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Usage of permeate water for treated domestic wastewater by direct capillary nanofiltration membrane in agriculture reuse

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ABSTRACT

Direct Capillary Nanofiltration (Direct CNF) is a new technique used for surface water and wastewater treatment in one step without pre-treatment. The CNF membrane module combines the favourable cleaning properties of capillary ultrafiltration membranes with the favourable separation properties of nanofiltration membranes in terms of removal of DOC, colour, bacteria, viruses and pesticides. In this study, Direct CNF has been applied to wastewater treatment under continuous and stable process conditions. The optimum conditions for Direct CNF when applied to domestic wastewater were 15 min filtration time, $20\,L/m^2\,h$ flux rate with undiluted domestic wastewater under stable conditions for 19 h and 20 min continuously. Direct CNF showed high removal for bacteria, heavy metals, COD, BOD_5, DOC and medium removal for manganese, calcium and magnesium while lower removal was detected for mono valent ions such as sodium and chloride. The permeate of CNF was high quality water that can be used for agriculture purposes but presence of a few counts of bacteria and high ammonia and total nitrogen concentrations shows that the permeate water needs further treatment. Accordingly, the permeate water was treated with granular activated carbon followed by low chlorine doses for disinfection to produce water that can be used for agriculture purposes of any type of crop.

Keywords: Capillary nanofiltration; Domestic wastewater; Agriculture reuse; Post treatment

1. Introduction

Membrane technology is widely accepted as a means of producing various qualities of water from surface water, well water, brackish water and seawater. Membrane technology is also used in potable water as well as industrial processes and in industrial wastewater treatment, and lately membrane technology has moved into the area of treating secondary and tertiary municipal wastewater and oil field-produced water [1].

Capillary nanofiltration (CNF) membrane is a type of pressure-driven membrane with properties in between reverse osmosis (RO) and ultrafiltration (UF) membranes. CNF offers several advantages such as low operation pressure, high flux, high retention of multivalent anion salts and an organic molecular weight above 300, relatively low investment and low operation and maintenance costs. Because of these

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advantages, the applications of NF worldwide have increased [2]. Also, it combines the favourable properties of the capillary UF membranes in terms of ease of cleaning with the favourable properties of the NF membrane in terms of the removal of bacteria, viruses, pesticides and heavy metals. The capillary NF membrane is presently available in the well-known 8 in. modules which are also being used successfully for capillary and the system setup.

The capillary NF module is operated in the same way as semi dead-end UF. During the production run the concentrate valve is closed and all the feed supplied to the system is withdrawn as permeate as shown in Fig. 1. In order to stabilise the flux and rejections at an acceptable level a small cross-flow velocity is applied over the module. If the rejection drops too much, the concentrate valve is opened and the system is flushed by means of air-enhanced forward flushing, a so-called AirFlush®. During this flushing procedure a backflush can also be carried out. Subsequently, the concentrate valve is closed and the production run starts again.

In a previous study [3] a new purification concept was introduced: direct CNF and its potential for direct treatment of surface water and effluent of a waste water treatment plant was shown. More recently [4], this CNF membrane was compared with commercial flat sheet NF membranes for the treatment of surface water showing excellent flux behaviour and comparable rejection characteristics.

CNF concept is developed further for surface water treatment by Dijkstra et al. [5] focussing on back flushing and chemical cleaning. Next, the removal rates for various components are studied by modelling and sampling the feed and permeate streams. Attention is paid especially to the membrane performance in comparison with the model. The final objective is to determine the operating conditions for a full scale plant. The last part is dedicated to the long term operation experiences of a fully-automated pilot installation with two 8 in. Capillairy NF modules.

CNF was applied for the first time to direct treatment of domestic wastewater [6] to obtain optimum conditions. The optimization of the chemical cleaning regime was carried out and the recovery was increased from 35% to 65%.

