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Direct water reclamation from sewage using ceramic tight ultra- and nanofiltration

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ABSTRACT

Sewage is a nutrient rich reliable water source that is rather consistent in quality, volume and temperature, and is available in large amounts in urban areas. Decentralised reclamation of water including its constituents from municipal sewage, further referred to as sewer mining, is a concept in which municipal sewage is considered a resource instead of a waste stream.

In this research, water reclamation in the sewer mining concept was studied using ceramic tight ultra- (UF) and ceramic nanofiltration (NF). In our current approach, ceramic membrane filtration is proposed as pre-treatment for reverse osmosis (RO) to produce demineralised water for industries from municipal sewage. The objectives of this research are to study (i) the membrane performance, (ii) the organic matter and ion rejection, and (iii) the biofouling potential of RO using permeate water from the ceramic filtration.

The application of ceramic tight UF and ceramic NF for direct treatment of domestic sewage has been demonstrated in this study. The cross flow ceramic tight UF and NF fed with filtered sewage, can be operated for 1–4 days without any cleaning required. The membrane performance remained high with chemical cleaning with NaClO (0.1%) and HCl (0.1 mol L^{-1}) solutions. On average about 81% of organic matter was rejected by both ceramic tight UF and NF membranes. Finally, the pressure drop increase in the MFS fed with ceramic NF permeate was low during an operation of 14 days. These results were comparable with the increase in pressure drop of an MFS fed with Dutch drinking water.

1. Introduction

Sewer mining is a decentralised water reclamation concept, where municipal sewage is considered a resource instead of a waste stream. In this concept, the sewage flow is fully or partly captured and directly treated for on-site usage, usually for non-potable purposes [4]. There is a growing interest in sewer mining concepts as a possible economic and sustainable alternative for the currently applied decentralised sewage treatment plants (STPs), while recovering nutrients, water and energy from the sewage flows.

Reclamation of treated sewage for drinking water purposes is a much more difficult approach than for non-potable use due to psychological barriers and the perceived health risk [21]. However, sewer mining is considered a breakthrough approach for the production of industrial water, minimising competitive fresh water claims in urban areas. In northern and central Europe, the majority of fresh water supply is used for industrial water [33], whereas for many applications, industrial water requires a low salt concentration and low hardness, i.e. demineralised water [3].

Reverse osmosis (RO) is commonly applied to produce demineralised water, considering its high rejection of impurities, manageable costs, and ease of operation [2,8,21]. However, RO membranes are sensitive to fouling which declines its efficiency, performance and salt rejection. Four types of fouling can be defined; particulate fouling, organic fouling, inorganic fouling (i.e. scaling), and biofouling [17]. Biofouling is caused by biological growth on the membrane and in the feed spacer. Not only the biofilm itself but also the extracellular polymeric substances (EPS) produced by the microorganisms, can deteriorate the membrane performance [9,26,28]. Membrane fouling is directly linked to the feed water quality, therefore extensive pre-treatment of RO-feed water is required [6,17].

Current RO-based water reclamation consists of conventional sewage treatment followed by multimedia filtration (MMF), microfiltration (MF) or ultrafiltration (UF) and RO [2]. In the

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conventional treatment, municipal sewage is usually led through primary settlers, activated sludge process, secondary settlers, whereas the sludge is stabilised in a digester prior to dewatering and disposal (Fig. 1) [25,34]. However, the conventional RO-based water recycle technology requires a large footprint and does not make optimal use of energy, water and nutrient reuse [19,27]. Therefore, alternative treatment concepts are in demand.

Ravazzini et al. [20] and Sayed et al. [24] suggested to disregard the conventional sewage pre-treatment by treating sewage directly with polymeric UF and nanofiltration (NF). However, they found that this process is not economically feasible, due to the duration of the membrane cleaning. The layer of polymeric membranes is very sensitive to chemical cleaning with the use of extreme pH, temperature or chemical concentrations [1,5,13]. Since the concentration of chemicals is directly linked to the duration of the chemical cleaning process, treating the membranes with low chemical concentrations will increase the duration of the chemical cleaning. Sayed et al. [24] found that due to the severe clogging of the membranes using sewage as feed water, chemical cleaning with a duration of 8 h was required after a filtration time of 8 h including hydraulic backwashing. This means that the filtration and relative production downtime are similar.

Forward osmosis (FO) is a new technique that can be used as an alternative water reclamation step in the proposed sewer mining concept [15]. FO is a membrane separation technique based on osmotic pressure; the feed solution is driven through the membrane by a draw solution that has a higher ion concentration than the feed solution. FO consumes much less energy than other membrane techniques, since osmotic pressure is the driving force and no additional pressure is required [14]. The permeate water can be reclaimed with RO, and FO is a suitable pre-treatment step for RO [11,15,35]. However, in order to remove water from sewage, a high ion concentration in the draw solution of FO is required. Due to the high ion concentration, a high pressure RO is necessary, which consumes a lot energy [11]. Furthermore, implementation of the FO is limited by its relatively low flux [18].

Ceramic tight UF or ceramic NF is considered a potential alternative of interest for water reclamation in the sewer mining concept [26]. Ceramic membranes, compared to polymeric membranes, are robust; they have a high mechanical strength, a high chemical and thermal resistance, and a homogeneous distribution of narrow pores [32]. The membrane is expected not to be damaged by high pressure, high temperatures or chemicals, enabling high pressure backwash and vigorous chemical cleaning of the membrane. Other benefits are the long life of the membrane (>15 years) and the recyclability of the membrane material. To date, ceramic membranes are less frequently utilised than polymeric membranes due to their higher price per m^2 [26]. In literature, conclusive definitions cannot be found to distinguish ceramic UF from ceramic NF. In this research, the following definitions were used based on the molecular weight cut-off (MWCO) of the membranes: ceramic membranes with a MWCO between 500 and 3000 Da are defined as tight UF membranes, and those with a MWCO smaller than 500 Da as NF membranes.

