Characterisation of RO fouling in an integrated MBR/RO system for wastewater reuse

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ABSTRACT

Membrane filtration has gradually gained acceptance as the preferred pre-treatment for reverse osmosis (RO). In this paper, an integrated membrane bioreactor (MBR)/RO system for wastewater reuse treating real sewage water has been evaluated and the RO fouling has been characterised. The MBR achieved low values of organic matter, total nitrogen, PO_4^{3-}, total organic carbon, turbidity and conductivity. Filtration with two different RO commercial membranes was performed after the MBR pre-treatment and the same average fouling rate (0.08 bar day^{-1}) was noted. These results gained from the characterisation of the high quality MBR/RO permeate show its potential for water reuse. Inorganic precipitation appears to be the predominant form of fouling in the RO membranes. Calcium phosphate and alumino-silicates were identified by a scanning electron microscope combined with an energy dispersive X-ray and polysaccharides, amide and aliphatic structures were detected with attenuated total reflection infrared microspectroscopy.

Key words | biological fouling, membrane bioreactors, microfiltration, permeate characterisation, reverse osmosis

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INTRODUCTION

The problem of water scarcity has generated great interest in new and alternative water sources in many Mediterranean regions. The focus has been on both wastewater reuse, and reclamation and desalination. The goal of wastewater reuse and reclamation is to capture water from non-traditional sources such as municipal or industrial wastewaters, and treat it in accordance to its subsequent uses. Within this framework, the use of membrane bioreactors (MBR) coupled with reverse osmosis (RO) membranes is becoming an increasingly attractive option for wastewater treatment and reclamation due to the high efficiency, ease of operation and cost effectiveness of this combination of technologies (Reith & Birkenhead 1998; del Pino & Durham 1999; Marti et al. 2011).

An MBR combines conventional activated sludge process and membrane separation. An MBR removes most of the organic matter and nutrients from raw wastewater (Monclús et al. 2010). However, very few published studies have

illustrated whether or not the permeate of an MBR treating real wastewater provides adequate quality to be directly treated by the RO membranes (Tam et al. 2007; Bonnélye et al. 2008; Dialynas & Diamadopoulos 2009) for further removal of dissolved solids, and organic and inorganic matter.

On the other hand, the main operational problem of RO is the fouling phenomenon resulting in a decrease in membrane permeability and a consequent increase in energy and chemical consumption. In terms of fouling components, the fouling can be classified into three different categories: biofouling, organic fouling and inorganic fouling. Inorganic fouling is caused by the precipitation of inorganic compounds. Organic fouling is due to the deposition of biopolymers (e.g. proteins and polysaccharides), humic acids, fragmentation compounds from the breakdown of humic substances in compounds of lower molecular weight, acids from the oxidation of organic carbon to carboxylic acids and neutral compounds of low molecular

weight (e.g. alcohols and oligosaccharides) on the membranes. Biofouling appears when microorganisms grow on the membrane surface (Fritzmann et al. 2007). The identification of the predominant agents in the feed water causing fouling on the membranes is an essential step in proposing efficient management strategies to minimise RO fouling, i.e. type of chemicals, doses and frequency of application, thus reducing losses of permeability and avoiding abrupt increases in working pressure.

The aim of this paper is to assess the use of an MBR as the pre-treatment for an RO system, and to provide further insights into which kind of fouling is predominant in the RO membrane.

MATERIALS AND METHODS

MBR/RO pilot plant

This study was carried out in a coupled MBR/RO pilot plant treating wastewater (4.3 m^3/day) from the sewer system of the full-scale Castell-Platja d'Aro wastewater treatment plant (WWTP) (Catalonia, NE Spain), previously described in Dolar et al. (2012). The plant consists of a primary wastewater settler followed by an MBR with UCT (University of Cape Town) configuration (i.e. anaerobic, anoxic, aerobic and membrane compartments) for biological nutrient removal. The membrane compartment holds 8 m^2 of flat sheet microfiltration (MF) membranes (LF-10, Kubota, nominal pore size of 0.4 μm). The MBR was operating with a hydraulic retention time (HRT) of 16 h, while the solid retention time was between 35 and 45 days during the experimental period. The MBR provides feed water for the coupled RO system, which is composed of one pressure vessel housing a 40 inch RO membrane along with an automatic cleaning system with RO permeate at low pressure, in case the water supply from the MBR should fail for any reason.