NF is an efficient and ecologically suited technology for decontamination and recycling of wastewater generated in many industries [7], e.g. treatment of fish meal wastewaters [8]. NF reduces the organic load in the wastewaters and promotes the partial desalination, making water re-usable. A major problem of wastewater treatment is the water recovery rate, which should be as close as possible to 100% [9].

In this paper we represent the efficiency of Direct CNF in the removal of organic, inorganic and biological parameters from domestic wastewater to produce high quality permeate which is compared with the guidelines for irrigation purposes in Egypt. Secondary treatment with granular activated carbon (GAC) followed by 2 mg/L chlorine dose was carried out to improve the quality of the permeate water.

2. Experimental

The Capillary Nanofiltration Membrane used was 4 in. NF50 M10 which was provided by X-Flow company (The Netherlands) which is composed of polyamide polyethersulphone coated with polyamide. The domestic wastewater was passed first through a 500 μm screen before going through the membrane. The performance data of the used membrane are shown in Table 1.

The optimum conditions applied in this experiment were 20 L/m^{2}h flux rate and 15 min filtration time with 100% domestic wastewater. The system was hydraulically cleaned for 2 min between each filtration runs and chemically cleaned using hydrochloric acid,

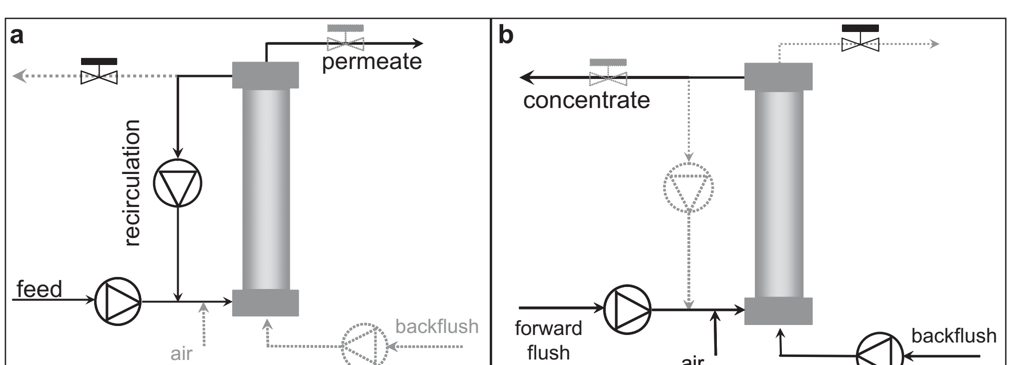

Fig. 1. Operation of direct capillary nanofiltration: (a) production; (b) cleaning.

Table 1
Performance data of NF50 M10 CNF membrane

Parameter	Unit	NF50 M10	Comment
Clean water flux	$L/m^2 h$. 100 kPa	16 ± 2	RO-water at 25°C
Initial water flux	$L/m^2 h$. 100 kPa	13 ± 2	0.35 wt.% NaCl at 25°C
Rejection of NaCl	%	35 ± 5	At 600 kPa and Re = 2500
Initial flux	$L/m^2 h$. 100 kPa	12 ± 2	0.5 wt.% $MgSO_4$ at 25°C
Rejection of $MgSO_4$	%	94 ± 2	At 600 kPa and Re = 2500
Maximum (TMP) transmembrane pressure	kPa	700	–
pH feed	–	4–10	at 25°C
Chlorine exposure	ppm h	Do not expose	–
Temperature	°C	1–40	–

hydrogen peroxide and sodium hydroxide for 2.5 h every 19 h with a recovery percent of 78%.

The pilot installation receives the domestic wastewater after a 500 μm strainer to avoid particles damaging the membrane surface and/or clogging the membrane fibres themselves.

The chemical analyses and bacteriological analyses were carried out according to the Standard Methods for the Examination of Water and Wastewater 22th Edition (2005).

The samples were collected every 10 min during the filtration time using an autosampler and were mixed together to make a composite sample which was analysed after that.

