



CAN MICROFILTRATION OF TREATED WASTEWATER PRODUCE SUITABLE WATER FOR IRRIGATION?

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ABSTRACT

Water requirement for irrigation dramatically exceeds the traditional resources of Tenerife island that are becoming more and more brackish. An important programme of wastewater recovery is actually implemented; it is focused on the reuse of the secondary treated wastewater of the city of Santa Cruz for the irrigation of banana and tomato crops. Considering the hard competition with South American producers, the programme demands water completely disinfected. Microfiltration meeting the required standards, this study was then devoted to preliminary results obtained by cross-flow filtering through a 0.14 mm inorganic composite membrane, i.e. Carbosep M14, which was indeed a total barrier for suspended solids, total coliform, fecal coliform and fecal streptococci. The removal of turbidity and total COD were also significant, 93% and 60%. There was no rejection of the soluble fraction of size lower than 0.01 mm. Some 45% abatement of phosphorus was also obtained. The microfiltered water was therefore perfectly adapted for irrigation. In spite of a fouling mechanism difficult to identify, a critical flux of 100 l/m² h was obtained at 1 bar driving pressure and 3 m/s cross-flow velocity and this value was close to the permeation rate for tap water. A phenomenological approach of the operation allowed us to define two dimensionless groups: the shear stress number and the fouling number. These numbers allowed us to display all the experimental results in only one curve. © 1998. Published by Elsevier Science Ltd. All rights reserved

KEYWORDS

Adimensional analysis; disinfection; inorganic membrane; irrigation; microfiltration; tertiary treatment.

INTRODUCTION

The water requirement of Tenerife island dramatically exceeds its water resources. The traditional underground resources recovered by water galleries or wells which represented up to now 99.5% of the total distributed water are actually unable to satisfy the demand for human, touristic and agricultural purposes. Moreover, as most of this resource is becoming brackish its use will be more and more hampered. Sea water desalination and wastewater reuse already produce some 1400 m³/year and, in 2002, the overall production will be increased up to 4000 m³/year. Taking into account the economic limitations of desalination, an important programme of wastewater reclamation has consequently been implemented for agricultural purposes. Started in 1993, the project is focused on the reuse of the effluent of the wastewater treatment plant of Santa Cruz to irrigate the southern banana and tomato crops. Since there is a strong economical

competition with the South American growers, the water reuse requires health guarantees acceptable by the international market.

Membrane processes have largely demonstrated their ability to replace a secondary clarifier. A more rational approach would be coupling together a fixed biomass reactor with membrane equipment (Bailey *et al.*, 1994). But both approaches mean a completely new design. More realistic in the next few years would be the use of membranes in the tertiary step of an existing wastewater treatment plant (Ben Aïm *et al.*, 1993). Membranes offer indeed the possibility of simultaneous clarification and disinfection without the risk of organo-halogenated compound formation. In Sydney, Australia, pilot studies have been conducted on a large scale (Peters and Pedersen, 1990). The effluent from the secondary sedimentation tank was fed to a microfiltration unit. A comprehensive microbiological testing programme established that all indicators of pathogenicity (bacteria and viruses) were removed by this system operated with gas backwashing. Testing for chemical quality showed significant reduction in BOD, turbidity and oil/grease as well as some reduction of heavy metals and phosphorus, suspended solids being completely abated.

However, the major hurdle in the extensive use of membranes is the continuous reduction of permeation flux caused by membrane fouling. Since the permeation flux is known to be the main factor determining the economic feasibility in practical aspects, specific applications of membrane processes may require careful attention to various causes of membrane fouling. In particular, membrane fouling is in close association with the physico-chemical properties of solutes and the membrane. For instance, the increase of solid concentration, the size reduction of solids or the size distribution of particles being filtered proved to reduce membrane permeability. A need for a comprehensive study of membrane fouling by secondary clarified suspensions still remains because there are many unknowns regarding the fouling mechanisms in such complex systems, particularly in the case of Santa Cruz where the wastewater conductivity is particularly large at about 1600 $\mu\text{s}/\text{cm}$ and where nitrification is not carried out. Moreover, in this first step towards the design of an efficient and economical treatment, an inorganic membrane is better adapted than an organic one. This work is devoted to the preliminary results which were obtained.