In this paper, the concept of the production of industrial water from raw sewage in small residential areas using ceramic tight UF or ceramic NF is presented. Fig. 1 gives an overview of the concept in which the conventional RO-based water reclamation process is replaced by a fine sieve, ceramic filtration and RO. This decentralised water treatment can supply demineralised water to nearby located industries, and the resultant cost saving in water transportation can be expected. In our proposed concept, the raw municipal sewage passes firstly through a coarse sieve and grit removal followed by a fine sieve of 1 mm to remove the largest fraction of suspended solids [22]. Then, the pre-treated sewage is directly subjected to ceramic filtration, which serves as the pre-treatment step for RO. The concentrate from the ceramic membrane and the debris from the fine sieve can then be stabilised in anaerobic digester systems in which the organic matter is largely converted into biogas by, for example, an up-flow anaerobic sludge blanket (UASB) reactor. In conventional STPs, a large part of the organic matter is mineralised to CO_2 in the aeration tanks and subsequently released to the atmosphere. By using ceramic filtration combined with anaerobic digestion of the sewage organic matter, energy recovery from the sewage constituents is maximised [16,23,27].

Due to the high foulant load of the sewage, fouling in the ceramic membrane and in the subsequent RO membrane is expected to be the main challenge in this ceramic filtration concept. The aim of this paper is therefore to investigate the feasibility of this concept by determining the (i) performance of ceramic tight UF and ceramic NF in sewage filtration, (ii) rejection of organic matter and ions of the ceramic membrane, and (iii) biofouling potential of the RO with ceramic NF pre-treatment. The energy production from the concentrate of ceramic membranes will be studied in the future stages of research, but is not within the scope of this paper.

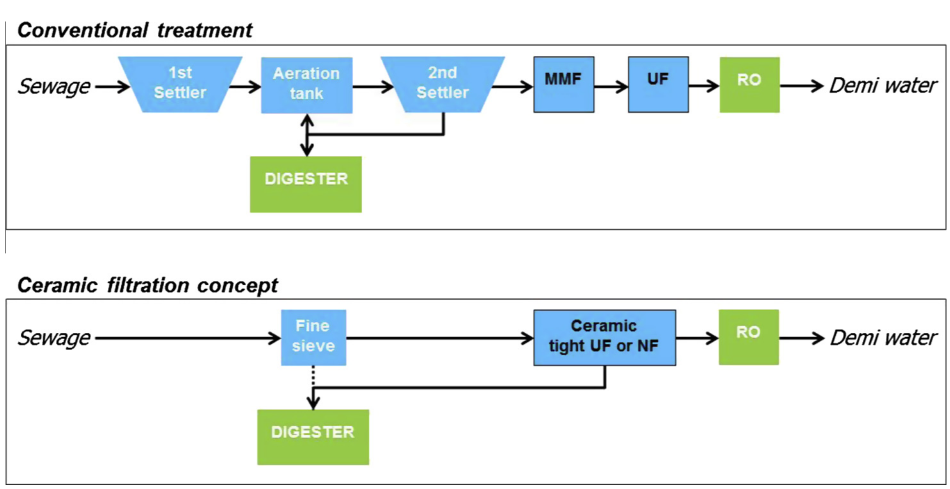

Fig. 1. Production of demineralised (demi) water from sewage using conventional treatment and the ceramic filtration concept. Raw municipal sewage first faces coarse sieve and grit removal before entering the first step of both treatment processes.

2. Materials and methods

2.1. Performance and cleaning

2.1.1. Membranes and filtration set-up

Ceramic tight UF and ceramic NF membranes were used during filtration in this research. Both types of membranes were made of TiO_2 and had a multi-channel configuration. Both ceramic tight UF and ceramic NF experiments were carried out with a cross-flow filtration system (Figs. 2 and 3). The filtration was conducted using a pneumatic diaphragm pump (Hydra-cell) with a pulsation dampener.

The ceramic tight UF membrane (TAMI Industry, France) had a MWCO of 3 kDa or mean pore size of 3.06 nm [26], 2 mm diameter channels, and an effective filtration area of 0.013 m^2. The ceramic NF membranes (Inopor, Germany) had a MWCO of 450 Da or mean pore size of 0.9 nm with 3.5 mm diameter channels, an open porosity of 30–40%, and an effective filtration area of 0.25 m^2. There were 4 ceramic NF membranes installed in tandem, which provided a total filtration area of 1 m^2.

The experiments using ceramic tight UF and ceramic NF membranes were conducted at different locations and with slightly different setup configurations:

The filtration experiments using the ceramic tight UF membrane were carried out in the Waterlab at TU Delft. A feed tank containing 50 L was installed. Both the permeate and the concentrate were fed back into the feed tank, except the sampling volume, which was a negligible amount (<0.1%). The chemical oxygen demand (COD) of the feed water remained constant over the duration of the experiments. Water temperature was controlled at