During the experiment, six samples were collected and analyzed. The removal percent of various compounds was calculated. The permeate was passed through a 16 mg/L granular activated carbon (GAC) column followed by a chlorine dose of 2 mg/L. The effluent in each step was compared to the standards of the Food and Agriculture Organization (FAO) 1985.

3. Results

3.1. Removal of wastewater by CNF

The removal percentages of the inorganic compounds of the wastewater using direct CNF are presented in Table 2. Table 3 shows the removal percentages of the organic and bacteriological parameters of the wastewater (see Table 4).

Retention of organic compounds was affected by some parameters such as molecular weight cut-off of the membrane and hydrophobicity. Since hydrophobic molecules generally have low retention, while hydrophilic molecules and low molecular weight compounds showed high retention [10] retention by different nanofiltration membranes is assessed taking molecular weight and hydrophobicity into account, which was studied by Moons and Van der Bruggen [11]. On the other hand, pH has a significant effect on retention as well as membrane structure and its permeability as mentioned by Mika et al. [12]. CNF reduced the organic loading rate in the wastewaters [6] and promoted their partial desalination, making water re-usable. A major problem of the wastewater treatment is the water recovery rate, which should be close to 80% [13]. To achieve this target, many researchers investigated an integrated membrane system, as example Rautenbach and Linn [14] and Rautenbach et al. [15] used a new concept of integrated membranes consisting of RO/NF/high-pressure RO. The integration can achieve water recovery rates of more than 95% in the case of dumpsite leachate, which promises an almost zero discharge process.

Separation by CNF membrane occurs primarily due to size exclusion and charge effect on electrostatic interactions [16]. Namely, the rejection of uncharged molecules is dominated by size exclusion, while that of ionic species is influenced by size exclusion and electrostatic interaction. And, electrostatic characteristics of CNF membranes have been known to play an important role in rejection anions, i.e., the negative zeta potential on the membrane surface varies with different pH and concentration of an electrolyte solution [17,18].

Direct CNF showed high removal for turbidity, suspended solids and divalent molecules such as iron, manganese, lead and copper compared to monovalent ions such as chlorine, sodium and potassium, which may be attributed to the ion size which plays a role in membranes with small pores, leading to large selectivity or due to applied pressure may influence the separation [19] Also, CNF showed high removal for organic contents and TOC had the highest removal (95.9%) meanwhile, COD and BOD showed nearly equal removal. The results showed a slight increase in the pH value in permeate water (from 7.21 to 7.32) which may be attributed to the decrease in the bicarbonate concentration from 524 mg/L to 400 mg/L

Table 2
Removal percents for some physico-chemical parameters after direct CNF

Parameters	Unit	Feed	Permeate	% of removal	Usual range in irrigation water[a]
pH	–	7.21	7.32	–	6.5–8.5
Turbidity	FTE	56	1.9	96.6	
Electrical conductivity	μS/m	1433	1143	20.2	3000
Total dissolved solids	mg/L	852	682	20.0	2000
Total suspended solids	mg/L	733	8	98.9	
Total hardness (as $CaCO_3$)	mg/L	182	92	49.5	
Calcium hardness (as $CaCo_3$)	mg/L	130	64	50.8	
Magnesium hardness (as $CaCO_3$)	mg/L	52	23	55.8	
Alkalinity	mg/L	524	400	23.7	
Chloride	mg/L	122	106	13.1	1100
Sulfate	mg/L	9.6	7.8	18.8	1000
Nitrate (NO_3–N)	mg/L	1.38	0.102	92.6	
Nitrite (NO_2–N)	mg/L	0.20	0.053	73.5	
Ammonia	mg/L	27	22	18.51	
Total Nitrogen	mg/L	30	23	23.33	30
Carbonate	mg/L	0.00	0.00	–	
Hydroxide	mg/L	0.00	0.00	–	
Bicarbonate	mg/L	524	400	23.7	600
Phosphate (PO_4–P)	mg/L	8.22	1.49	81.9	
Iron	mg/L	1.27	0.02	98.4	5
Manganese	mg/L	0.37	0.07	81.1	
Calcium	mg/L	51	25	51.0	400
Magnesium	mg/L	12	5.5	54.2	60
Copper	mg/L	0.040	0.004	90.0	0.1
Lead	mg/L	0.0034	0.0009	73.5	2
Sodium	mg/L	137	109	20.4	900
Potassium	mg/L	35	28	20.0	
Sodium absorption ratio	–	–	7.25	–	15

[a]Food and Agriculture Organization (FAO), 1985.