Two different commercial RO membranes from two of the major manufacturers of RO membranes were purchased for the study, namely a Ropur TR70-4021-HF (Toray Industries Inc., Japan), aromatic polyamide, spiral wound module and a Hydranautics ESPA2-4040 (Hydranautics, USA), composite polyamide, spiral-wound module. Details of the membranes are given on the manufacturers' websites, listed at the end of the paper.

ESPA2-4040 membranes have been previously demonstrated to be suitable for application in an integrated MBR/RO pilot test treating municipal wastewater (Freeman et al. 2011). According to the technical information provided by the manufacturer, TR70-4021-HF is characterised by a high permeate flow rate and therefore is expected to exhibit a relatively low fouling propensity.

The entire pilot plant was equipped with a programmable logic controller and a supervisory control and data acquisition system for digital and analogical data. The data acquisition and monitoring system of the RO system enables the monitoring of online flow rates (Sitrans Flowmeter Magflo Siemens, Denmark), conductivities (Digital Conductivity system 3400 sc, Hach Lange, Germany), temperature and pH (pHD sc Differential pH and ORP Analysis System, Hach Lange, Germany) of the influent and permeate streams, as well as both MF (MBR) and RO membrane pressures.

The mean temperature inside the reactor was 22.2 ± 4.9 °C during the entire sampling campaign. The pilot plant was lodged inside a container equipped with a thermal insulation and air conditioning system (heating and cooling system). While the temperature in the reactor varied with a maximum of ±2 °C during the day, the standard deviation nevertheless reflects the annual variation. Sludge from the Girona WWTP was inoculated 3 months prior to the RO fouling experiments being run. The MBR permeate was used as the feed stream to the RO membrane and had no additional treatment (i.e. dosage of antiscalants, acid or disinfection agents), in order to avoid any influence on the quick formation of fouling on the membrane surface. The initial set-point of the working pressure was positioned at 10 bar. See Table 1 for further information on the operating conditions of the coupled MBR/RO system.

Analytical methods

Analyses were performed according to standard methods (APHA 2005), i.e. total chemical oxygen demand (COD; APHA standard method 5220B), total Kjeldahl nitrogen (TKN; APHA standard method 4500-Norg.B), total organic carbon (TOC; APHA-S310), bicarbonate and carbonate (APHA standard method 2320B, titration method), dissolved cations (ionic chromatography ISO 14911 (ISO 1998)), dissolved anions (ionic chromatography ISO 10304–1 (ISO 2007)), turbidity (APHA standard method 2130), aerobic bacteria (ISO 6222 (ISO 1999)) and ammonium concentration (by boric acid (Büchi, Postfach, Switzerland; APHA standard method B324)). The ammonia in the distillate was determined with a titrimetric method (Tritino 719S Metrohm, Herisau, Switzerland) using H_2SO_4 and a pH meter. Nitrites (NO_2^--N), nitrates (NO_3^--N) and phosphates (PO_3^{-4}-P) were analysed using ion chromatography (Metrohm 761-Compact; APHA standard method 4110B). Total nitrogen (mg TN L^{-1})

Table 1 │ Operational conditions of the MBR/RO pilot plant during the experimental period

Parameters	Units	Experimental period
Operational period	days	358
HRT	hours	14–18
MBR permeate flux	L m^{-2} h^{-1}	16–21
Anaerobic recirculation	% of the inflow	129
Anoxic recirculation	% of the inflow	92
External recycle	% of the inflow	136
Wastage pump	% of the inflow	1.8
DO aerobic set point	mg O$_2$ L^{-1}	1.5
pH	–	7.3 ±1.6
Temperature	°C	22.2 ± 4.9
Permeate/relaxation time	minutes	9/1
RO recovery	%	40–60
RO membrane pressure	bar	10–15

was calculated as the sum of TKN, nitrite and nitrate concentrations.

