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Reverse osmosis membranes for treatment of produced water: a process analysis

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ABSTRACT

The purpose of this paper was to develop and present a process suitable for the purification of the so-called produced waters, a by-product of crude oil extraction, by devising a treatment train aimed at industrial and agricultural water reuse. If compared to municipal wastewaters, produced waters have a very high salinity that requires specific attention for designing and managing the specific treatment device. Membranes, commonly used in the production of desalted water, appear to be a suitable technique to deal with these issues. In this paper, we propose a comprehensive process scheme for produced water treatment train: A Vibratory Shear Enhanced Processing (VSEP) membrane system is in charge of the secondary treatment, whereas a reverse osmosis (RO) unit realizes the tertiary treatment. Material and energy balances are carried out on the whole process, while the RO process is simulated by the IMSDesign Software by Hydranautics. We analyzed three different scenarios, at increasing produced waters salinity, getting a stream outlet as purified water with such low pollutants concentration and salinity to be reusable for different purposes. The RO process is carried out with a single-step or a double-step filtration; a cost analysis, performed on the different case studies, allowed computing the final specific costs per cubic meter of treated water, showing that a double filtration step allows a lower salinity water, albeit raising the costs up to about 5 €/m^3, a high price justified only if a ultrapure water should be required for specific applications.

Keywords: Produced water; Hydrocarbon; VSEP membrane; Water reuse; Reverse osmosis; Process analysis

1. Introduction

A large amount of wastewaters, known as "produced waters", comes out from oil plants and platforms, considered a by-product of crude oil extraction; currently, they are treated and disposed in deep wells on the onshore platforms or directly discharged into the sea. Specifically, 65% of this water is re-injected to the well for pressure maintenance, 30% of total is injected to deep well for final disposal in the case of proper aquifer conditions, and the rest of the water is discharged to surface water [1].

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Extraction technology and reservoir characteristics affect the amount of produced waters [2], and up to tenfold the amount of produced oil [3]. Produced waters account for around 70% of total oil production wastewaters volume.

Moreover, leaks and accidental spills occur regularly during all the activities connected to petroleum industry, for example, exploration, production, refining, transport, and storage of petroleum and petroleum products [4]. In 2003, the amount of natural crude oil seepage was estimated to be about 6×10^5 metric tons [5]. Thus, hydrocarbons released into the environment, whether accidentally or due to anthropic activities, are the main source of water pollution.

Nowadays, as water demand increases day by day, it is essential to recover and reuse water [6]. Thus, many countries with oil fields are generally water-stressed countries; therefore, they are increasingly focusing their efforts to find efficient and cost-effective treatment methods to remove pollutants as a way to supplement their limited freshwater resources [7]. In addition, it is crucial to find new technologies that aim not only to the environmental sustainability, but also comply with the more stringent rules and regulations of the field. The permit limits of oil and gas (O&G) for treated produced water according to the United States Environmental Protection Agency regulatory limits are 29 mg/L for a monthly average and 42 mg/L for a daily maximum [8].

Produced waters are characterized by a high content of salts and oil, which forces to draw a purposed treatment train, different, for example, from those commonly used for municipal wastewater treatment. Typically, produced water contains high concentrations of aromatic hydrocarbons, for example, BTEX (benzene, toluene, ethylbenzene, xylene), NPD (naphthalene, phenanthrene dibenzothiophene), PAH (polycyclic aromatic compounds) [9,10], minerals, radioactive substances, dissolved gases, scale products, waxes, microorganisms, and dissolved oxygen [11]. The salt concentration may range from a few to 300,000 mg/L; the total organic carbon (TOC) concentrations are between 0 and 1,500 mg/L and O&G concentrations between 2 and 565 mg/L [2].

Biological, physical, and chemical methods are available to specifically remove hydrocarbons from produced water. In offshore extraction facilities, due to space constraints, compact physical and chemical treatment technologies mostly apply [7] (photoelectrocatalytic processes, hydrocyclones, coagulation, and flocculation). Most of these techniques are only suitable for pretreatment of wastewater for *in situ* reuse, for example, reinjection to enhance oil recovery yield [2].