Table 3
Removal percents for organic matter after direct CNF

Parameters	Unit	Influent	Permeate	% of removal	Usual range in irrigation water[a]
Total organic carbon	mg/L	122	5	95.9	
Chemical oxygen demand (COD)	mgO_2/L	743	93	87.5	
Biological oxygen demand (BOD_5)	mgO_2/L	382	53	86.1	

[a]Food and Agriculture Organization (FAO), 1985.

which leads to a decrease in the carbonic acid produced in water and increase in pH according to the following equation.

$$HCO_3^- + H_2O = H_2CO_3 + OH^-$$

3.2. Comparison of permeate water with FAO standards

On comparison of the water quality of the effluent with that of the usual range in irrigation water of FAO,all the studied parameters are within the usual range in the irrigation water of FAO.

Table 4
Removal percents for bacteriological parameters after direct CNF

Parameters	Unit	Influent	Permeate	% of removal	Usual range in irrigation water[a]
Total bacterial count at 22°C	mL	5.7×10^6	1×10^3	99.98	
Total coliform	100 mL	377	1	99.7	10^3
E. Coli	100 mL	348	0	100	

[a]Food and Agriculture Organization (FAO), 1985.

Table 5
Guidelines for interpretation of water quality for irrigation

Potential irrigation problem	Units	Degree of restriction on use		
		None	Slight to moderate	Severe
Salinity				
Ec_w[a]	dS/m	<0.7	0.7–3.0	>3.0
or				
TDS	mg/L	<450	450–2000	>2000
Infiltration				
SAR[b] = 0–3 and EC_w		>0.7	0.7–0.2	<0.2
3–6		>1.2	1.2–0.3	<0.3
6–12		>1.9	1.9–0.5	<0.5
12–20		>2.9	2.9–1.3	<1.3
20–40		>5.0	5.0–2.9	<2.9
Specific ion toxicity				
Sodium (Na)				
Surface irrigation	SAR	<3	3–9	>9
Sprinkler irrigation	me/I	<3	>3	
Chloride (Cl)				
Surface irrigation	me/I	<4	4–10	>10
Sprinkler irrigation	m^3/l	<3	>3	
Boron (B)	mg/L	<0.7	0.7–3.0	>3.0
Miscellaneous effects				
Nitrogen (NO_3–N)[c]	mg/L	<5	5–30	>30
Bicarbonate (HCO_3)	me/I	<1.5	1.5–8.5	>8.5
pH	Normal range 6.5–8			

[a]EC_w means electrical conductivity in deciSiemens per meter at 25°C.
[b]SAR means sodium adsorption ratio.
[c]NO_3–N means nitrate nitrogen reported in terms of elemental nitrogen.

Many schemes of classification for irrigation water have been proposed, FAO (1985) [20] classified irrigation water into three groups based on salinity, sodicity, toxicity and miscellaneous hazards, as shown in (Table 5). These general water quality classification guidelines help to identify potential crop production problems associated with the use of conventional water sources.

Correlation between SAR and EC indicated that soil permeability (including infiltration rate and surface crusting) hazards caused by sodium in irrigation water cannot be predicted independently of the dissolved salt content of the irrigation water or that of the surface layer of the soil as shown in Fig. 2 which is given by Rhoades [21].