The salt rejection was calculated with the following formula:

$$\% \, \text{Rejection} = \frac{W_F - W_P}{W_F} \times 100$$

where W_F is the salt concentration in the feed stream and W_P is the salt concentration in the permeate stream.

The recovery was calculated with the following formula:

$$\% \, \text{Recovery} = \frac{Q_P}{Q_F} \times 100$$

where Q_P is the flow rate in the permeate stream and Q_F is the flow rate in the feed stream (Fritzmann *et al.* 2007).

Fouling characterisation

Membrane autopsies were performed at the end of the RO operation to investigate the fouling formation on the membranes' surface. The measurements of the inorganic and organic fractions were performed at the end of each test. For the inorganic fouling a scanning electron microscope (SEM) (Ivnitsky *et al.* 2005) (Stereoscan S360, Cambridge Instruments) combined with energy dispersive X-ray (EDX) analysis (INCA Energy Serie200, Oxford Instruments) was used. In the case of organic fouling, the samples were characterised using a Fourier transform infrared (IR) microscope (Illuminate IR, Smiths detection)

interfaced with a mercury–cadmium–telluride photoconductive detector and equipped with a microscope with a motorised x-y stage, 20× and 50× objectives, and a slide-on attenuated total reflection diamond objective (Smiths detection). The membrane samples were placed on the stage of the microscope and a specific position was selected with the assistance of the live camera (Leica OM 2500, Modulo FT-IR, Renishaw plc, Wotton-under-Edge, UK). The microscope was software-controlled using Wire 3.2 version software (Renishaw plc, Wotton-under-Edge, UK). The spectra acquired ranged between 4,000 and 800 cm^{-1} wavelengths with 4 cm^{-1} resolution. A background scan was taken before every sample scan to prevent the environment from affecting the sample spectrum. The spectrum of each sample was obtained by taking the average of 128 scans to improve the signal-to-noise ratio. The spectrometer was completely software controlled by synchronised IR basic version 1.1 software (SensIR Technologies, Smiths detection). The time required to take one spectrum measurement was about 40 s.

RESULTS AND DISCUSSION

Performance of membrane processes

The coupled MBR/RO system was operated for 358 consecutive days (Figure 1). The experiment started with a chemical cleaning of the MF membranes and then a second chemical cleaning was needed after 5 months of operation, when the transmembrane pressure reached 180 mbar. The daily mean values of the total suspended solids ranged between 4 and 11 g L^{-1} due to the seasonal variation in treating real domestic influent. Indeed, simultaneously with this study, a successful validation of an automatic membrane air-scour control method (Ferrero *et al.* 2011) was carried out from the beginning of the 6th month until the end of the experiment. No noteworthy negative interactions occurred with the membrane performance when the air-scouring was automatically regulated. The permeate characterisation of the MBR is shown in Table 1.

Two different RO membranes were tested. First, a TR70 membrane was used over 35 days and later an ESPA2 membrane was used over 58 days, before the RO system was stopped and the membranes were extracted to perform the autopsies.