On the other hand, membrane technology may be successfully used to remove hydrocarbons from oil-contaminated wastewater, also in the presence of a high salinity. Membrane processes offer several advantages over conventional treatments, such as compact module, lower energy consumption, environmental friendliness, and high-quality product, independently on fluctuations in feed quality. Moreover, membrane equipments have a smaller footprint, energy costs are often lower, and the plant can be highly automated [12]. For these reasons, microfiltration (MF) and ultrafiltration processes have been increasingly used in potable water production and wastewater treatments as an alternative technology to conventional treatments, getting rid of the chemical pre-treatments and sedimentation, aimed at removing suspended solids, microorganisms, and natural organic matter [13,14].

Moreover, processes based on water separation from saline solution by reverse osmosis (RO) membrane processes are widely spread on the industrial scale, applied not only to sea waters (high salinity), but also to brackish and low-salinity waters, which are characterized by different compositions, thus needing specific pre-treatments [6,15].

Because of the presence of dissolved and suspended oil in untreated produced water, the membrane equipment may foul, thus increasing operation costs. On the other hand, the problem of membrane fouling is a key issue, which frequently limits the widespread of such an effective technique, thus being a hot topic for research purposed at overcoming or limiting it [15–17].

On this regard, the vibrating membrane process Vibratory Shear Enhanced Processing (VSEP®) limits membrane fouling, removing the main contaminants from wastewater without the addiction of antiscalant chemical substances; thanks to the design characteristics, the fouling common to all membrane processes is largely reduced [18]. The pressure vessel moves in a vigorous vibratory motion, tangential to membrane surface, thus creating shear waves, preventing membrane fouling [19]. The retentate volume sent to disposal is reduced about to one third of the feed, so corresponding power duties are very low [20].

The aim of this paper was to analyze the possibility of adapting membrane processes, for example, VSPE and RO, using as feed produced water, properly pretreated, to provide water of high quality, reusable as process water and/or in agriculture. We devise a treatment train comprising a two-step membrane filtration stage (VSEP followed by RO). We analyze the mass balances applied to the whole process; according to removal efficiencies from literature, we perform the

RO process simulation by the IMSDesign Software by Hydranautics™ [21]. Results show that in all case studies (at different salinity of the feed stream), the treatment results into stream purification within the limits for reuse; thus, the process is viable to get pure or ultrapure water, for multipurpose reuse (industrial or agricultural).

2. Process simulation

2.1. Synthetic feed water characteristics

The composition of produced water varies considerably depending on the geographic location of the reservoir, the geological structure of the soil, the characteristics of the extracted hydrocarbons, the production process, and the exploitation degree of the well.

Thus, we adopted a model solution to simulate the real produced water properties, reported in Table 1 [7,11].

2.2. Primary treatment

Fig. 1 reports the block scheme of the process. The produced water, leaving the three-phase separator which separates oil, gas, and water in the stream from the cross-wellhead, is sent to the gravity separator (American Petroleum Institute (API) separator), which removes, from water surface, oils, and other light fractions with a lower density than water. These fractions, removed by an oil skimmer, are then sent to the oil recovery stage. The settled particles (i.e., those with a larger density than water) are conveyed on the bottom of the separator and, drawn with a screw pump, transferred to the oil sludge processing. The separation performance depends primarily on the type and condition of the oil to be treated and the size of the unit separation. The size of the separator API is calculated according to the standard of API 421-90, in order to separate oil particles with a diameter larger than 150 microns. With these processes, it is possible to obtain removal percentages of 80% for oil and grease and of 90% for the total suspended solid (TSS).