MATERIAL AND METHODS

The experimental unit provided by Tech-Sep consisted of a 40 cm long and 6 mm diameter tubular Carbosep membrane in a closed loop where the permeate and the retentate were both recirculated (Fig. 1). A thermoregulation at 30°C, a temperature currently observed, was maintained in a 25 litre tank. Such a large volume allowed multiple sampling without hampering the filtration runs. The selected M14 membrane was an inorganic composite membrane whose zirconia-active layer was deposited on a carbon support. Its rated pore membrane as given by the manufacturer was 0.14 μm . The membrane was cleaned prior to each experiment using a 3% solution of sodium hydroxide, followed by washing with a 3% solution of nitric acid, followed by through rinsing with tap water. Tap water (900 $\mu\text{s}/\text{cm}$ conductivity) permeability experiments were carried out to assess the cleanliness of the membrane.

The suspension was the effluent of the Santa Cruz wastewater treatment plant which includes a pretreatment and a primary sedimentation followed by an activated sludge whose volumetric loading rate is about 1.6 kg BOD₅/m³ day. The main characteristic parameters of this effluent are given in Table 1. The high conductivity values were essentially due to sodium and hydrogenocarbonate.

All the physical, chemical or biological analyses were carried out in accordance with the Standard Methods (1995). The pollutants size distribution was measured by filtering through different cellulose membranes whose mean pore diameters were respectively 1.2 and 0.22 and 0.2 and 0.05 and 0.01 μm . The particle size distribution was quantified with a Coulter LS laser granulometer which gave a distribution by particle numbers. pH was obtained with a pH-meter Metrohm and turbidity with a turbidimeter HACH DR 3000.

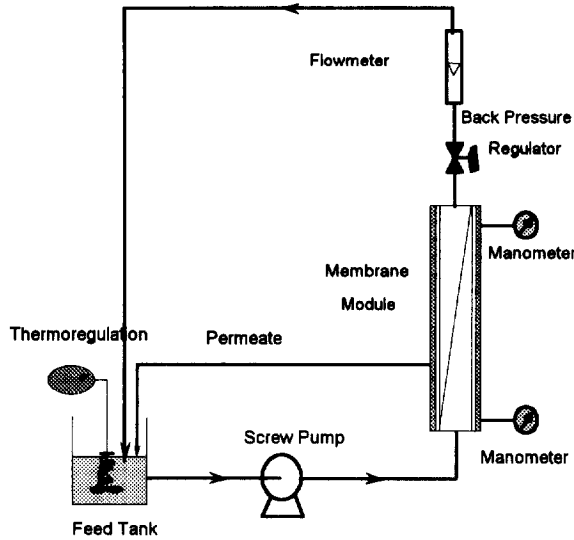


Figure 1. Experimental set up.

Table 1. Main characteristics of the suspension and the filtrate

Parameters	Feed			Filtrate		
	Maximum	Minimum	Average	Maximum	Minimum	Average
pH	8.24	7.92		8.61	8.16	
Conductivity, $\mu\text{S}/\text{cm}$	1810	1440	1620	1780	1440	1610
Turbidity, FTU	53	38	43	5	2	3
S.S. content, mg/l	40	20	28	Nil	Nil	Nil
Total COD, mg/l	110	80	89	39	27	34
N-NH ₃ , mg/l	35	26	30	32	24	28
PO ₄ ³⁻ , mg/l	59	37	41	35	28	32
Fecal coliform col/100 ml	66 000	8 000	25 800	Nil	Nil	Nil
Total coliform col/100 ml	152 000	9 100	62 280	Nil	Nil	Nil
Fecal streptococci col/100 ml	88 000	14 000	52 750	Nil	Nil	Nil

