

## Potabilization of low NOM reservoir water by ultrafiltration spiral wound membranes

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### Abstract

Membrane technologies such as ultrafiltration offer an interesting alternative to integral treatment of surface water destined for human consumption. With this in mind, a pilot-scale ultrafiltration module was set up, equipped with spiral-wound polyethersulphone membranes (16.6 m<sup>2</sup>) with an effective pore size of 0.05 μm. The system operated continuously with a stable production of 0.9 m<sup>3</sup>/h (54 l/h) and a constant transmembrane pressure of −0.2 bar. The effluent obtained showed a total absence of faecal contamination indicators of both bacterial and viral origin, and also presented an excellent physico-chemical quality, independently of the quality of influent. Total aerobic bacteria counts revealed the problem of bacterial contamination in the membrane permeate zone, which could be controlled through daily chemical cleansing of the membrane. The chief problem presented by this type of system, applied as exclusive treatment, is low effectiveness in the retention of natural organic matter (NOM), in which respect the quality of the effluent was observed to depend on the quality of influent. This constitutes the principal limitation for applying the system to surface water due to the risk of disinfection by-products formation during the final post-chlorination. However, spiral wound ultrafiltration (SWUF) membranes could be used for low NOM reservoir water total treatment offering several advantages over conventional technologies.

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### 1. Introduction

With the detection of certain compounds in potentially harmful concentrations, it has become evident that absence of faecal contamination indicators and high organoleptic quality are not a sufficient guarantee for water destined for human consumption. In the light of recent findings related to organic and inorganic contaminants, particularly those regarded as disinfection by-products, there is a need to improve the performance of potabilization systems in this respect.

At present, the most widespread disinfection method throughout the world is the use of chlorine and its derivatives, owing to its effectiveness and particularly to its low cost. However, evidence of the formation of chloroform as a by-product of chlorination in water destined for human consumption [1,2]

has raised alarm with regard to the generation of chemical disinfection by-products, and a wide range of substances are now recognised to pose a serious threat to public health [3].

As a result of this problem, it is necessary to find ways of preventing the formation of disinfection by-products in the water without reducing effectiveness in the elimination of microorganisms. Various alternatives are currently being explored, with some researchers proposing the improvement of the quality of water with respect to the content of organic compound precursors [4], elimination of generated by-products [5], or the total or partial substitution of the practice of chlorination with alternative processes of a similar or different nature [6,7].

Membrane technologies, particularly ultrafiltration, represent an interesting alternative to the disinfection of water for human consumption [8]. These systems do not employ chemical oxidants, thereby reducing the generation of by-products. Instead they are based on a screening process which is highly effective at retaining both viruses and bacteria and avoids the

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problem of resistance, while improving the physico-chemical characteristics of the water [9].

Given that membrane technologies make it possible obtain a completely disinfected and clarified effluent, their application may lead to simplified potabilization facilities. However, since the treated water does not contain a residual concentration of disinfection, a final post-chlorination phase would need to be included in order to guarantee sanitary standards in the distribution network. Authors such as Galambos et al. [10] highlight the difficulty of retaining humic acids through membrane filtration, indicating that the disinfection process would also need to eliminate the possible precursors of chlorination by-product generation.

At the present time, low-pressure membrane technologies are used increasingly in the treatment of water for human consumption [8], applied in combination with other processes such as coagulation or ozonation [11,12]. The systems have also been applied at laboratory scale [6] and pilot-scale [13], with results indicating the possible application of ultrafiltration as an exclusive treatment for potabilization.

With these considerations in mind, it was decided to set up a pilot-scale ultrafiltration system working with surface water from a reservoir, in order to evaluate both the behaviour of the system and the final quality of the effluent obtained over 180 days' of continuous operation.