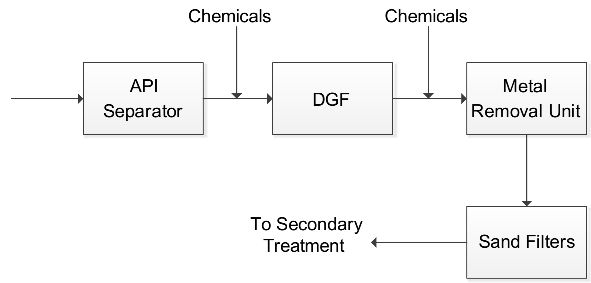


Fig. 1. Primary treatment scheme.

Table 1
Simulated produced water composition

Parameter	Value (mg/L)
Oil and grease	565
Total suspended solid	1,000
Chemical oxygen demand	3,000
Biochemical oxygen demand	1,500
Total organic carbon	1,500
Ammonia NH_4^+	200
BTEX	2.0
Total dissolved solid	37,500

The water separated from oil reaches the free surface on the side opposite to the power supply, and by means of a pump, it is sent to a mixing tank, where appropriate amounts of coagulants and flocculants are added to facilitate the particles coalescence by sedimentation.

Then, water enters into the dissolved gas flotation (DGF) stage, where nitrogen or natural gas (to avoid explosions upon contact with hydrocarbons) is blown to separate oil, suspended solids, and other macromolecules. With this treatment, we get the removal of 90% of the oil and fat TSS and 20–25% of chemical oxygen demand (COD), biochemical oxygen demand (BOD), and TOC.

The produced water coming from the DGF stage is further deoiled and then sent in another mixing tank, where chemical reagents are added (polyelectrolytes, caustic soda, and aluminum chloride); then, the stream is treated in a sedimentation tank to remove metals. Before the secondary treatment stages, produced water passes through sand filters to ensure a further reduction of pollutants. With this system, the abatement percentages reported in the literature are around 5% removal of total dissolved solid (TDS).

2.3. VSEP membranes

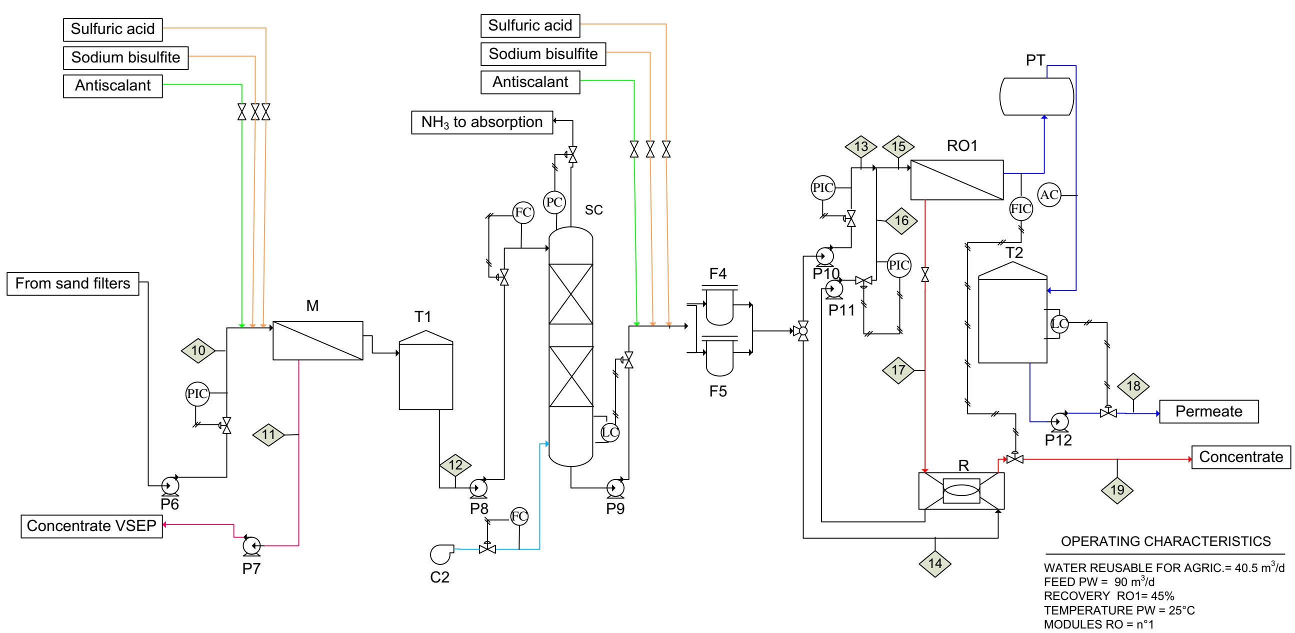
In the VSEP membrane system, patented by New Logic Research, the feed slurry remains nearly stationary, moving in a leisurely, meandering flow between parallel membrane leaf elements. Shear cleaning action is created by vigorously vibrating the leaf elements in a direction tangent to the faces of the membranes: The propagation of shear waves from the sinusoidal membrane surface favors the suspension of particles on it, facilitating the flow, thus reducing the membrane fouling.