RESULTS AND DISCUSSION

Membrane rejection

Irrespective of the operating parameters, the M14 membrane was a total barrier for the suspended solids (Table 1). This is a classical result already observed (Ben Aïm *et al.*, 1993) and explained by the particle size distribution (Fig. 2). In consequence, the turbidity abatement was larger than 93%. The total COD removal was about 60%; the analysis of the size distribution of the effluent and the filtrate showed that the abated fraction corresponded essentially to species larger than 1.2 μm and that species smaller than 0.05 μm were not rejected (Fig. 3). The suspended solids deposition did not then constitute a dynamic membrane able to separate species smaller than the rated pore membrane as observed in water potabilization by microfiltration

through a 0.2 μm membrane (Elmaleh and Naceur, 1992). The membrane was also a total barrier for the total coliform, the fecal coliform and the fecal streptococci (Table 1). This disinfection effect is particularly interesting since the filtered water is intended for irrigation. It allows us to discard the tedious chlorination step. Virus can be also removed by microfiltration owing to the deposited layer but not immediately after a backflush (Peters and Pedersen, 1990). Some 45% reduction of phosphorus was also observed and was probably due to the complexation of phosphorus compounds. The slight nitrification was probably due to oxidation in the filtration loop. On the other hand, the conductivity removal was negligible since microfiltering could not eliminate small ions.

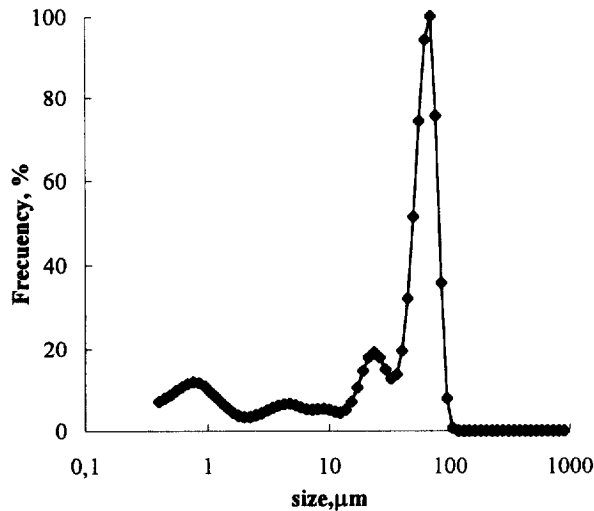


Figure 2. Particle size distribution.

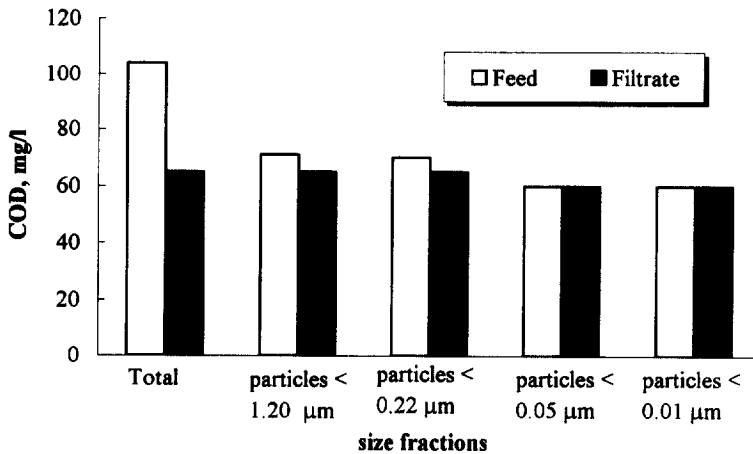


Figure 3. Size distribution by filtration in series.