Figure 2(a) illustrates the TR70 membrane performance. It can be seen that membrane pressure increased during the entire period, starting at 10 bar and reaching 12.5 bar in 29
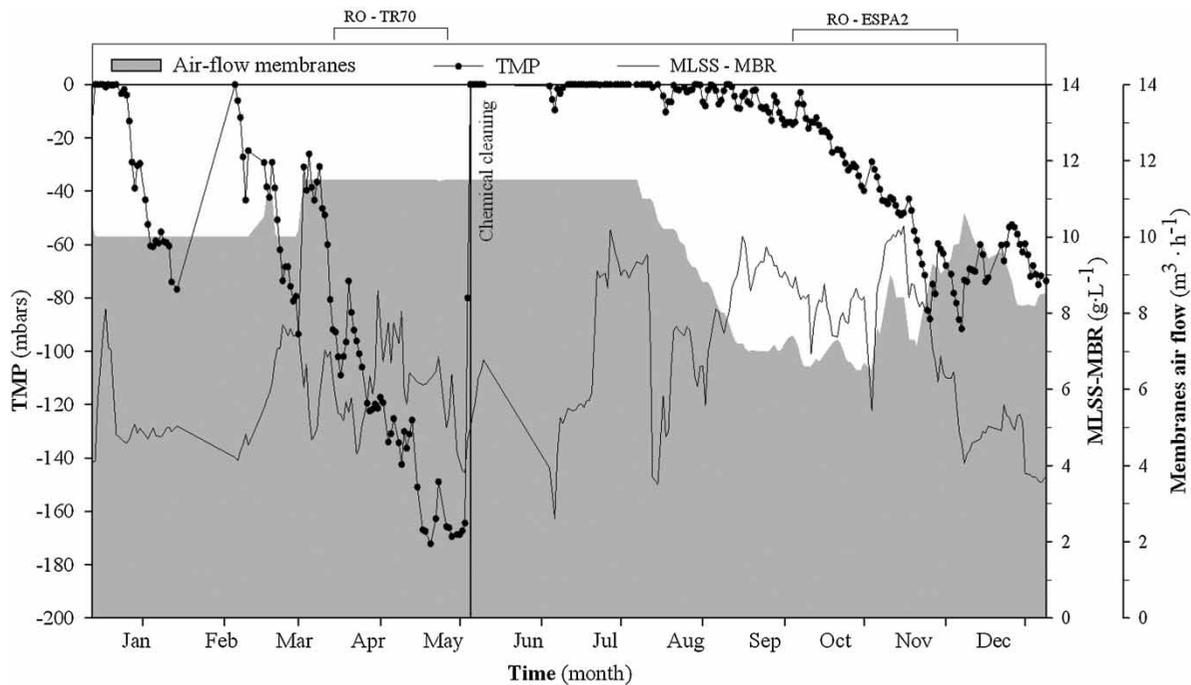
Figure 1 | MBR operation performance (MLSS: mixed liquor suspended solids).

days (0.086 bar day^{-1}). Thus, on day 29, a pressure rectification was applied and the pressure was reduced to 10 bar. After that, a rapid pressure increase was noted again, so it was decided to stop the process and remove the membrane because the pressure had increased by more than 15% over the initial starting pressure. On the other hand the salt rejection decreased, although it was over 99% during the entire experiment, and the permeate flow rate remained constant (200 L h^{-1}).