The VSEP system is characterized by a "plate and frame" configuration: The porous polymeric membranes (nanofiltration) are arranged in series on flat supports separated by spacer networks which, by providing some resistance to the module, determine a structure with compartments, where powered and permeate are collected in different zones of these rooms. With a recovery factor of 80%, the vibrating membrane system is able to break down more than 90% of TSS, COD, BOD, and TOC, albeit it is ineffective on TDS.

2.4. Process scheme

The simulation runs rely on the process scheme in Fig. 2. Produced water (stream 10), coming from primary treatments, aimed at removing the largest part of contaminants (suspended particles, inorganic compounds, and heavy metals), is sent through a high-pressure pump to the VSEP membranes system. At this stage, the retentate water stream (11), still rich in pollutants, is disposed of, while the permeate stream (12) is sent to a stripping column to remove ammonia. The stripping column is equipped with a cartridge filter (MF up to 5 microns) to remove solids drawn from the stripping column.

To prevent precipitation of low-soluble salts on the membrane surface, antiscalant and chemicals are added to water in the upstream of RO stage. The permeate is then pressurized and sent to the RO stage (stream 15). The process scheme includes also an energy recovery device (R) and a booster pump. Part of the produced water coming from the pre-treatment is pressurized by a high-pressure pump, while the other part is put under pressure by the energy recovery system operating in series with the booster (stream 14). The retentate from the RO stage (stream 19) is sent to disposal, while the permeate (stream 18) is conveyed to a storage tank, ready to use.



	10	11	12	13	14	15	16	17	18	19
FLOWRATE [m^3/d]	100	10	90	39.6	50.4	90	50.4	49.5	40.5	49.5
Oil & Grease [ppm]	0.34	3.31	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
TSS [ppm]	3.02	30.24	0	0	0	0	0	0	0	0
COD [ppm]	453.26	4082.66	50.37	50.37	50.37	50.37	50.37	50.37	50.37	50.37
BOD_5 [ppm]	226.63	2041.33	25.18	25.18	25.18	25.18	25.18	25.18	25.18	25.18
TOC [ppm]	1133.16	10206.7	62.97	62.97	62.97	62.97	62.97	62.97	62.97	62.97
TDS [ppm]	35803.4	38120	35546	35546	35546	35546	35546	64498	160.2	64498

NOTES

PRODUCED WATER
AIR
CHEMICALS
PERMEATE RO1
CONCENTRATE RO1
ANTISCALANT
CONCENTRATE VSEP

LEGEND

P CENTRIFUGAL PUMP
PT PERMEATE TANK
F FILTER
C AIR BLOWERS
T STORAGE TANK
RO REVERSE OSMOSIS
M MEMBRANE VSEP
SC STRIPPER
R ENERGY RECOVERY

Fig. 2. Process scheme—case study A.