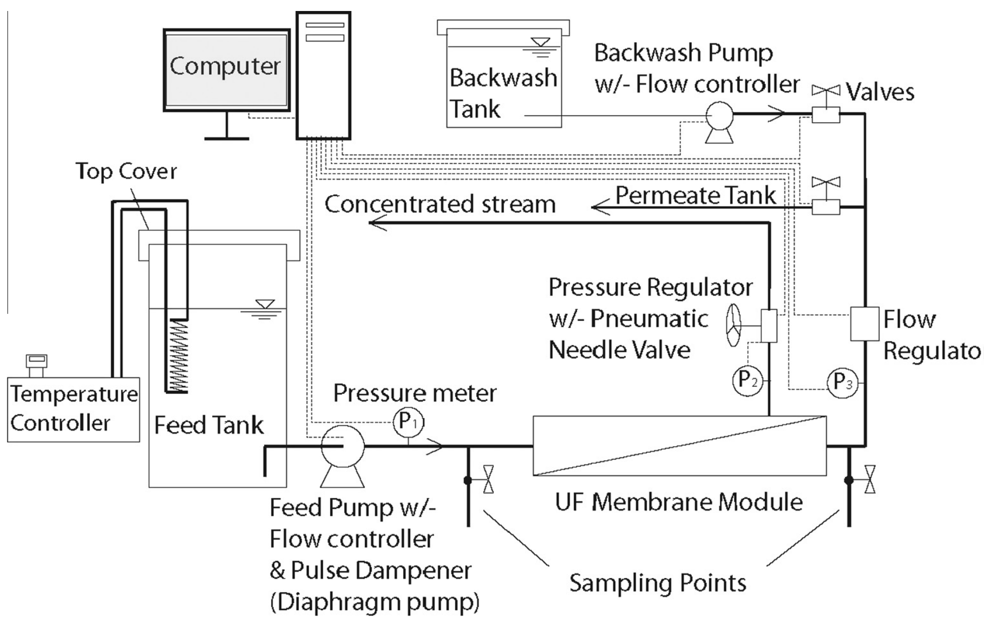

Fig. 2. Schematic representation of the cross-flow ceramic tight UF filtration system.

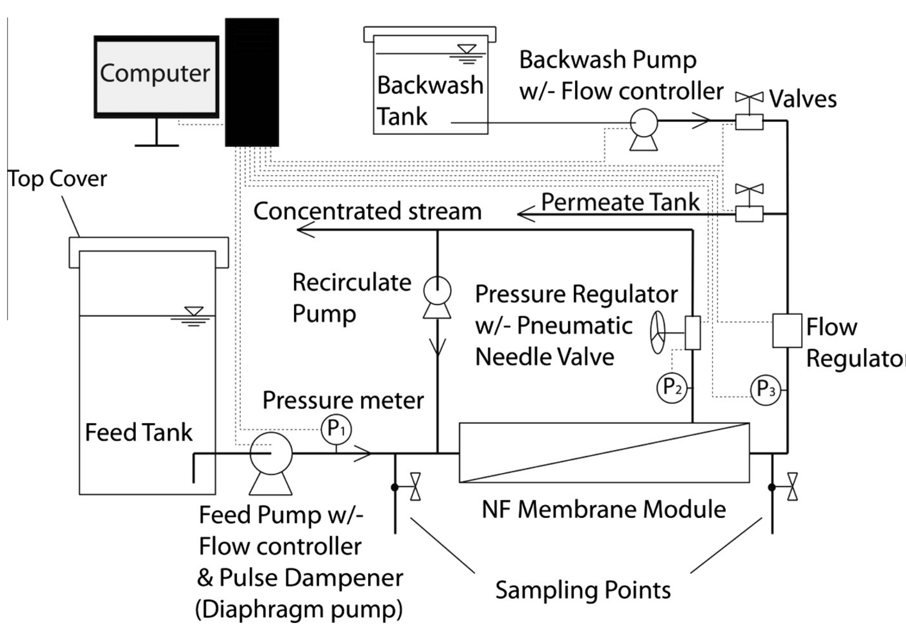

Fig. 3. Schematic representation of the cross-flow ceramic NF filtration system.

20 ± 1 °C using a temperature controller (Fig. 2). The single pass water recovery of the ceramic tight UF filtration systems was <1%.

The filtration experiments using the ceramic NF were conducted at Harnaschpolder STP in Den Hoorn, the Netherlands, due to the large filtration area of the ceramic NF membranes (1 m^2). The permeate- and concentrate streams were directly discharged to the influent stream of the STP. The NF filtration system used a recirculation pump to reach a water recovery of 50% (Fig. 3).

2.1.2. Raw sewage

Sewage, which was fed to the ceramic tight UF membrane, was collected on a daily basis, after the influent screening (6 mm) at the Harnaschpolder STP. The collected sewage (50 L) was transported to the Waterlab at TU Delft and pre-filtrated with a fine sieve of 1 mm mesh width, before it was fed to the membranes. Key physicochemical properties of the pre-sieved sewage are summarised in Tables 1 and 2.

The feed water to ceramic NF system was directly pumped from the well after the influent screening (6 mm) at the Harnaschpolder STP. Prior to feeding to the ceramic NF membranes, the sewage flew through a 0.5 mm sieve to remove granulates that may clog the membrane channels. Daily, 24 h mixed samples were collected by an automatic sampling machine for characterization.

Table 1
Key physicochemical properties of 1 mm-sieved sewage during experiments B and D (see Section 2.1.3) using the ceramic tight UF system.

Parameter	Experiment B, Fig. 5		Experiment D, Fig. 7				
	Day 1	Day 2	Day 1	Day 2	Day 3	Day 4	Day 5
Total COD (mg L^{-1})	632	348	383	863	444	670	527
Dissolved COD (mg L^{-1})	254	157	153	294	127	265	129
DOC (mg L^{-1})[a]	20	12	12	22	17	19	6.0
Conductivity (mS cm^{-1})	1655	924	790	1305	1321	1273	831
pH (–)	7.4	7.4	7.8	7.1	7.5	7.3	7.3
NH_4^+ (mg L^{-1})[a]	28	38	42	57	61	62	13
PO_4^{3+}-P (mg L^{-1})[a]	8.5	3.7	5.3	8.1	8.3	8.9	4.7
Mg^{2+} (mg L^{-1})[a]	21	10	8.4	15	16	14	9.2

[a] Concentration of dissolved fraction.

Table 2
Key physicochemical properties of 1 mm-sieved sewage during experiments A and C (see Section 2.1.3) using the ceramic NF system. Data measured by external laboratory of WWTP Harnaschpolder.