ESPA2 membrane performance is presented in Figure 2(b). It shows that the pressure also increased over the entire period, starting at 9.5 bar and achieving values of up to 15 bar after 58 days (an average rate of 0.086 bar day^{-1}). At the beginning of the ESPA2 experiment, a faster increase of the pressure (1 bar day^{-1}), as a result of the membrane compaction, was noted. Some authors have described membrane compaction in previous studies (Ladner *et al.* 2010). Membrane compaction shows a quick increase in the working pressure and a decrease in the permeate production. A manual pressure rectification was applied in the experiment, reducing the pressure to 10 bar thus allowing the membrane to depressurise and
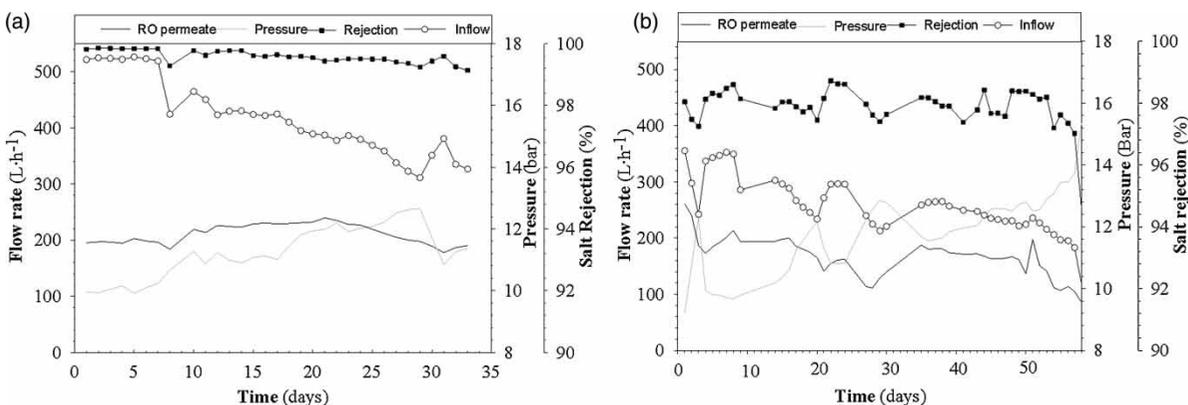

Figure 2 | Performance of the TR70 Ropur (a) and ESPA2 hydranautics (b) membranes.

consequently recover the permeate production. Another possible cause for the initial loss of permeability could be the adsorption of organics onto the membrane surface. This phenomenon is common in any RO system treating municipal waste water, as reported in the surveys carried out by membrane manufacturers (http://www.membranes.com). By the end of the experiment, the membrane pressure had increased by more than 50% over the initial starting pressure.

The salt rejection varied between 98 and 96%, similar to the inflow variation, due to the manual pressure rectification actions. The RO permeate flow rate was initially 200 L h^{-1} but decreased to 100 L h^{-1} at the end of the test; hence the RO system recovery being reported as a range in Table 1. During both tests, the increase in working pressure due to the build-up of fouling led to a decrease in the influent flow rate delivered by the centrifugal pressure pump, according to the pump's characteristic curve. In fact, it was not equipped with a frequency driver. The difference in the permeate flow rate for the two membranes is probably due to the intrinsic characteristics of the TR70–4021-HF, which allows a higher flow and, hence, is likely to be less prone to abrupt loss of permeability.

Both RO membranes showed the same average fouling rate (0.086 bar day^{-1}). According to Freeman *et al.* 2011, this value is below a severe fouling rate, as only after 35 days in the case of the TR70 and 58 days in the case of the ESPA2 were clear symptoms of fouling shown.

Influent and effluent water characterisation

The water characterisation values of the WWTP influent, MBR permeate and RO permeate are summarised in Table 2. The selected WWTP sewer receives wastewater of domestic (95%) and industrial (5%) origin from the Santa Cristina d'Aro municipality (located in the middle of Costa Brava). The C:N:P ratio of the influent was 100:13:0.92, with insufficient organic matter for a successful nitrogen removal (Metcalf & Eddy 1991). Despite this limitation, biologic removal achieved a 74% removal of total nitrogen and a 46% removal of phosphorous. Ammonium and nitrate were then retained by the RO membranes, with rejections higher than 90%. In addition, the MBR permeate showed low levels of TOC (4.54 mg C L^{-1}), turbidity (0.475 NTU) and conductivity (793 µS cm^{-1}). The coupled MBR/RO system showed excellent removal rates (>96%) for all compounds, in agreement with other studies (Häyrynen *et al.* 2008).

The MBR/RO permeate presented high quality reusable water throughout the entire experimental period. This quality fulfils all the requirements (except for two parameters, *Escherichia coli* and nematodes, which were not evaluated in this study) of the Spanish legislation for reuse of reclaimed water (RD 1620 2007).

