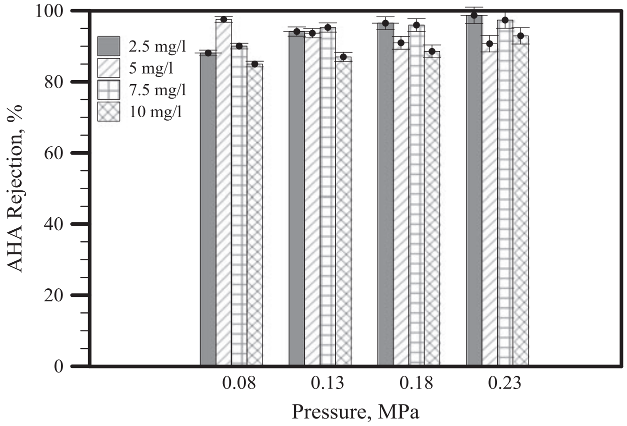

Fig. 8. Influence of TMP and AHA concentration on AHA rejection for 20 nm tubular membrane.

Slightly different performance was detected with lower membrane rating (20 nm). As showed in Fig. 9, experimental runs have allowed to detect an increasing of conductivity rejection as a function of TMP, reaching better performances (up to 80%) for higher pressure values. This is probably due to the higher membrane selectivity, which is able to retain, with increasing the pressure, higher amounts of salts in retentate flux. However, to enhance conductivity rejection, more advanced membrane process technologies are necessary, like nanofiltration or reverse osmosis.

Turbidity measurements confirmed high fouling power of such a feeding water, due to the presence of

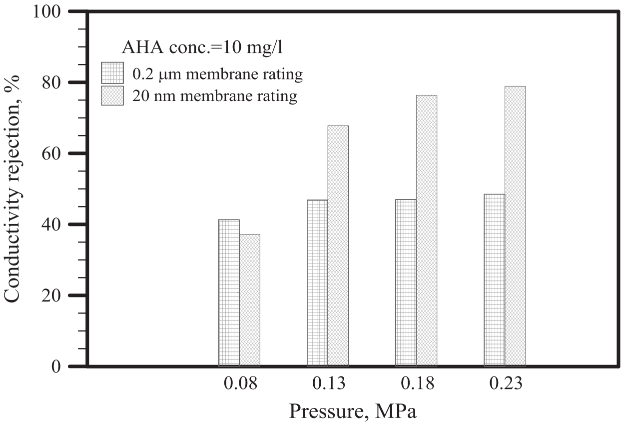

Fig. 9. Influence of TMP on conductivity rejection for 0.2 µm and 20 nm tubular membrane.

kaolite, α-alumina and humic acid. Experimental runs showed that no influence of TMP was detected on turbidity rejection in case of 0.2 µm rating membrane: particularly its value was always about 100% for all the AHA concentration and TMP examined in this paper. As expected, similar results were achieved for 50 and 20 nm membrane rating values.

3.3. Process analysis

On the basis of the obtained experimental results, a process analysis has been carried out, in order to propose a suitable process scheme of surface water treatment potabilization, in which the classical disinfection step is revised with the introduction of a membrane filtration stage, thus reducing the chlorine dosage. The analysis has been carried out using the SuperPro Designer simulator [17].

Fig. 10 shows a simplified classical disinfection block scheme, in which superficial water (flowrate = 2.4 m^3/s, $T = 25°C$, $p = 1$ atm), whose characteristics are reported in Table 2, passes through a grid for removing particulate matter (about 20% of TSS removal); then, it is pre-disinfected by the addition of chlorine in the typical concentration range. The organic matter (NOM) reacts with hypochlorous acid, to form by-products; afterwards water is clarified to remove the biomass with a typical efficiency (>80%). The produced sludge is usually discharged without the need of any additional treatment. In sequence, the clarified water is forced to pass through a sand filter and, like in the most advanced treatment scheme, through an activated carbon bed for a tertiary treatment; finally, water undergoes a further chlorination step. This scheme, widely diffused, implies a huge amount of chlorine addition, main responsible, by reacting with NOM, for the DBPs formation.

Fig. 11 proposes a new treatment sequence, in which after a preliminary clarification, water is sent into a double membrane filtration step, consisting of an UF module followed by reverse osmosis module, in order to obtain high quality water. Finally, a chlorination step is provided to ensure residue chlorine. Obviously, in this case, the chlorine addition will be lesser with respect to the previous scheme. The considered rejection coefficient towards NOM in UF step is about 80%, the same value detected in experimental plan. If water is intended for human consumption, the following step, reverse osmosis treatment, can be bypassed. On the contrary, if water is directed to be used for more advanced processes, for example to produce steam in a boiler, the reverse osmosis treatment is necessary to reach a very low salt content, compatible with that required purity.