Influence of operating parameters

The flux variations during a filtration run were quite classical: decreasing with time and tending toward a steady value (Fig. 4). However the flux steadiness was reached faster at low driving pressure and high cross-flow velocity. The important concept of critical flux corresponds to a permeation rate which does not decrease with elapsed time and keeps consequently a value very close to the one observed at the beginning

of the run. This concept is particularly relevant while operating a membrane unit since no additional energy or counter-washing or any other remedial action is required to recover the initial flux. Such a critical flux of slightly more than $100 \text{ l/m}^2 \text{ h}$ was observed at 1 bar and 3 m/s cross-flow velocity (Fig. 4). This value should be compared with previous studies. While filtering clarified secondary wastewater with a $0.2 \mu\text{m}$ Kerasep membrane, Pouet (1994) showed that the flux depended on the loading rate of the activated sludge plant; for a low loading rate, the flux reached $200 \text{ l/m}^2 \text{ h}$ at 1.7 bar and 3 m/s while the effluent of a high loaded activated sludge filtered under the same conditions led to $150 \text{ l/m}^2 \text{ h}$. In both cases, these permeation rates could not be maintained and a high frequency counter washing was required. In these cases too, the suspended solid concentrations were significant higher than in this study, i.e. 60 to 65 mg/l versus 30 mg/l (Table 1). It should be assumed that, in this study, the suspension chemical characteristics and its interaction with the membrane were particularly adapted to the production of a high critical flux as shown by Bowen *et al.* (1996) who demonstrated that the osmotic modelling accounted for multiparticle electrostatic interaction, dispersion forces and configurational entropy effects. Their model yielded a prediction for the filtration rate of colloids as a function of particle size, zeta potential or surface charge and ionic strength. Unfortunately, a secondary effluent is too complex and insufficiently characterized to allow the use of such a modelling with no adjustable parameters but this approach could give some hints.

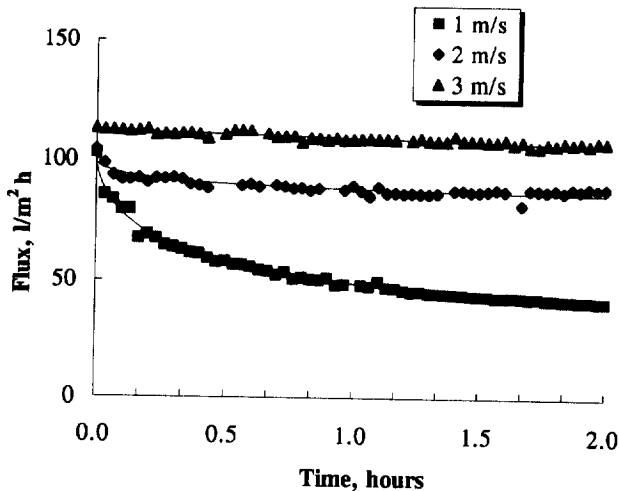


Figure 4. Flux against time at 1 bar driving pressure.

Under the same cross-flow velocity, the flux reached a maximum limiting value for all membranes at a driving pressure between 1 and 3 bar (Fig. 5). At low cross-velocity, the permeation rate was nearly independent of the driving pressure. Nevertheless, a slight tendency to decrease was observed while the pressure was increased. For most suspensions, the permeation rate increased with the pressure approaching a limiting flux. The relative decrease in Fig. 5 at higher pressure was also observed while filtering anaerobic cells (Elmaleh and Abdelmoumni, 1997). It was attributed to a compression and a deformation of the deposited particles beyond a critical pressure. It was also predicted by Bowen *et al.* (1996) for some values of the membrane zeta potential.

At cross-flow velocities less than 2 m/s, the flux was independent of the driving pressure (Fig. 6). A plateau appeared between 2 and 3 m/s. An interesting result was that there was nearly no fouling resistance at a cross-flow velocity of 3 m/s with a 1 bar driving pressure as shown in Fig. 6, where the filtration flux was the same as the tap water flux of the membrane. Such a result could be explained by a predominant reversible particle deposition on the membrane surface.