Characterisation of the RO fouling

Analysis of the deposit from the membranes' surface

The results of the analysis of the deposits on the surface of the membranes are shown in Figure 3. With respect to the fouling type classification, in this study the contribution due to biofouling has been included in the organic category. The predominant fouling compounds in both membranes were organic matter (33.82% for TR70 and 35% for ESPA2), followed by HCl-insoluble matter (20.37 and 36%, respectively) and finally the rest of the precipitates: calcium, magnesium, carbonates, aluminium, iron, phosphate and others (45.82 and 29.12%, respectively). The high concentration of HCl-insoluble matter at ESPA2 can be attributed to differences in the sewage water. Heavy rains occurring locally during the study probably caused these differences, although further research would have to be done to confirm this opinion.

Characterisation of the organic fouling

The IR spectra of the different membranes are shown in Figure 4. The TR70 spectrum shows a broad absorption band at 1,643 cm^{-1} linked to Amide I group vibrations of peptides and β-pleated secondary protein structures. The C–O–C stretching polysaccharides can be seen at 1,047 cm^{-1}. The ESPA2 spectrum shows the presence of Amide I group at 1,636 cm^{-1}, and the band at 1,023 cm^{-1} shows the presence of C–N aliphatic amines. According to (Ivnitsky *et al.* 2005), the presence of polysaccharide, amide and aliphatic structures reflects a defined biofilms structure.

Characterisation of the inorganic fouling

SEM images were taken to reveal the morphology of the fouled membrane. Figure 5 presents the SEM images from the fouled membranes. Figure 5(a) shows the TR70 fouled surface membrane, which contains a fine matrix with encrusted rounded forms. Figure 5(b) shows that the surface of the ESPA2 membrane was significantly different: it was covered with a fine matrix with a grey relief and large depositions on top.

Table 2 | Water characterisation of the MBR and RO

Parameters	Units	Influent (Mean ± SD)	MBR permeate (Mean ± SD)	% Removal	RO permeate (Mean ± SD)	% Removal
COD_{total}	mg L^{-1}	327 ± 234	25 ± 17	92	n.d.	100[a]
$COD_{soluble}$	mg L^{-1}	141 ± 88	–	–	–	–
NH_4^+	mg N-NH$_4^+$ L^{-1}	35 ± 24	2.09 ± 2.50	94	n.d.	100[a]
NO_3^-	mg N-NO$_x$ L^{-1}	0.5 ± 0.7	8.75 ± 5.95		n.d.	100[a]
Total nitrogen	mg N L^{-1}	43 ± 25	11.16 ± 6.35	74	n.d.	100[a]
PO_4^{3-}	mg P-PO$_4^{3-}$ L^{-1}	2.94 ± 1.45	1.58 ± 1.36	46	n.d.	100[a]
TOC	mg C L^{-1}	21 ± 16	4.54 ± 2.31	78	0.76 ±0.54	96
TC	mg C L^{-1}	101 ± 25	49.72 ± 10.04	51	–	–
IC	mg C L^{-1}	79 ± 13	45.1 ± 9.9	43	–	–
pH		–	7.65 ± 0.21	–	7.03 ±4.9	–
Conductivity	μS cm^{-1}	–	793 ± 125	–	14.7 ±10.4	98
Calcium	mg Ca L^{-1}	–	71 ± 25	–	n.d.	100[a]
Magnesium	mg Mg L^{-1}	–	14.5 ± 4.9	–	n.d.	100[a]
Sodium	mg Na L^{-1}	–	89 ± 19	–	n.d.	100[a]
Potassium	mg K L^{-1}	–	12.5 ± 0.7	–	n.d.	100[a]
Sulphate	mg SO$_4$ L^{-1}	–	90.5 ± 24.7	–	n.d.	100[a]
Chloride	mg Cl L^{-1}	–	54.5 ± 60.1	–	n.d.	100[a]
Fluoride	mg F L^{-1}	–	n.d.	---	n.d	100[a]
Bicarbonates	mg HCO$_3$ L^{-1}	–	164 ± 27	–	n.d.	100[a]
Carbonates	mg CO$_3$ L^{-1}	–	0	–	0	–
Silicium	mg SiO$_2$ L^{-1}	–	10.85 ± 7.28	–	n.d.	100[a]
Phosphate	mg PO$_4$ L^{-1}	–	2.07 ± 2.4	–	n.d.	100[a]
Manganese	mg Mn L^{-1}	–	0.02 ± 0.003	–	n.d.	100[a]
Temperature	°C	–	22.2 ± 4.9	–	–	–
Aerobic bacteria (22 °C)	Ufc mL^{-1}	–	1.73×10^3	–	4.69×10^2	73
Turbidity	NTU	–	0.475 ± 0.34	–	0.14 ±0.02	71