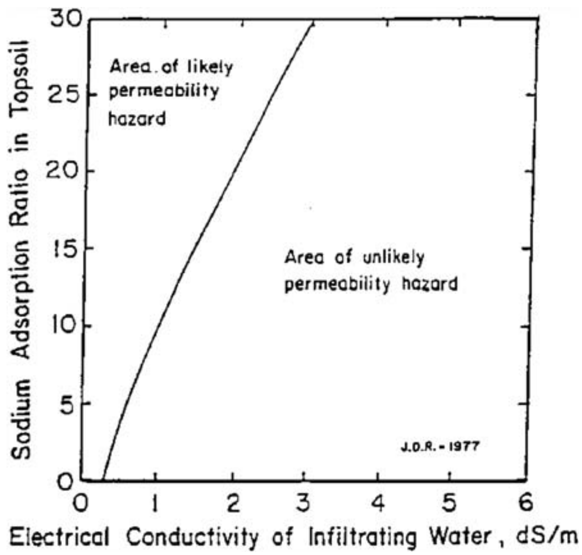

Fig. 2. Threshold values of sodium adsorption ratio and total salt concentration on soil permeability hazard (Rhoades 1982).

3.3. Comparison of effluent water quality with Egyptian Standards

Quality of permeate water compared with ministerial decree number 44/2000 for the conditions and precautions required in the reused wastewater in irrigation is showed in Table 6. The results showed that the quality of permeate water is even better than that obtained with advanced treated water in all the studied parameters except in chemical oxygen demand and biological oxygen demand content. Accordingly, permeate water can be used for irrigation of woody trees in desert soil which are 5 km far from localities and making a fence around the farm and prevent entry of cattle and tresspassers people according to Egyptian standard Ministerial Decree No.44/2000. Accordingly, permeate water needed to be secondary treated to improve the water quality.

Permeate water was passed through a 16 mg/L column of granular activated carbon with a contact time of 8 min followed by 2 mg/L chlorine with a contact time of 15 min in order to decrease the COD, BOD, ammonia and total nitrogen concentrations and the quality of the effluent is showed in Table 7. Fig. 2 shows the removal of some studied physico-chemical parameters in different treatment steps.

The analysis of the final effluent showed the total removal of bacteria, COD, BOD_5 (Table 6). It is shown in Fig. 3 that the concentration of nitrate is increased by 1.58 times and this was attributed to the oxidation of ammonia into nitrate. The final concentration of nitrite and nitrate was very low compared with the original ammonia concentration and this may be attributed to incomplete oxidation of ammonia with chlorine to form, the weak oxidant and disinfectant chloramines that will increase the oxidation rate and increase disinfection.

The results showed that the final effluent can be used for irrigation of edible raw plants, corticosteroid plants, all crops, orchids, forages (feed) and green pastures for any soil type without any caution. Further analysis of the final effluent indicated that water can be drinkable according to the Egyptian standard limits (Ministerial Decree 458/2007).

Finally, we can conclude that domestic wastewater can be treated to produce water that can be used for irrigation of any type of crops without any caution in any soil using direct CNF by treatment of the permeate with 16 mg/L GAC and 2 mg/L chlorine. The schematic diagram of the system will be as shown in Fig. 4.

Table 6
Comparison of permeate water quality with Egyptian Standards (ministerial decree number 44/2000)

Parameters	Unit	Permeate	Primary treated water	Secondary treated water	Tertiary (advanced) treated water
Chemical oxygen demand	mgO_2/L	93	600	80	40
Biological oxygen demand	mgO_2/L	53	300	40	20
Total suspended solids	mg/L	8	350	40	20
Total dissolved solids	mg/L	682	2500	2000	2000
SAR	%	7.25	25	20	20
Chloride	mg/L	106	350	300	300
Iron	mg/L	0.02	–	5	5
Manganese	mg/L	0.07	0.20	0.20	0.20
Copper	mg/L	0.004	–	0.20	0.20
Lead	mg/L	0.0009	10	5	5
Total coliform	Unit/100 mL	1	–	1000	100