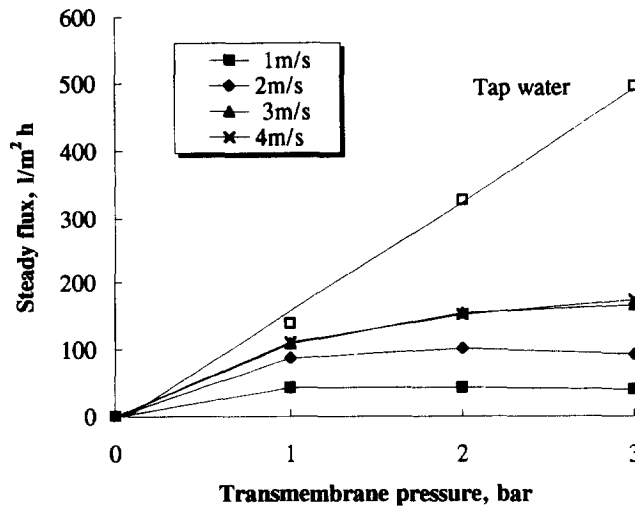


Figure 5. Steady flux against driving pressure at different cross-flow velocities.

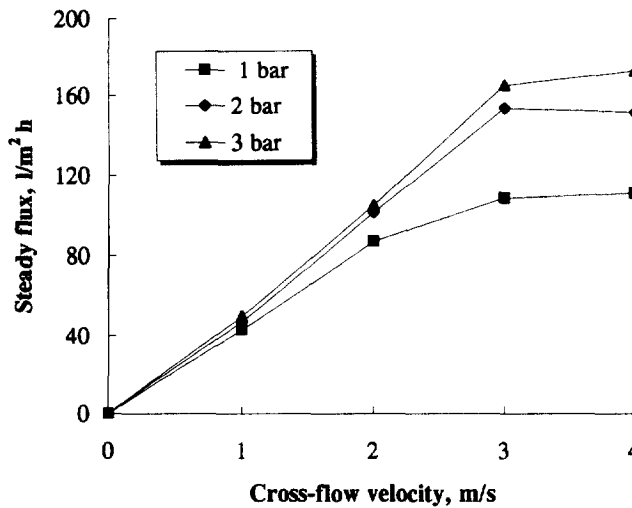


Figure 6. Steady flux against cross-flow velocity at different driving pressures.

A helpful model to determine the fouling mechanism of particles filtration was recently proposed by Song and Elimelech (1995). This theory shows that the extent of concentration polarization, as well as the behaviour of the permeate flux, are characterized by a dimensionless number called the filtration number:

$$N_F = \frac{4\pi r^3}{3kT} P \tag{1}$$

where r is the particle radius, k the Boltzmann constant, T the temperature and P the driving pressure. The filtration number can be considered as the ratio of the energy needed to bring a particle from the membrane surface to the bulk suspension to the thermal energy of the particle. It is shown that for N_F lower than 15, the membrane resistance or the polarization layer resistance dominate, whereas when N_F is higher than 15, the

cake layer resistance dominates. In this study, for all runs the filtration number was considerably larger than 15. Although this theory was established for spherical non-interacting particles which do not penetrate the active layer, it reinforces the assumption about the importance of particle deposition. Figure 7 shows particularly that the filtration of prefiltered wastewater through the same M14 membrane allows us to recover a flux close to the tap water flux.

Nevertheless, part of the fouling could be due to metabolites even if the suspended solid concentration is low (Bowen *et al.*, 1996). The fouling mechanism of such a water is definitely too complex to be modelled by pure physico-chemical considerations. A phenomenological chemical engineering approach could then be helpful.

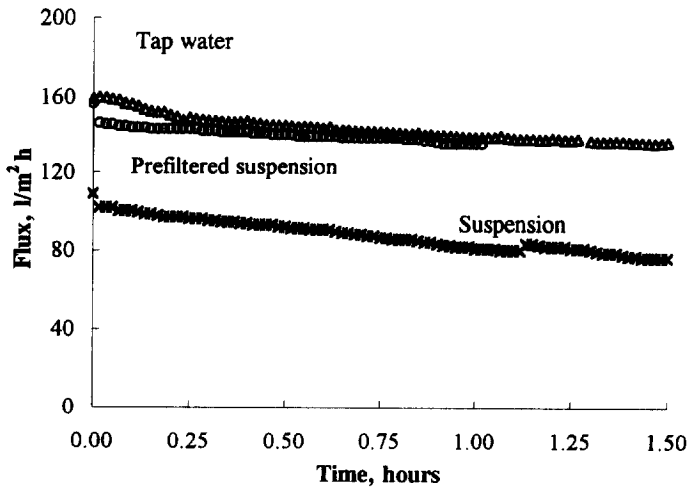


Figure 7. Flux against time for tap water, suspension and prefiltered suspension (1 bar and 3 m/s).