Parameter	Experiment A, Fig. 4				Experiment C, Fig. 6			
	Day 1	Day 2	Day 3	Day 4	Day 1	Day 2	Day 3	Day 4
Total COD (mg L^{-1})	700	590	610	590	420	420	480	430
TSS (mg L^{-1})	270	270	280	280	210	200	230	250
Total-N (mg L^{-1})	66	62.9	63.3	61.7	39	34	46	48
N-Kjehldahl (mg L^{-1})	66	62.9	63.3	61.7	39	34	46	48
Total-P (mg L^{-1})	9.4	8.9	8.8	8.8	5.1	5	6.5	6.5

2.1.3. Experimental protocol

The filtration experiments were carried out at a cross-flow velocity of 1 m s^{-1}. Three different cleaning methods were examined: (i) filtration with only hydraulic backwash, (ii) filtration with only chemical cleaning, and (iii) filtration with forward flush cleaning. The experiments testing different cleaning methods were performed with a constant feed pressure of 8 bar. Experiments were performed with 3 kDa ceramic tight UF and/or 450 Da ceramic NF membranes. Chemical cleaning was performed with both membrane types, whereas hydraulic backwash and forward flush were performed with ceramic tight UF and ceramic NF, respectively. Both hydraulic backwash and forward flush were determined to be ineffective, therefore the experiments were not repeated for the other type of membrane. Table 3 gives an overview of the performed experiments.

The effect of hydraulic backwash cleaning was tested using the 3 kDa ceramic tight UF (Table 3, experiment B). The hydraulic backwash was carried out using permeate water at a flux of 70 L/(m^2 h) for 3 min every hour with a total experimental duration of 21 h. Then, forward flush cleaning was assessed using the 450 Da ceramic NF membrane (Table 3, experiment C). In this experiment, continuous filtration for 96 h (3 days) was conducted with a release of the feed pressure for 5 min after every 24 h of filtration. After the 96 h of filtration the membrane was chemically cleaned twice. Next, the effect of chemical cleaning on both ceramic tight UF and NF membranes was examined, while different cleaning intervals were used (Table 3, experiment A and D). During experiment D the 3 kDa ceramic tight UF membrane and a 22 h cleaning interval was used, whereas during experiment A, the 450 ceramic NF membrane was chemical cleaned once at the end of the experiment after 115 h. Chemical cleaning was carried out with a NaClO solution (0.1%, 15 min backwash and 45 min soaking) followed by an HCl solution (0.1 mol L^{-1}, 15 min backwash).

The relative production downtime (RPD) (min/h) was calculated using the following equation:

$$\mathrm{RPD} = \frac{t_{\mathrm{cleaning}}}{t_{\mathrm{filtration}} + t_{\mathrm{cleaning}}} \tag{2.1}$$

where t_{cleaning} is the duration of the cleaning and $t_{\mathrm{filtration}}$ is the filtration time between cleaning two cleaning intervals (Table 3).

Membrane filtration performance was measured at a temperature-corrected permeability to 20 °C using the following equation:

$$L_{20^\circ\mathrm{C}} = \frac{J \cdot e^{-0.0239\cdot(T-20)}}{\Delta P} \tag{2.2}$$

where $L_{20^\circ\mathrm{C}}$ is the permeability at 20 °C (L/(m^2 h bar), T is temperature of water (°C), J is membrane flux (L/(m^2 h)), e^x is exponential function, and ΔP is transmembrane pressure (bar).

Table 3
Specifications of performed filtration experiments with different cleaning methods.

Experiment	Cleaning method	Ceramic membrane type	Constant pressure	Cleaning interval	Cleaning time	Total filtration time	Relative production downtime
A, Fig. 4	No cleaning	NF	No	115 h	–	–	–
B, Fig. 5	Hydraulic backwash	Tight UF	8 bar	1 h	3 min	21 h	3 min/h
C, Fig. 6	Forward flush	NF	8 bar	24 h	5 min	96 h	0.2 min/h
D, Fig. 7	Chemical cleaning	Tight UF	8 bar	22 h	1 h	120 h	2.6 min/h

2.2. Rejection of organic matter and ions

The morphology and elemental composition of the cake layer that formed on the membrane surface of ceramic tight UF was examined by removing and collecting the cake layer with forward flush after 21 h of ceramic tight UF (3 kDa). The cake layer was analysed using a scanning electron microscopes coupled with an energy dispersive X-ray (SEM-EDX) analyser (Ametek EDAXTSL). The samples were scanned at 10 kV accelerating voltage and 500× magnification. The scan area was approximately 0.6 × 0.6 mm and three random areas were measured on each sample. The composition of the cake layer was measured as described in the next paragraph.

The concentrations of organic matter and ions (nitrate, orthophosphate, sulphate, magnesium, and calcium) in the feed and permeate water from the ceramic tight UF membranes was measured to calculate the rejection percentages. This experiment was repeated 3 times using a ceramic NF membrane under similar conditions. During the first repetition using a ceramic NF membrane, the same parameters were measured. However, during the second and third repetition different parameters were measured, DOC and/or total phosphate were added and fewer ions were measured (no sulphate, magnesium and calcium).

Chemical oxygen demand COD was measured by COD test cells (Spectroquant). Water samples were filtered by 0.45 μm glass fibre syringe filters (Whatman) for the measurements of dissolved COD, dissolved organic carbon (DOC), and ions. The DOC was measured by a total organic carbon (TOC) analyser (TOC-VCPH, Shimadzu Instruments). Ions, including NH_4^+, PO_4^{3-}, Ca^{2+} and Mg^{2+}, were measured by ion chromatography (Metrohm Instrument). The pH and conductivity were measured using a multi-meter (WTW inoLab 720).

2.3. Biofouling potential in the RO membrane

The MFS [29] containing an RO membrane sheet (Filmtec BW30LE) was used to simulate the biofouling potential in spiral wound RO membrane systems. The MFS is proven to be able to mimic polymeric membrane conditions and to measure biofouling in the feed spacer of RO-membranes [7,10,12,31,30]. However, since no permeate is produced with the MFS, particulate fouling, organic fouling, inorganic fouling (i.e. scaling) cannot be simulated with the MFS.