## 2. Materials and methods

### 2.1. Description of pilot-scale installation

The experimental pilot system (Fig. 1) was designed to produce a constant working flow of 0.9 m<sup>3</sup>/h, by means of a pump of surface water (1 m<sup>3</sup>/h, 30 m.c.a.) deriving from the reservoir of Canales (Granada, Spain). The plant comprised a ring-filter macrofiltration pretreatment phase (150 µm) and an ultrafiltration system using a SpiraSep module (TriSep Corporation).

The ultrafiltration module was equipped with spiral-bound polyethersulphone membranes (TriSep Corporation) with an effective pore size of 0.05 µm, installed in a 151-l capacity tank. The area of filtration was 16.6 m<sup>2</sup> (54 lmh), operating in a vacuum with a transmembrane pressure of -0.2 bar. Working conditions consisted of production periods of 60 min (0.9 m<sup>3</sup>/h) with continuous aeration, followed by backwashing phases of 2 min (2 m<sup>3</sup>/h) using filtered water. Chemical cleaning was carried out daily with chlorine (100 mg/l), and once a week using citric acid (pH 4.5).

### 2.2. Experimental methodology

The system operated continuously for a period of 180 days. Samples of both the influent and the effluent were taken on a daily basis. In all water samples, turbidity, total suspended solids, colour, permanganate oxidability and particles were analysed as physico-chemical parameters, while faecal coliforms, *E. coli*, enterococci, *Clostridium perfringens*, somatic coliphages and total aerobic bacteria were analysed as microbiological parameters.

For physico-chemical analysis, water samples were collected in thoroughly cleansed plastic bottles and analysed immediately. Analytic determination of turbidity was carried out using the quantitative diffuse radiation method described in Regulation UNE-EN ISO 7027: 2001. Suspended solids concentration was established by a filtration method using 0.45 µm filters, as reflected in *Standard Methods For the Examination of Water and Wastewater* [14]. For colour determination, the technique described in Regulation UNE-EN ISO 7887: 1995 was used, while determination of permanganate oxidability was based on the method described in Regulation UNE-EN ISO 846:1995.

Particle size distribution (PSD) of the permeate was conducted using a LiQuilaz-E20 particle counter (Particle Measuring Systems). The measuring principle is based on laser light extinction. A volume of 10 ml set at a fixed rate was analysed for each sample, which resulted in minimum counted particles of 100 ml<sup>-1</sup> and maximum of 100,000 ml<sup>-1</sup>. Particles were in a size range of 2 and 125 µm and the system was calibrated by inert latex particles of defined size.

For bacteriological and viral analyses, water samples were collected in sterile glass bottles (1 l) and analysed immediately after collection. The presence of faecal coliforms and *E. coli* was studied using the membrane filtration procedure (UNE-EN ISO 9308-1: 2001).

Enterococci were analysed using the method described in Regulation UNE-EN ISO 7899-2: 2001. Total aerobic bacteria count was carried out at 22 °C using the method described in Regulation UNE-EN ISO 6222: 1999. For determination of *C. perfringens*, samples were passed through membrane filtration (0.45 µm pores) and incubated anaerobically in PAB medium (Oxoid CM 587), supplemented with TSC (Oxoid SR0881E) and EYE (Oxoid SR047C) growing media, at 44 ± 1 °C for 24 h. After incubation, a count was made of colonies presenting the colour black. Somatic coliphages were examined using a modified form of the double agar layer method described by Adams [15], with *Escherichia coli* C (ATTC 13076) as host bacteria. Previous to analyses, 10 ml of sample was placed in a tube containing 2 ml of chloroform. The tube was shaken vigorously and then left to settle for 20 min. After heating at 45 °C, the chloroform was removed. This sample treatment removed all bacteria prior to coliphage determination.

Throughout the sampling period, chlorination curves were carried out for both the influent and the effluent, using aliquots of 1 l, and applying a quantity of NaClO (1 mg Cl) for the addition of chlorine to the water. After 30 min contact at slow agitation (30 rpm), free chlorine and total chlorine were quantified by means of the volumetric evaluation method using *N,N*-diethyl-1, and 4-phenylenediamine, as described in Regulation UNE-EN ISO 7393-1.