3. Process analysis

Mass balance reported in Fig. 2 clearly shows the reliability of the VSEP membrane system for the produced water treatment: The vibrating membranes are able to reduce the TSS content of produced water treated up to 100%, while BOD_5, COD, and TOC contents are reduced up to 90% (according to the removal efficiency of the membrane system from literature [18]). Mass balance also highlights the unreliability of VSEP membrane system for the removal of TDS, which must be got rid of in a dedicated RO section.

In the following, we discuss three case studies for the desalination process of produced waters with three salinity levels (35,546, 71,127, and 106,670 ppm). The sizing of the RO process was performed using the IMSDesign Software by Hydranautics™ [21].

The membranes for RO are the SWC4 Max and SWC5 Max (see Fig. 3) by Hydranautics™ Nitto Group Company, allowing a salt rejection of 99.8%. Membrane geometry (spiral wound) ensures a lower membrane fouling, because of the tangential water flux to membrane surface, which allows high velocities and turbulent flow regimes.

Each membrane has a length of 1 m and an active area of 40.8 m^2; the composite membrane comprises an active layer of polyamide (providing the membrane selectivity) and a porous layer of structural support (polyethersulfone), with a low resistance to water flow. These membranes are chemically and physically stable; they do not hydrolyze water, tolerate pH values in the range of 3–11, and are virtually immune toward biological degradation [22].

3.1. Case study A (TDS 35,546 ppm)

In this case study, we considered one RO stage in order to meet the water quality required by the Legislative Decree No. 152/2006 (Ministry of Environment and Protection of Land and Sea in concert with the Ministries of Economic Development, Infrastructure and Transport and Economy and Finance of Italy), related to "water for irrigation purposes" (no food applications). The mass balance for this case study is reported in Fig. 2, with the flowrates and compositions of all the main streams.

The ionic composition of produced water at $T = 25°C$ and pH 8.1 is shown in Table 2. The salt concentration of the produced water is reduced from 37,500 to 35,803 ppm (see stream 10) by upstream primary treatments, previously described. Table 3 reports the operative membrane parameters.

Table 2
Ionic composition of produced water for case study A (see Fig. 2, stream 12)

Cations	Value (ppm)	Anions	Value (ppm)
Ca^{2+}	410	HCO_3^-	152
Mg^{2+}	1,310	SO_4^{2-}	2,000
Na^+	10,987.9	Cl^-	20,260.2
K^+	390	F^-	1.4
NH_4^+	0	NO_3^-	0.6
Ba^{2+}	0.050	B^{3+}	4
Sr^{2+}	13	SiO_2^{2-}	0.5
Total TDS = 35,546 ppm			

Table 3
Operative RO membrane parameters—case study A

Stage number	1	Feed flowrate (m^3/d)	90
Vessel number	1	Permeate flowrate (m^3/d)	40.5
Elements number	3	Retentate flowrate (m^3/d)	49.5
Total active area (m^2)	122.4	Recovery (%)	45
TDS feed (ppm)	35,546	Feed pressure (bar)	57

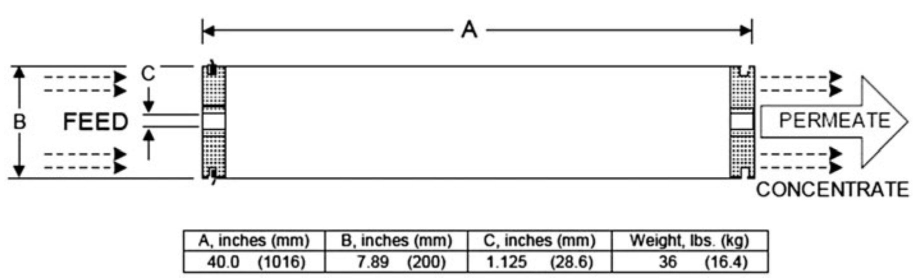
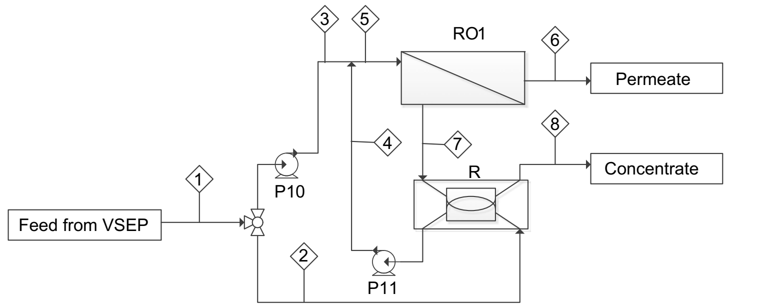