The membrane element contained one RO spacer sheet with a thickness of 0.78 mm and a mesh size of 3 × 3 mm. The external dimensions of the MFS unit are 0.7 × 0.3 × 0.04 m and the effective membrane length and width are 0.20 × 0.04 m. The unit was covered to prevent daylight from affecting the biological growth. Permeate water of the ceramic NF experiment was used as feed water for the MFS. The feed water flow of the MFS was 16 L h^{-1}, and the cross flow velocity in the feed spacer was 0.14 m s^{-1}. The installation was set up in such a way that no air could come in the system, so oxygen was prevented from entering. The oxygen concentration, flow velocity and transmembrane pressure were continuously monitored during the experiment, which was carried out two times; one run was 7 days and the other run was 14 days. The results were compared with MFS results from Vrouwenvelder et al. [29].

3. Results and discussion

3.1. Performance and cleaning

Experiments were performed using a ceramic NF membrane; the results are presented in Fig. 4. The permeability reduced 58%

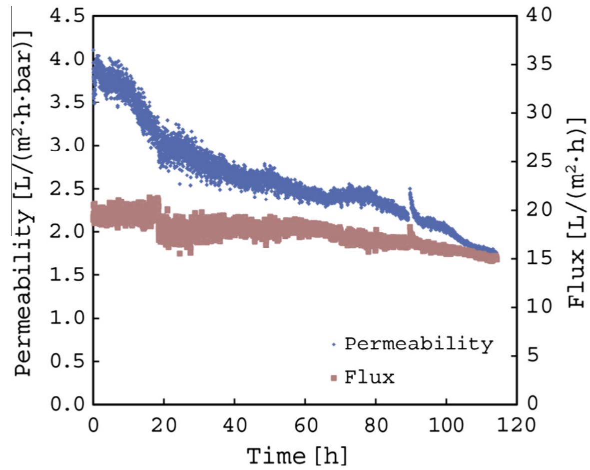

Fig. 4. Temperature corrected permeability and flux over time of ceramic NF filtration (450 Da) membrane during the sewage filtration. No membrane cleaning was conducted within the 120 h of filtration.

from 4.1 to 1.7 L/(m^2 h bar) in 4 days, with an average reduction rate of 0.02 L/(m^2 h bar) per hour without cleaning the membrane. Sayed et al. [24] studied direct sewage filtration with polymeric NF membranes. They observed a permeability reduction rate of 0.7 L/(m^2 h bar) per hour. Under similar filtration flux of 15–20 L/(m^2 h), the ceramic NF filtration exhibited a significant lower fouling rate compared to the polymeric NF system.

During the experiment the pump pressure was maximum 8.5 bar; this was reached after about 10 h, causing the flux to drop. The flux started at a constant value of 20 L/(m^2 h) and after 10 h slowly dropped to 15 L/(m^2 h) at the end of the experiment (at 115 h).

Hydraulic backwash is the most common used method in practice to remove the cake layer in MF and UF membranes [17]. Depending on the feed water quality, hydraulic backwashing is performed from every 15 min to once a day. In this study, 1 mm pre-filtered sewage was used as feed water. An hourly hydraulic backwash was applied on ceramic tight UF (3 kDa) at constant pressure. Fig. 5 shows that the permeability of the ceramic tight UF membrane with and without hourly hydraulic backwash is comparable during a period of 21 h. Thus, the hydraulic backwash was not effective in recovering the membrane permeability.

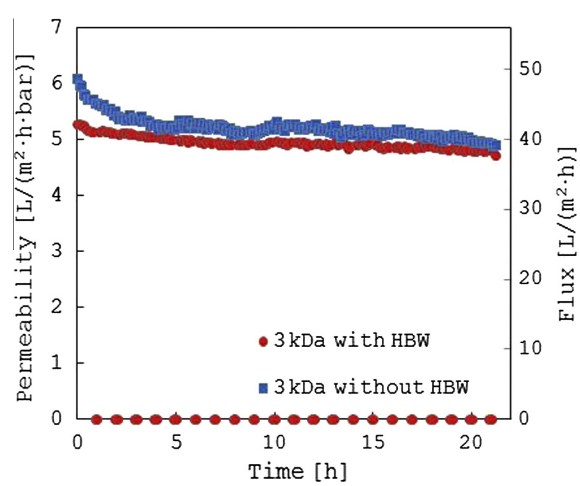

Fig. 5. Temperature corrected permeability and flux over time of ceramic tight UF filtration (3 kDa) membrane during the pre-filtered sewage filtration at constant pressure of 8 bar, operated with (red) and without (blue) hourly hydraulic backwash (HBW). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The performance of the ceramic tight UF membrane was still adequate after 21 h of filtration without cleaning. Therefore, filtration without hydraulic backwash is recommended to obtain a higher water recovery, because no permeate is used for backwashing.

The effects of daily forward flushing on the permeability of the 450 Da ceramic NF membrane was studied over a 4 days period at constant pressure. Fig. 6 shows that the permeability decreased from 5.9 to 2.5 L/(m^2 h bar) within the first 24 h and maintained above 2 L/(m^2 h bar), during the rest of the filtration period. The daily forward flush recovered the permeability with 22 ± 2%. Thus, using forward flush to remove part of the loose cake layer formed on the membrane surface did not significantly enhance the permeability of the membrane. After a continuous operation of 4 days, chemical cleaning was executed twice with NaClO (0.1%) and HCl (0.1 mol L^{-1}). The permeability after these chemical cleanings was measured to be 5.7 L/(m^2 h bar), resulting in a membrane recovery of 97%.