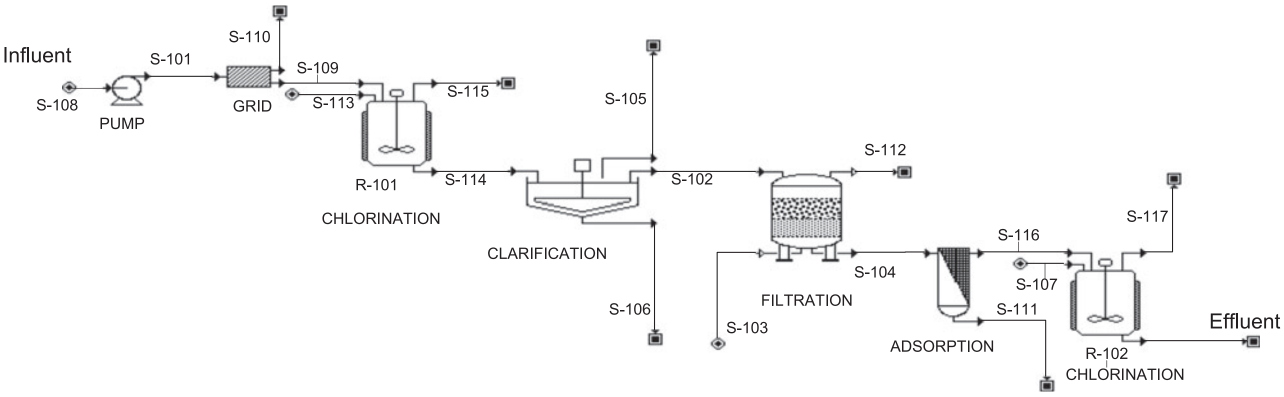

Fig. 10. Sketch of a conventional surface water potabilization process.

Table 2
Material balance relative to the process scheme in Figs. 10 and 11

Components	Units	Influent (Figs. 10 and 11)	Effluent (Fig. 10)	Effluent (Fig. 11)	Limits D. Lgs 2001, n. 31
Organic matter (AHA)	mg/l	5.00	0.50	–	–
Nitrates	mg/l	2.72	1.10	0.16	50
Biomass	mg/l	210.50	–	–	–
BOD_5	mg/l	4.00	0.10	3.10	–
Water hardness	°F	55.40	41.60	2.37	15–50
Chlorides	mg/l	928.70	27.70	2.19	250
Conductivity	μS/cm	3,540.00	800	130	up to 2,500
COD	mg/l	61.80	3.90	10.8	–
Phosphorus	mg/l	0.15	–	0.10	–
Sulphates	mg/l	183.40	5.50	1.71	250
DPB	mg/l	–	3.80	–	–
TSS	mg/l	26.00	2.20	0.10	–

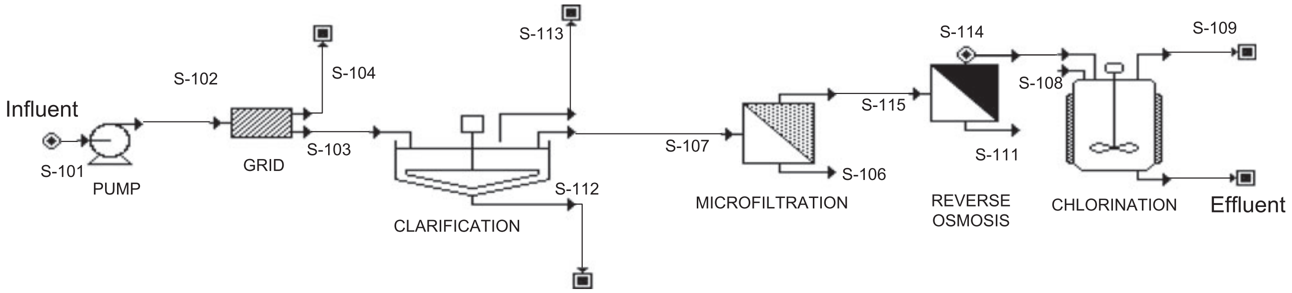

Fig. 11. Sketch of a membrane surface water potabilization process.

From the overall material balance, by considering the stoichiometry of disinfection reaction for the two proposed configurations, results shown in Table 2 have been obtained. The table reports the concentrations of the inlet stream (surface water) and the outlet streams of the conventional and new processes, compared with the emission limits imposed by Law in Italy as for drinking waters (D. Lgs. 2001, n. 31). The analysis of the table reveals how for both cases the water characteristics are very satisfactory; for the first scheme, some by-products are formed with a higher chlorine consumption (≅25 kg/h in the proposed scheme), while as for the second scheme, no by-products are formed, and about one-tenth of chlorine is needed (≅2.5 kg/h). However, energy expenses are higher, due to the energy required to overcome the water osmotic pressure in the RO stage. Nevertheless, if only UF is provided, TMP values are low (few bars) and thus energy costs between two plant configurations are comparable; if a reverse osmosis is needed, ultrapure water for advanced uses is obtained, thus justifying the higher costs.

4. Conclusions

Filtration tests with a model solution have demonstrated that the examined MF and UF membrane showed similar behaviours towards AHA rejection and turbidity: regarding these parameters, performances of the three membranes investigated seemed not to be influenced by TMP and AHA concentration values considered in this paper. Slightly differences can be detected as for conductivity rejection, where UF membranes (20 nm rating) seemed to be more effective in salts separation. However, performances achieved would suggest to use a more advanced filtration process (NF or RO) to reach higher conductivity rejection values.

All in all for the model solution investigated, MF membrane with low TMP can be surely recommended to perform an effective pretreatment step, before starting more advanced water treatment processes.

A process analysis carried out by using SuperPro Designer simulator, compares two schemes for surface water treatment: a classical process layout and a different process scheme, aimed at water reuse, in which chlorine is added in a small amount, only to ensure the effect of a chlorine residual concentration. A mass balance carried out on the membrane process scheme shows that in this last case, the problem of DBP formation, caused by the reaction of chlorine and NOM, is overcome; moreover, water of high quality is obtained, also usable for industrial purposes.

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