All data obtained in this study were analysed using the statistical program STAGRAPHS Plus 3.0 for Windows.

## 3. Results and discussion

One of the principal operational problems of membrane systems is fouling and clogging of the membranes. Although this may improve the capacity for retaining particles, it leads to a

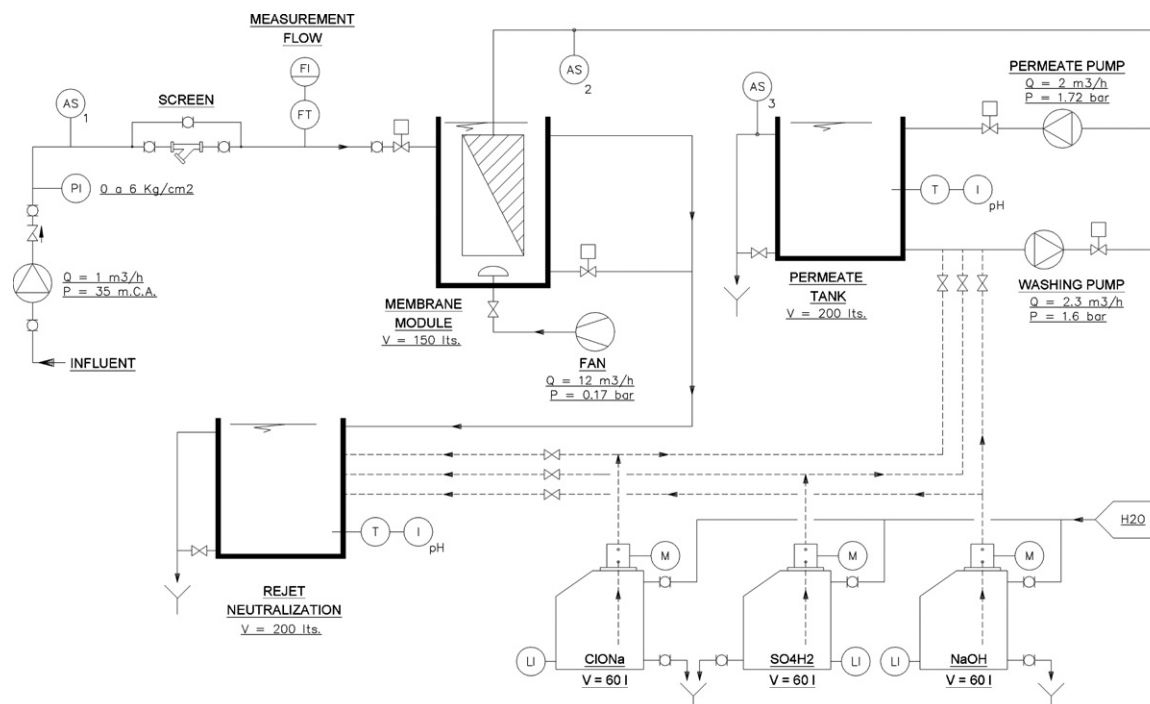


Fig. 1. Schematic diagram of experimental pilot plant.

rapid loss of permeability [16] with a consequent increase in transmembrane pressure [17]. The SWUF system did not present problems of permeability loss during the 180 days of operation, and the working flow of  $0.9 \text{ m}^3/\text{h}$  with a transmembrane pressure of  $-0.2 \text{ bar}$  was constantly maintained. The absence of any problems deriving from fouling and clogging indicates that time periods established for backwashing and chemical cleaning phases were adequate for the correct operation of the system.

In these conditions, the experimental system achieved a recovery of 93%, generating a retentate of 70 l for each  $\text{m}^3$  of effluent. On occasion, the retentates presented a high concentration of chlorine or a slightly acidic pH, both of which were neutralized by mixing with other retentates produced after backwashing. Basing the prediction of retentate concentration on a simple mass balance, which has proved valid for parameters such as suspended solids and turbidity [16], the retentate generated by the pilot system presents a considerable microbial concentration, at approximately  $10^8 \text{ cfu/ml}$ , and a suspended solids concentration of approximately  $200 \text{ mg/l}$ . However, the organic content is low ( $6.5 \text{ mg O}_2/\text{l}$  as permanganate oxidability) in consequence of the quality of influent. A retentate of these characteristics could be adequately managed through discharge into the urban sanitary network.