Fig. 3. SWC MAX membrane sketch.



	Units	1	2	3	4	5	6	7	8
Flowrate	m^3/h	3.80	2.80	1.0	2.80	3.80	0.95	2.85	2.85
Pressure	bar	1	1	80.4	80.4	80.4	1	80.1	1
TDS	ppm	71127	71127	71127	71127	71127	449	94686	94686

Fig. 4. Sketch of the RO section for the case study B.

3.2. *Case study B (TDS 71,127 ppm)*

The sketch of the RO section for this case study is reported in Fig. 4. The ionic composition of produced water analyzed in the case is reported in Table 4. Also in this case, it has been considered only one stage of RO. The operative membrane parameters are reported in Table 5.

Table 4
Ionic composition of produced water for case study B

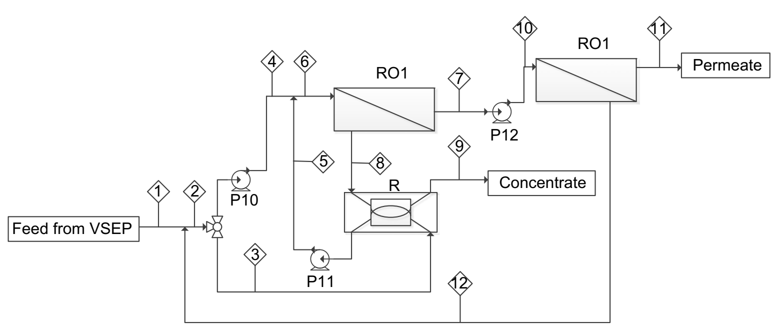
Cations	Value (ppm)	Anions	Value (ppm)
Ca^{2+}	820	HCO_3^-	304
Mg^{2+}	2,620	SO_4^{2-}	4,000
Na^+	21,975.8	Cl^-	40,520.4
K^+	780	F^-	2.8
NH_4^+	0	NO_3^-	1.2
Ba^{2+}	0.1	B^{3+}	8
Sr^{2+}	26	SiO_2^{2-}	1
Total TDS = 71,127 ppm			

3.3. *Case study C (TDS 106,670 ppm)*

The sketch of the RO section for this case study is reported in Fig. 5. The ionic composition of produced

Table 5
Operative RO membrane parameters—case study B

Stage number	1	Feed flowrate (m^3/d)	91.2
Vessel number	1	Permeate flowrate (m^3/d)	22.80
Elements number	3	Retentate flowrate (m^3/d)	68.4
Total active area (m^2)	122.4	Recovery (%)	25
TDS feed (ppm)	71,127	Feed pressure (bar)	80.4



	Units	1	2	3	4	5	6	7	8	9	10	11	12
Flowrate	m^3/h	3.40	3.80	1.0	2.80	1.0	3.80	0,57	3.23	3.23	0.57	0.19	0.4
Pressure	bar	1	1	80.4	80.4	80.4	80.4	1	80.1	1	2.7	1	2.7
TDS	ppm	103348	106670	106670	106670	106670	106670	94686	108785	108785	94686	55.1	134901

Fig. 5. Sketch of the RO section for the case study C.

water in this case study is reported in Table 6. In this case, because of the high water salinity, we consider two RO stages, the permeate coming out from the first stage, entering the second with a larger recovery factor (35%).