n.d. – not detected (below detection limit).
[a]Approximate.
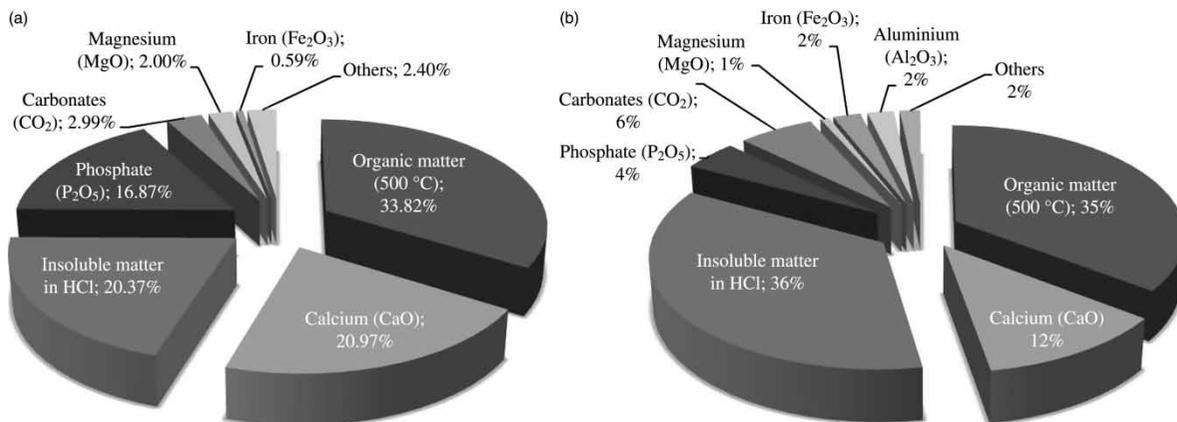
Figure 3 | Deposit composition from membrane surfaces: TR70 (a) and ESPA2 (b).
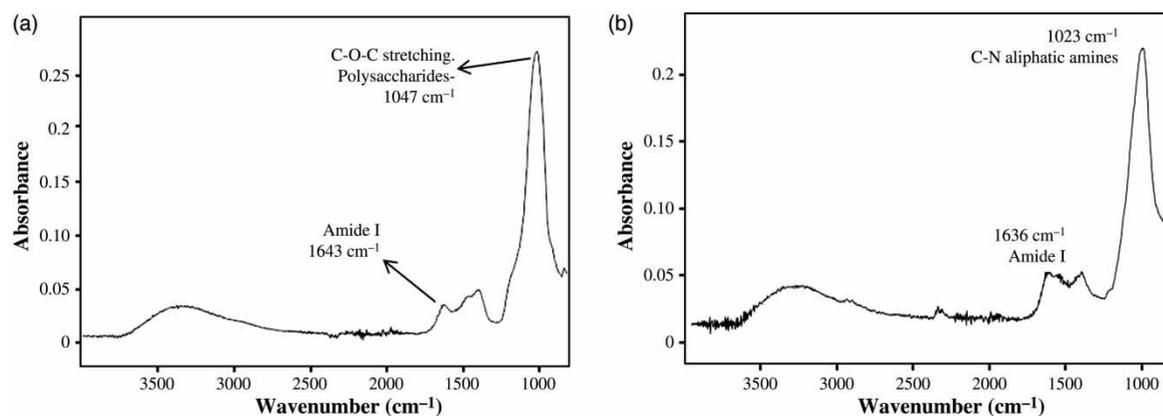
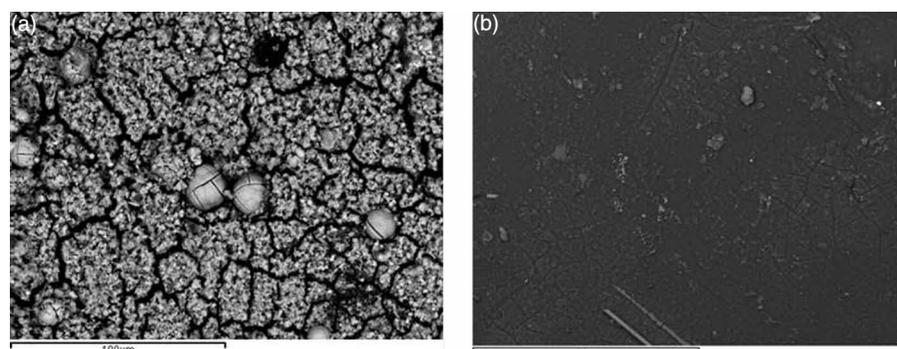**Figure 4** │ Typical attenuated total reflection infrared spectra of the TR70 membrane (a) and ESPA2 (b).