Interpretation by dimensional analysis

At steady state, the flux is given by:

$$J_s = \frac{P}{\mu (R_m + R_f)} \quad (2)$$

Factorial analysis of the mass, length and time dimensions shows that two dimensionless quantities can be obtained (Asaadi and White, 1992); $\rho u^2/P$ and $\mu R_f u/P$. The quantity $\rho u^2/P$ similar to the inverse of an Euler number or of a Number of Energy Units (Le Goff, 1979) is called the shear stress number N_s , whereas $\mu R_f u/P$ is equal to u/J_f . J_f being the flux through the fouling layer; this number therefore compares the flux transported by cross-flow to the flux through the fouling layer and is called the fouling number N_f . The experimental results recalculated in terms of these dimensionless groups gave a satisfactory plot where two parts could be put into evidence (Fig. 8). When the shear stress number was less than 30 000, the fouling number decreased linearly. For higher values of N_s , the fouling number remained steady at about 0.04 which meant that the fouling could not any more be decreased by cross-flow. This was probably related to bacteria or metabolite adsorption against the membrane wall. While treating data obtained in microfiltration of inorganic solid particle suspensions, Asaadi and White (1992) found straight lines with negative slope, the intersection with the N_x -axis Ghaffor (1996) who filtered hydrocarbon and biological mixed suspensions found straight lines with positive slope putting into evidence the impossibility of elimination of fouling. Both numbers N_s and N_f do not take into account, at least explicitly, the physico-chemical interactions

between the suspension and the membrane and therefore do not look a satisfactory approach to such a complex operation as microfiltration but the good correlation between them allows us to predict easily the steady flux for different values of operating variables.

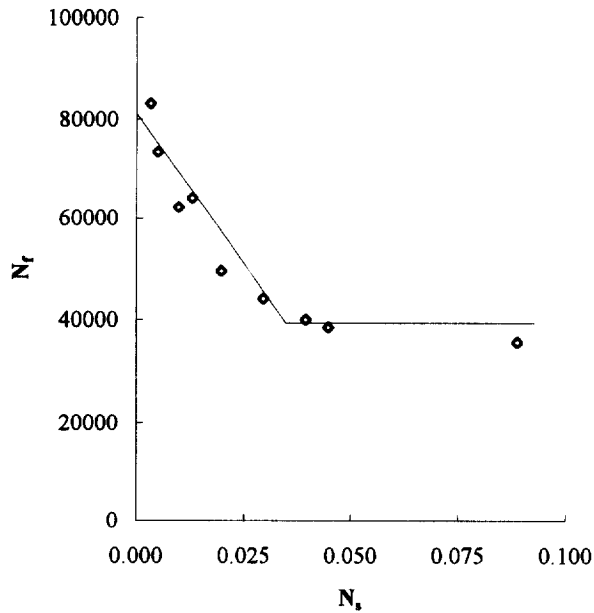


Figure 8. Fouling number against shear stress number (all experimental conditions).

CONCLUSIONS

The M14 membrane was a total barrier for suspended solids and eliminated 92% of the turbidity.

All the indicators of fecal contamination were eliminated: total coliform, fecal coliform and fecal streptococci. The permeate could then be considered as disinfected and in conformity with the quality specifications for agricultural use in Tenerife.

The total COD abatement was about 60% and essentially concerned species larger than 0.22 μm .

The M14 membrane showed a critical flux of about 100 $\text{l/m}^2 \text{ h}$ at 1 bar and 3 m/s cross-flow velocity.

The flux decline was essentially due to a particle deposition and it can be significant eliminated operating at 1 bar driving pressure and 3 m/s cross-flow velocity.

All the experimental results could be plotted on one only curve using dimensionless groups called respectively the shear stress number and the fouling number. This curve allowed us to predict the flux in any operating conditions.

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