Table 6
Ionic composition of produced water for case study C

Cations	Value (ppm)	Anions	Value (ppm)
Ca^{2+}	1,230	HCO_3^-	456
Mg^{2+}	3,930	SO_4^{2-}	6,000
Na^+	32,977.4	Cl^-	60,781.4
K^+	1,170	F^-	4.2
NH_4^+	0	NO_3^-	1.8
Ba^{2+}	0.15	B^{3+}	12
Sr^{2+}	39	SiO_2^{2-}	1.5
Total TDS = 106,670 ppm			

Table 7
Operative RO membrane parameters—case study C

	First stage	Second stage
Membrane type	SWC4 Max	SWC5 Max
Stage number	1	1
Vessel number	1	1
Membrane number per vessel	6	3
Total active surface (m^2)	244.8	22.3
Feed flowrate (m^3/d)	91.2	13.7
Permeate flowrate (m^3/d)	13.68	4.79
Retentate flowrate (m^3/d)	77.5	8.9
Recovery factor (%)	15	35
Feed pressure (bar)	89.3	2.7

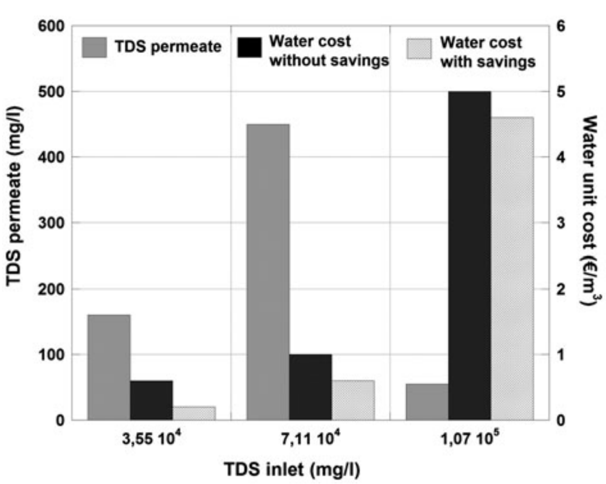


Fig. 6. TDS permeate and water unit cost, without and with savings coming from water reuse, for the three case studies.

Table 8
TDS for the three case studies

	Case study A (one stage)	Case study B (one stage)	Case study C (double stage)
TDS permeate (ppm)	160.2	449.5	55.1

The features of the two membrane stages are reported in Table 7: to meet the requirements for this case study, the feed pressure to the first stage must be reduced, by recirculating 0.4 m^3/h of the retentate flow from the second stage, thus the feed pressure to the first stage drops to 89.3 bar (Fig. 5).

For comparison, Table 8 reports the collective results, in terms of final TDS content of produced water, for all case studies: The processed produced water always meets the requirements by law for the reuse (TDS from 400 to 2,000 ppm).

4. Cost analysis

We performed a cost analysis for the outlined three cases (Table 9): The total plant cost is represented by the fixed capital investment (FCI), computed as the sum of the manufacturing FCI (direct costs, for example, purchased-equipment cost for the case studies A, B, C, and auxiliaries) and the non-manufacturing FCI (indirect costs) [23].

To assess the plant economical sustainability, investment costs have to be compared with the economical save coming from water reuse (Fig. 6): hence, we computed the payback period, for example, the period (years) required to recover the initially invested capital (cost breakdown).

Putting water cost 0.37 €/m^3, the return times for cases A, B, and C are, respectively, 8, 15, and 88 years, and the savings R coming from the water reuse are as follows:

(1) $R(A) = 40.8\ m^3/d \times 365\ d \times 0.37$ €/m^3 = 5,510 €/year.
(2) $R(B) = 22.80\ m^3/d \times 365\ d \times 0.37$ €/m^3 = 3,079 €/year.
(3) $R(C) = 4.79\ m^3/d \times 365\ d \times 0.37$ €/m^3 = 647 €/year.