Figure 5 │ SEM images of membrane: (a) TR70 membrane (650×), (b) ESPA2 membrane (75×).

In order to identify the chemical components of the surface fouled layer, elemental analysis was performed by EDX analysis. For TR70, the composition of the rounded forms corresponds mainly to calcium (Ca) and phosphorous (P), and the matrix was composed of silicon (Si), magnesium (Mg) and aluminium (Al). For ESPA2, the large depositions correspond to calcium, phosphorous and magnesium. The grey relief was identified as magnesium, aluminium, silicon, phosphorous, calcium and iron. In this analysis a sulphur element was also detected. It may be a component of the polysulfone layer of the ESPA2 membrane material.

The presence of Ca and P on the membrane surface may be because the MBR treats domestic wastewater. According to Jacob (2011), phosphate in the feed water of an RO system results in calcium phosphate fouling. Phosphate is multivalent and takes many different forms with varying solubility. The crystallisation is very difficult and the most common form is as an amorphous deposit on the membrane surface.

Fe, Al, Mg and Si on the membrane indicate the presence of aluminosilicates (clay). They rapidly attach to the membrane and cause a reduction in flux and an increase in work pressure as the permeability of the membrane is reduced. Clay comes from surface waters, rivers and reservoirs but can also find its way into sewers. The MBR pretreatment is not 100% efficient in removing them.

CONCLUSIONS

An integrated MBR/RO system for wastewater reuse treating real sewage water has been evaluated and the RO fouling has been characterised. This study shows that the MBR system provides an adequate pre-treatment for the RO unit; low average values of organic matter, total nitrogen, phosphates, TOC, turbidity, and conductivity were attained. The same average fouling rate (0.086 bar day^{-1}) was achieved testing two different RO membranes (Ropur TR70-4021-HF and Hydranautics ESPA2-4040).

The permeate of the MBR/RO system shows high quality standards for wastewater reuse for water reclamation purposes.

Inorganic precipitation appears to be the predominant form of fouling in the RO unit, followed by organic matter and then HCl insoluble matter. Furthermore, calcium phosphate and aluminosilicates were identified by the SEM-EDX analysis as the main inorganic foulants, while attenuated total reflection infrared microspectroscopy analysis detected polysaccharides, amide and aliphatic structures as the main organic foulants, thus reflecting the build-up of a biofilm structure on the membrane. Aluminosilicates were detected when the RO membrane autopsies were carried out, showing that the MBR pretreatment is not 100% efficient in their removal from wastewater.

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