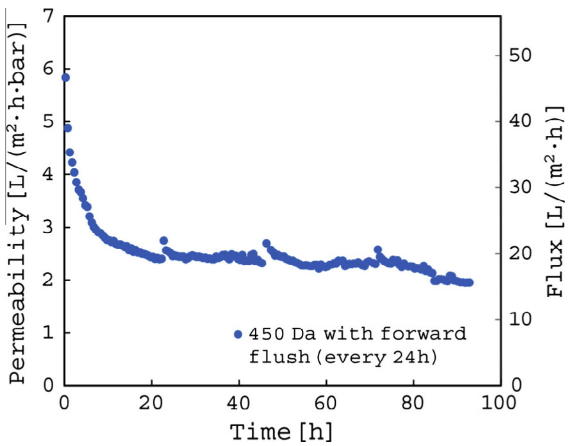

Fig. 6. Temperature corrected permeability and flux over time of ceramic NF filtration (450 Da) membrane during the pre-filtered sewage filtration at constant pressure of 8 bar, cleaned with forward flush per every 24 h.

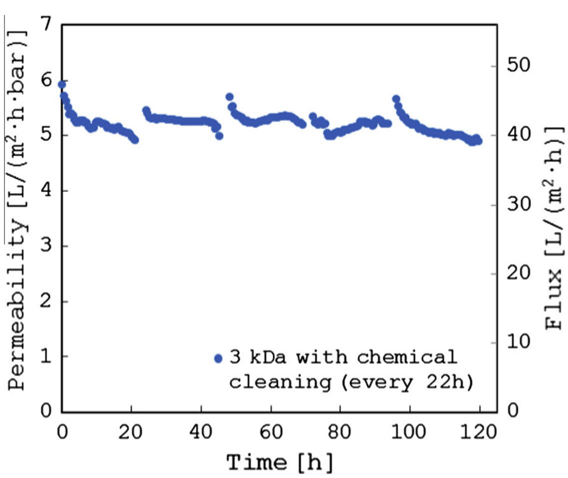

Fig. 7. Temperature corrected permeability and flux over time of ceramic tight UF filtration (3 kDa) membrane during the sewage filtration at constant pressure of 8 bar, cleaned with chemical cleaning per every 22 h.

Fig. 7 shows a reduction of 16% in permeability of the 3 kDa ceramic tight UF membrane within 24 h (from 5.8 to 4.8 L/(m^2 h bar)) and a recovery of 93 ± 3% due to chemical cleaning. The relative production downtime during this chemical cleaning experiment was 2.6 min/h. This value can be compared with 3 and 0.2 min/h of the hydraulic backwash and the forward flush experiments respectively (Table 3).

3.2. Rejection of organic matter and ions

The cake layer formed on the ceramic tight UF membrane was examined after 22 h of filtration. It was clearly visible that the cake layer consisted of thin compressed grains (Fig. 8). This morphology was probably caused by the high operational pressure of 8 bar. Table 4 shows the elemental composition of the cake layer; the cake layer consisted for the major part (98% of the total mass weight) of organic material (elements C, O, and P) and for a minor part of 2% of inorganic elements (Na, Al, Si, S, and Cl). This indicates that no scaling by salts occurred.

Furthermore, the rejection of organic matter and ions by the ceramic 450 Da ceramic NF (repeated 3 times) and 3 kDa ceramic tight UF membranes was measured. Table 5 presents that both membranes rejected about 81% of total COD in all cases. However, the tighter 450 Da membrane seems to withhold a higher percentage of dissolved COD due to the greater steric exclusion of organic molecules in the 450 Dalton pores.

The ortho-phosphate rejection percentages varied for three experiments using the 450 Da ceramic NF membranes, even though the experimental conditions were similar. During the first 450 Da ceramic NF experiment, a 97% rejection of phosphate was measured, while only 14 and 9% rejection was measured during the second and third experiments, respectively. During the first 450 Da ceramic NF experiment a new membrane was used, while the other experiments used older membranes. This indicates that the high rejection of phosphate decreased with the fouling of the membrane.

The rejection of ions (i.e. calcium, magnesium, and ammonium) was low, below 10% (Table 5). These ions arrive at the same concentration in the permeate and should therefore be removed during RO treatment. The rejection percentages varied between the

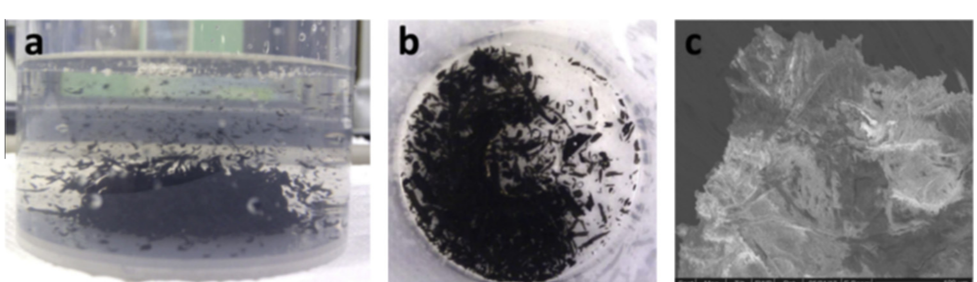

Fig. 8. Morphology of the cake layer formed on the ceramic tight UF membrane (3 kDa) after 22 h of filtration of sewage; visual observations (a and b) and microscopy image (c).

Table 4
Elemental composition of the cake layer on the 3 kDa ceramic membrane after 22 h of filtration of municipal sewage.

Element	Weight percentage, %
C	54.5 ± 0.7
O	43.3 ± 1.3
Na	0.4 ± 0.1
Al	0.6 ± 0.02
Si	0.4 ± 0.1
P	0.3 ± 0.2
S	0.3 ± 0.4
Cl	0.2 ± 0.3

Table 5
Rejection of compounds in sewage by the 3 kDa ceramic tight UF membrane and repeated 3 times with the 450 Da ceramic membrane (average ± standard deviation from at least duplicate measurements).