Table 7
Removal percents for some studied parameters after treatment of permeate with GAC and chlorine

Parameters	Unit	Feed water	Permeate	Permeate + GAC + Cl_2	% of removal
Chemical oxygen demand	mgO_2/L	743	93	0	100
Biological oxygen demand	mgO_2/L	382	53	0	100
Total suspended solids	mg/L	733	8	10	98.6
Total dissolved solids	mg/L	852	682	701	17.7
SAR	%	–	7.25	7.33	–
Chloride	mg/L	122	106	116	4.9
Iron	mg/L	1.27	0.02	0.004	99.7
Manganese	mg/L	0.37	0.07	0.009	97.6
Copper	mg/L	0.04	0.004	0.003	92.5
Lead	mg/L	0.0034	0.0009	0.0008	76.5
Total coliform	Unit/100 mL	377	1	0	100

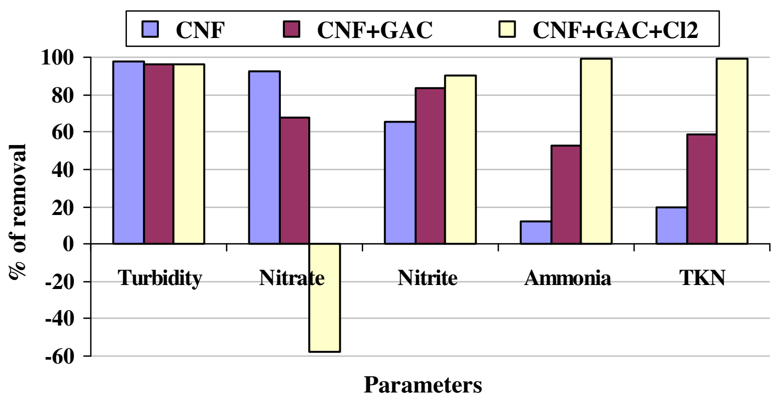

Fig. 3. Removal of physico-chemical parameters during treatment steps.

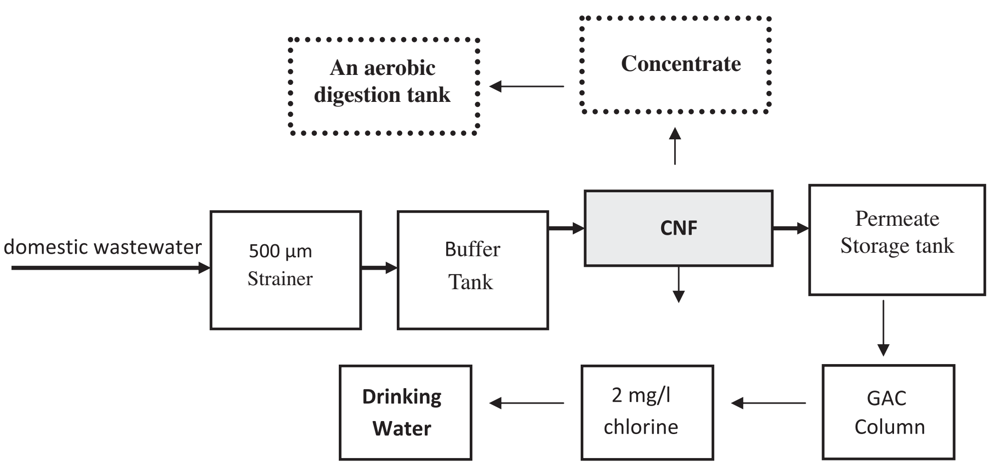

Fig. 4. Schematic diagram for CNF pilot installation.

4. Conclusion

Direct CNF was successfully applied to domestic wastewater processes to produce high quality water in a continuous process. The permeate can be used for irrigation of any type of crops without any cautions in any soil and also can be drinkable by further treatment using 16 mg/L GAC column with 8 min of contact time followed by oxidation and disinfection using 2 mg/L chlorine with 15 min reaction time.

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