Analysis of particle distribution in the influent indicated an absence of particles with a size above  $33 \mu\text{m}$ . As particle size decreases, particle concentration increases according to a logarithmic progression (Fig. 2). With respect to the effluent, particle distribution analysis also revealed a logarithmic progression, although in this case the size did not exceed  $11 \mu\text{m}$ . These results indicate the effectiveness of the ultrafiltration membrane at eliminating particle material from the influent. However, it

is noteworthy that the effluent presented particles well above the size of the membrane pore, whose origin would seem to be highly diverse.

The medium number of particles between 2 and  $125 \mu\text{m}$  in the effluent was 50 units/ml. However, it was observed that a large quantity of particles enters the effluent immediately after chemical cleaning with hypochlorite (Fig. 3) or citric acid. Similarly, after the hourly backwashing phases (70 and 140 min after chemical cleansing in Fig. 3) a slight increase in the number of particles between 2 and  $125 \mu\text{m}$  was noted. Backwashing water was obtained from the bottom of the effluent tank, where particles of the effluent were concentrated. So, during backwashing particles are retained in the clean side of membrane which appeared in the effluent after backwashing. On the other hand, a substantial proportion of the particles exceeding the membrane pore size were swept into the effluent after chemical cleaning. The origin of the particles swept into the effluent would appear to be incrustations produced in the permeate zone, the development of biofilms, accumulation of organic matter, wear and

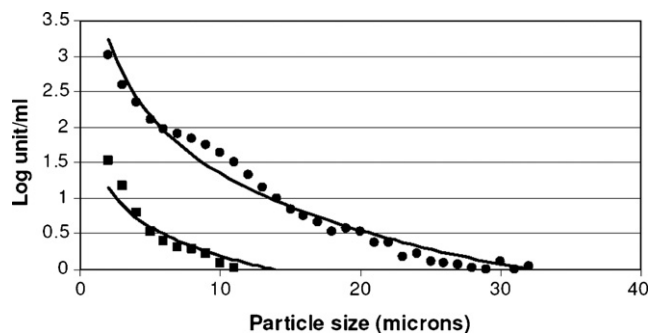


Fig. 2. Influent (●) and effluent (■) particle size distributions from 2 to  $125 \mu\text{m}$ .

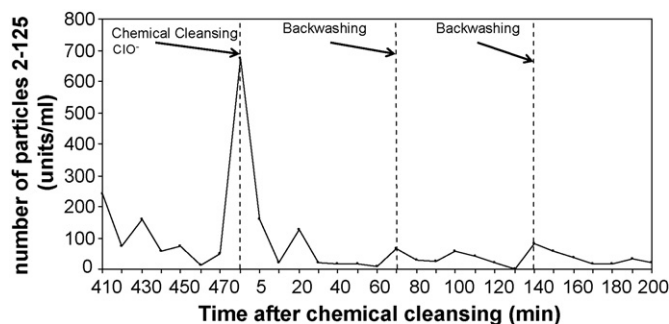


Fig. 3. Effluent 2–125  $\mu\text{m}$  particle count (unit/ml) after hypochlorine chemical cleansing and backwashing.

tear of materials, etc., which were removed by the action of chemical reagent. This problems affect to treated water quality momentarily and must be taken into account when ultrafiltration membranes are used for water purification.