Parameter	Rejection, %			
	3 kDa	450 Da (1)	450 Da (2)	450 Da (3)
Total COD	81 ± 4	81 ± 2	80 ± 3	81 ± 3
Dissolved COD	42 ± 17	49 ± 5	55 ± 15	45 ± 8
DOC	–	–	49 ± 11	–
Conductivity	7.6 ± 1	0.9 ± 4	–	4.6 ± 5
NH_4^+	11 ± 3	8.2 ± 0.3	6 ± 2	6.5 ± 5
Total P	–	–	37 ± 1	40 ± 4
PO_4^{3-}-P	17 ± 7	97 ± 4	14 ± 5	9.3 ± 7
SO_4^{2-}	22 ± 9	28 ± 33	–	–
Mg^{2+}	0.4 ± 2	2.0 ± 0.1	–	–
Ca^{2+}	11 ± 8	1.1 ± 1	–	–

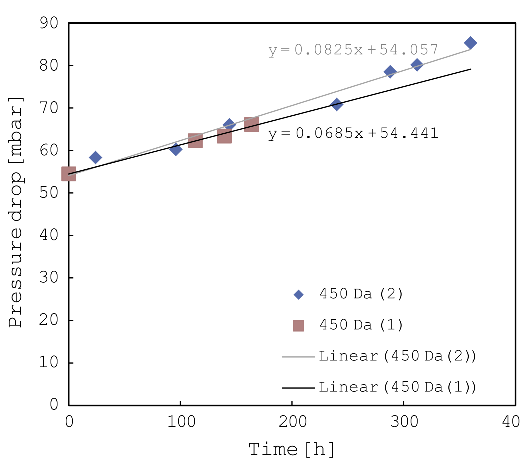

Fig. 9. Pressure drop in the membrane fouling simulator (MFS) with RO membrane over time with effluent water from 450 Da ceramic NF used as feed water. Experiment was repeated two times for 7 days (450 Da (1)) and 15 days (450 Da (2)).

different datasets, which can be explained by the variation in concentrations in the feed water. The water quality of sewage varies during the day.

3.3. Fouling potential in the RO membrane

Ceramic NF is proposed as pre-treatment for RO, according to the ceramic filtration concept (Fig. 1). Permeate of the ceramic NF membrane was used for investigating the fouling potential in RO membranes using MFS tests. The results of the MFS tests showed a gradual, small pressure increase over time for both experiments (Fig. 9), with a slope of 0.07–0.08 mbar/h. This pressure drop slope was comparable with MFS results fed with Dutch drinking water, 0.09 mbar/h [29], under similar operational conditions. Drinking water has a low nutrient concentration and therefore the fouling potential of RO fed with drinking water is low [17,29].

Biological growth causes resistance in the RO membrane feed spacer leading, to a higher pressure drop over the membrane [8,29]. The low increase in pressure drop indicates that the biological growth was minimal in the RO membrane during the experimental period. The absence of severe biofouling was probably because of the low oxygen concentrations (<0.01 mg/L) in the ceramic NF permeate. The MFS experiments showed that the biofouling potential of ceramic NF permeate in an RO was low and run times of at least 14 days without chemical cleaning could be expected.

4. Conclusions

In this paper, the use of ceramic tight UF and ceramic NF filtration for direct treatment of domestic sewage has been studied. From the results of this study, the following conclusions were drawn.

- Ceramic NF (450 Da) membranes were suitable for treating raw municipal sewage. Regular chemical cleaning (with NaClO (0.1%) and HCl (0.1 mol L^{-1})) between every 1 and 4 days could maintain the high performance of the ceramic membranes with at least 93% permeability recovery of the membrane. Hydraulic backwashing and forward flush removed part of the cake layer, but this did not restore the permeability of the membrane.
- The organic matter rejection for both tested ceramic tight UF and ceramic NF membranes was high (81% COD rejection). This led to a high organic load in the concentrate stream, which is potentially beneficial for further anaerobic digestion.
- The permeate water of the ceramic NF had a low fouling potential for RO treatment. The biofouling potential of the permeate water, measured using the Membrane Fouling Simulator (MFS), was comparable with Dutch drinking water.

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References

[1] W.S. Ang, S. Lee, M. Elimelech, Chemical and physical aspects of cleaning of organic-fouled reverse osmosis membranes, J. Memb. Sci. 272 (2006) 198–210.
[2] C.R. Bartels, M. Wilf, K. Andes, J. Iong, Design considerations for wastewater treatment by reverse osmosis, Water Sci. Technol. 51 (2005) 473–482.
[3] D. Bixio, H. Cikurel, M. Muston, V. Miska, D. Joksimovic, A. Ravazzini, A. Aharoni, D. Savic, C. Thoeye, H. Building, N.P. Road, Municipal wastewater reclamation : where do we stand ? An overview of treatment technology and management practice, Water Sci. Technol. Water Supply 5 (2005) 77–86.
[4] R. Butler, T. MacCormick, Opportunities for decentralized treatment, sewer mining and effluent re-use, Desalination 106 (1996) 273–283.
[5] J.P. Chen, S. Kim, Y. Ting, Optimization of membrane physical and chemical cleaning by a statistically designed approach, J. Memb. Sci. 219 (2003) 27–45.
[6] H. Choi, K. Zhang, D. Dionysiou, D. Oerther, G. Sorial, Effect of permeate flux and tangential flow on membrane fouling for wastewater treatment, Sep. Purif. Technol. 45 (2005) 68–78.