Regardless of particles analysis quality of treated water by SWUF membranes was suitable with EU potable standard (Table 1), mainly with regard to microbiological parameter. One of the great advantages of SWUF membranes is their capacity to disinfect water without problems such as the resistance of micro-organisms to the disinfection process or dependence on the quality of influent [9], due to the higher size of bacteria (0.3–10  $\mu\text{m}$ ) than membrane pore (0.05  $\mu\text{m}$ ). This capacity was confirmed in the present experiment, with an elimination rate of 100% for the faecal indicators faecal coliphorms, *E. coli*, enterococci, and *C. perfringens*. The rate remained stable throughout the experimental period regardless of the quality of influent, which presented considerable fluctuations (Table 1).

As with bacterial indicators of faecal contamination, the experimental system demonstrated a high level of effectiveness with regard to the retention of somatic coliphages (Table 1). This result may be attributed to the average pore size of the membrane, similar to coliphage size (0.05–0.1  $\mu\text{m}$ ) and to the low concentration of such indicators in the influent. 100% of the samples analysed presented total absence of this type of viral indicator, thereby endorsing the microbiological quality of effluent obtained by SWUF membranes with regards both to bacterial and viral content.

Table 1  
Summary of influent and effluent characteristics from the ultrafiltration system

Parameters	Raw water			Treated water			Yield (%)	EU potable standard
	Maximum	Minimum	Average	Maximum	Minimum	Average		
Turbidity (NTU)	12.8	3.9	6.4	0.95	0.07	0.24	96.25	1.0
TSS (mg/l)	81.6	2	12.03	0	0	0	100	–
Colour <sub>436nm</sub> ( $\text{m}^{-1}$ )	2.2	0.2	0.48	0.4	0	0.0001	99.97	–
MnO <sub>4</sub> <sup>-</sup> Ox. (mg O <sub>2</sub> /l)	1.55	0.4	0.96	1	0.17	0.55	42.5	5.0
A <sub>254</sub>	0.039	0.009	0.019	0.021	0.006	0.011	38.1	–
Faecal coliforms (cfu/100 ml)	185000	9	2098	0	0	0	100	0.0
<i>E. coli</i> (cfu/100 ml)	170000	8	1006	0	0	0	100	0.0
Enterococci (cfu/100 ml)	6500	1	127	0	0	0	100	0.0
<i>C. perfringens</i> (cfu/100 ml)	49	7	27	0	0	0	100	0.0
Somatic coliphages (pfc/100 ml)	180	0	16	0	0	0	100	–
Total aerobic count (cfu/ml)	3750000	120	71301	5	0	1.6	99.9	100.0

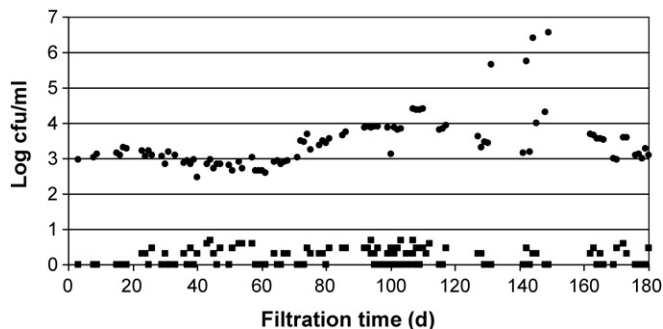


Fig. 4. Variation of influent (●) and effluent (■) total aerobic count over time.

A microbiological parameter of particular importance when working with membranes is the total aerobic bacteria count. This parameter presented positive counts in 48% of the samples analysed, with maximum values reaching 5 cfu/ml (Fig. 4), without correlation with the quality of influent ( $r^2 = 0.001$ ). Although these values do not indicate poor water quality, they do reflect the level of cleanliness of the membrane, which is a highly important aspect of potabilization by means of ultrafiltration. Authors such as Jacangelo et al. [18] or Gómez et al. [9] have observed contamination of the membrane in the permeate zone which could even lead to the presence of faecal contamination indicators in the effluent, although these are not related to the quality of the influent or to problems with the membrane. Since our experimental system did not operate in sterilized conditions, conditions leading to the development of biofilms could arise, as occurs with water distribution systems [19], resulting in a loss of water quality. As suggested earlier, presence of such bacteria in the effluent would explain the presence of particles exceeding the membrane pore size. Given that the control of this type of contamination relies on membrane cleaning processes, the greater or lesser presence of aerobic bacteria will depend on the efficiency of these processes, particularly of those using chlorine.