[7] J. Duiven, B. Rietman, W. Van De Ven, Application of the membrane fouling simulator to determine biofouling potential of antiscalants in membrane filtration, J. Water Supply Res. Technol. - AQUA 59 (2010) 111–119.
[8] H.-C. Flemming, Reverse osmosis membrane biofouling, Exp. Therm. Fluid Sci. 14 (1997) 382–391.
[9] M. Herzberg, M. Elimelech, Biofouling of reverse osmosis membranes: role of biofilm-enhanced osmotic pressure, J. Memb. Sci. 295 (2007) 11–20.
[10] W.A.M. Hijnen, D. Biraud, E.R. Cornelissen, D. Van Der Kooij, Threshold concentration of easily assimilable organic carbon in feedwater for biofouling of spiral-wound membranes, Environ. Sci. Technol. 43 (2009) 4890–4895.
[11] R.W. Holloway, A.E. Childress, K.E. Dennett, T.Y. Cath, Forward osmosis for concentration of anaerobic digester centrate, Water Res. 41 (2007) 4005–4014.
[12] S. Huang, N. Voutchkov, S.C. Jiang, Investigation of environmental influences on membrane biofouling in a Southern California desalination pilot plant, Desalination 319 (2013) 1–9.
[13] Q. Li, M. Elimelech, Organic fouling and chemical cleaning of nanofiltration membranes: measurements and mechanisms, Environ. Sci. Technol. 38 (2004) 4683–4693.
[14] K. Lutchmiah, E.R. Cornelissen, D.J.H. Harmsen, J.W. Post, K. Lampi, H. Ramaekers, L.C. Rietveld, K. Roest, Water recovery from sewage using forward osmosis, Water Sci. Technol. 64 (2011) 1443–1449.
[15] K. Lutchmiah, A.R.D. Verliefde, K. Roest, L.C. Rietveld, E.R. Cornelissen, Forward osmosis for application in wastewater treatment: a review, Water Res. 58 (2014) 179–197.
[16] P.L. McCarty, J. Bae, J. Kim, Domestic wastewater treatment as a net energy producer-can this be achieved?, Environ Sci. Technol. 45 (2011) 7100–7106.
[17] S.R. Pandey, V. Jegatheesan, K. Baskaran, L. Shu, Fouling in reverse osmosis (RO) membrane in water recovery from secondary effluent: a review, Rev. Environ. Sci. Bio/Technol. 11 (2012) 125–145.
[18] J.J. Qin, S. Chen, M.H. Oo, K.A. Kekre, E.R. Cornelissen, C.J. Ruiken, Experimental studies and modeling on concentration polarization in forward osmosis, Water Sci. Technol. 61 (2010) 2897–2904.
[19] M. Raffin, E. Germain, S. Judd, Wastewater polishing using membrane technology: a review of existing installations, Environ. Technol. 34 (2013) 617–627.
[20] A.M. Ravazzini, A.F. van Nieuwenhuijzen, J.H.M.J. van der Graaf, Direct ultrafiltration of municipal wastewater: comparison between filtration of raw sewage and primary clarifier effluent, Desalination 178 (2005) 51–62.
[21] L.C. Rietveld, D. Norton-Brandão, R. Shang, J. van Agtmaal, J.B. van Lier, Possibilities for reuse of treated domestic wastewater in the Netherlands, Water Sci. Technol. 64 (2011) 1540–1546.
[22] C.J. Ruiken, G. Breuer, E. Klaversma, T. Santiago, M.C.M. van Loosdrecht, Sieving wastewater–cellulose recovery, economic and energy evaluation, Water Res. 47 (2013) 43–48.
[23] W. Rulkens, Increasing the environmental sustainability of sewage treatment by mitigating pollutant, Pathways 23 (2006) 650–665.
[24] S. Sayed, S. Tarek, I. Dijkstra, C. Moerman, Optimum operation conditions of direct capillary nanofiltration for wastewater treatment, Desalination 214 (2007) 215–226.
[25] R. Shang, W.B.P. van den Broek, S.G.J. Heijman, S. van Agtmaal, L.C. Rietveld, Wastewater reuse through RO: a case study of four RO plants producing industrial water, Desalin. Water Treat. 34 (2011) 408–415.
[26] R. Shang, A.R.D. Verliefde, J. Hu, Z. Zeng, J. Lu, A.J.B. Kemperman, H. Deng, K. Nijmeijer, S.G.J. Heijman, L.C. Rietveld, Tight ceramic UF membrane as RO pre-treatment: the role of electrostatic interactions on phosphate rejection, Water Res. 48 (2014) 498–507.
[27] W. Verstraete, P. Van de Caveye, V. Diamantis, Maximum use of resources present in domestic "used water", Bioresour. Technol. 100 (2009) 5537–5545.
[28] J.S. Vrouwenvelder, Biofouling of Spiral Wound Membrane Systems, 2009, http://repository.tudelft.nl/view/ir/uuid%3Aae10097a-68e8-41a2-a51e-593c985c818d/.
[29] J. Vrouwenvelder, J. Vanpaassen, L. Wessels, a. Vandam, S. Bakker, The membrane fouling simulator: a practical tool for fouling prediction and control, J. Memb. Sci. 281 (2006) 316–324.
[30] J.S. Vrouwenvelder, S.M. Bakker, L.P. Wessels, J.a.M. van Paassen, The membrane fouling simulator as a new tool for biofouling control of spiral-wound membranes, Desalination 204 (2007) 170–174.
[31] J.S. Vrouwenvelder, F. Beyer, K. Dahmani, N. Hasan, G. Galjaard, J.C. Kruithof, M.C.M. Van Loosdrecht, Phosphate limitation to control biofouling, Water Res. 44 (2010) 3454–3466.
[32] R. Weber, H. Chmiel, V. Mavrov, Characteristics and application of new ceramic nanofiltration membranes, Desalination 157 (2003) 113–125.
[33] T. Wintgens, D. Bixio, C. Thoeye, P. Jeffrey, R. Hochstrat, T. Melin, Integrated Concepts for Reuse of Upgraded Wastewater – AQUAREC, 2002.
[34] T. Wintgens, T. Melin, A. Schäfer, S. Khan, M. Muston, D. Bixio, C. Thoeye, The role of membrane processes in municipal wastewater reclamation and reuse, Desalination 178 (2005) 1–11.
[35] M. Xie, L.D. Nghiem, W.E. Price, M. Elimelech, A forward osmosis-membrane distillation hybrid process for direct sewer mining: system performance and limitations, Environ. Sci. Technol. 47 (2013) 13486–13493.