Application of SWUF membranes brings about a considerable improvement in specific properties of the water such as turbidity or suspended solids concentration. In the present experiment, a more or less stable quality was obtained for these parameters, indicating a high level of performance (Table 1).



This quality was maintained throughout the experimental period, independently of operational time or quality of influent, which fluctuated considerably. This behaviour is characteristic of screening systems, as has been noted by other researchers [16]. Application of SWUF membranes to reservoir surface water may therefore be considered sufficient to guarantee a total absence of suspended solids and a constant quality with regard to water turbidity.

The method for colour determination described in Regulation UNE-EN ISO 7887 involves determining the spectral absorption coefficient at three different wavelengths: 436, 525 and 620 nm. However, the influent in the present experiment showed null values for 525 and 620 nm, presenting absorption at 436 nm only. This behaviour is characteristic of natural water of brown or yellowish colour, whose intensity depends on organic content. The SWUF membrane produced a significant improvement in the colour (Table 1) independent of the quality of influent ( $r^2 = 0.014$ ). Spectral absorption coefficient values ( $\alpha_{436}$ ) in the effluent were below  $0.4 \text{ m}^{-1}$ , indicating a level of less than 2 mg Pt/L. This considerable improvement may be attributed to the origin of the elements producing colour in the influent, which may be screened by a membrane with an average pore size of  $0.05 \text{ }\mu\text{m}$  without the need for preliminary treatment such as coagulation, although this has been frequently combined with membrane ultrafiltration for the elimination of colour [20].

The presence of organic material in the treated effluent may be regarded as very low, with average values between  $0.55 \text{ mg O}_2/\text{l}$  for permanganate oxidability or absorbance of 0.011 at 254 nm. However, this low concentration is attributable to the low concentration in the influent, and retention of NOM actually presents the poorest performance level of all the parameters assayed (Table 1). This situation has likewise been observed by other authors [17] for similar parameters such as chemical oxygen demand (COD), even when working with ultrafiltration membranes with a low molecular cut-off [6,16,21]. As may be seen in Fig. 5, a strong correlation exists between the quality of the effluent and that of the influent ( $r^2 = 0.8015$ ), so that high concentrations of NOM in the influent are likely to result in high concentrations in the effluent.

In experiments with ultrafiltration and nanofiltration membranes, Galambos et al. [10] and De la Rubia et al. [21] found that the ultrafiltration membrane presented low effectiveness at eliminating dissolved organic materials such as natural humic acids. This system remove NOM by a sieving mechanism, so

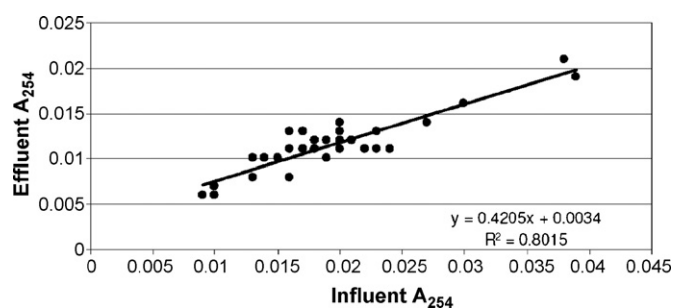


Fig. 5. Influent and effluent  $A_{254}$  correlation.

ultrafiltration membranes are only effective for large molecular size NOM. Similarly, Mijatovic et al. [22] studied the effectiveness of ultrafiltration membranes in the elimination of humic acids in natural water. Using the permanganate oxidability index as a measure of the concentration, results indicated average elimination levels of approximately 25%. With these studies in mind, it is clear that the present experimental system produced an effluent of high quality because the influent was also of high quality. Consequently, application of SWUF membranes as exclusive potabilization treatment cannot be considered suitable for surface water with a high concentration of NOM. Under this conditions, the potabilization treatment must be improved by pre-treatment or post-treatment to membrane, increasing the capacity of the system over NOM. The combination of ultrafiltration membranes with previous coagulation–flocculation [11] or ozonization could be an alternative, like the application of nanofiltration membranes which are suitable to eliminate NOM [21], although others factor such us ionic strength of water, membranes permeability and pretreatment prior to membrane must be taken into account [23].

An advantage of using SWUF membranes in potabilization is that they permit a reduction in the chemical disinfectants applied to the water, given that the disinfection is physical. However, the presence of organic material in the effluent after ultrafiltration treatment may lead to the development of micro-organisms [24], principally through the formation of biofilms [19]. These micro-organisms would affect not only the membrane permeate zone, but also storage systems and the water distribution network, with the consequent risk for public health [19].

One of the mechanisms to ensure that the water remains disinfected throughout the distribution system is the addition of a dose of chlorine to the effluent from the potabilization system. The present experiments show that the demand for chlorine in water with a low NOM content is reduced after ultrafiltration treatment, but does not disappear completely, as may be seen in Fig. 6. Effluent from SWUF membranes continues to present chlorine demand, and the demand increases in line with the increase of NOM in the water to be treated. Organic compounds of this type are regarded as precursors of the generation of chlorination by-products [25], which is also influenced by the dose of chlorine to be applied [26]. Consequently, ultrafiltration membranes alone cannot be considered adequate treatment for water with a

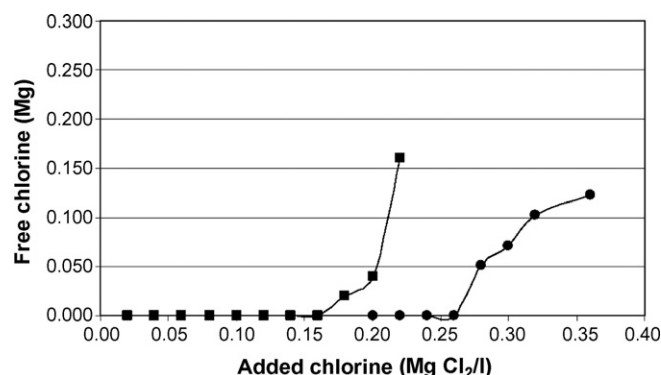


Fig. 6. Influent (●) and effluent (■) free chlorine after chlorination.

high NOM content, owing to the potential risk of disinfection by-products being generated in the chlorination post-treatment, which would be necessary to guarantee the microbiological quality of the water reaching consumer. However, potabilization of low NOM reservoir water can be simplified by SWUF membranes since it can be applied as only treatment together with the obligatory post-chlorination. This system reduces the surface needed for water treatment and the labour at the same time as guarantee a constant water quality independently of the quality of influent mainly the microbiological quality without problems of resistance by target micro-organisms.

#### 4. Conclusions

The results obtained in this study demonstrate the excellent performance of SWUF membranes at eliminating bacteria and viruses, producing perfectly disinfected water. A similarly high level of performance is shown for physico-chemical parameters such as turbidity, colour and suspended solids, in respect of which the quality of the effluent is independent of the quality of influent. The technology is therefore particularly appropriate for surface water with a high variability in quality. However, attention must be drawn to certain problems such as the microbial contamination of the permeate zone, which may lead to an increase in the presence of total aerobic bacteria, although this may be controlled through periodical chemical cleansing. A second and more important problem presented by this technology is its low performance in the retention of NOM, which did not exceed 42% in the experimental system, while the quality of the effluent with regard to these compounds depends on the quality of the influent. Consequently, in water with a high NOM content, the SWUF effluent would present a demand for chlorine requiring the addition of disinfectant concentrations in excess of the quantity required to maintain residual concentration.

We may conclude, however, that SWUF membrane is suitable as exclusive treatment for surface water with low NOM content, obtaining a perfectly disinfected effluent of high physico-chemical quality.

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