Table 9
Total plant cost

Item description	Unit cost (€)	Number of items A and B	Total unit cost A and B (€)	Number of items C	Total unit cost C (€)
Membrane RO	480	3	1.440	9	4.320
Vessel membrane RO	1.500	1	1.500	2	3.000
Sand microfilter	1.337	2	2.674	2	2.674
High-pressure RO pump	7.528	1	7.528	1	7.528
Booster pump	815	1	815	2	1.630
Tool for chemical preparation and dosing	3.062	1	3.062	1	3.062
Tool for antiscalant preparation and dosing	3.062	1	3.062	1	3.062
Purchased-equipment cost			*20.081*		*25.276*
Construction expenses, installation, piping, and valves	40%		8.032		10.110
Instrumentation and control electrical plant	30%		6.024		7.583
Process buildings	7%		1.406		1.769
Transportation	7%		1.406		1.769
Total			*36.949*		*46.507*
Contingency	10%		3.695		4.651
Contractor's fee	12%		4.434		5.581
Total plant cost			*~45.000*		*~56.700*

Considering the parameters listed in Tables 9 and 10, it is possible to estimate of the cost of water production (Table 11 and Fig. 6): In Fig. 6, it is possible to derive that the case study C, with a double filtration step, is characterized by the highest cost, but the lowest TDS content.

Estimated cost of the water must be deducted from the cost savings due to water reuse (see above):

(1) Case study A: annual cost of water − Cost savings $R(A) = 2.845$ €/year = ~0.2 €/m^3.

(2) Case study B: annual cost of water − Cost savings $R(B) = 4.941$ €/year = ~0.6 €/m^3.

(3) Case study C: annual cost of water − Cost savings $R(C) = 8.105$ €/year = ~4.6 €/m^3.

Water cost with savings coming from water reuse is also shown in Fig. 6, thus confirming that the double step of the case study C is largely the most expensive, purposed to high-purity water production only; on the other hand, if the water reuse is thought for agricultural purposes, processes A and B provide water with good properties at much lower costs.

Table 10
Cost coefficients

	Case study A	Case study B	Case study C
Production × year (m^3/year)	14,892	8,322	1,748
Total plant cost (from Table 9) (€)	45.000	45.000	56.700
Depreciation (%)	3	3	3
Annual membrane replacement (%)	20	20	20
Electric energy cost (€/kWh)	0.077	0.077	0.077
Energy (kWh)	1,890	2,653	2,984
Labor	20% 1 person for 6 months	20% 1 person for 6 months	20% 1 person for 6 months
Maintenance (%)	1.50	1.50	1.50

Table 11
Water production cost

	Case study A		Case study B		Case study C	
	Cost × year	Unit cost (€/m^3)	Cost × year	Unit cost (€/m^3)	Cost × year	Unit cost (€/m^3)
Annual plant fee[a]	1,350	0.09	1,350	0.16	1,700	0.97
Membrane replacement[b]	288	0.02	288	0.04	864	0.50
Energy	146	0.01	204	0.02	230	0.13
Chemicals[c]	894	0.06	499	0.06	105	0.06
Labor	5,000	0.33	5,000	0.60	5,000	2.85
Maintenance	676	0.05	676	0.08	851	0.50
Total	8,354	~0.6	8,017	~1.0	8,750	~5.0

[a]Total plant cost × depreciation.
[b]Membrane cost × 20%.
[c]A unit cost of 0.06 €/m^3 is considered.

5. Conclusions

The reuse of water is a hot topic in the industrial practice; the produced waters in oil extraction and production represent a good water source for reuse, when properly treated.

We report a preliminary study regarding the reliability and feasibility, on an economical ground too, of membrane processes, embedded in a treatment train. The analysis of three case studies opens to many applications of the process for waters at different salinity and for different final fates of the treated waters, showing the costs are lower when simpler processes are applied for non-food water reuse purposes.

This approach may foster the application of innovative membrane devices (VSEP) coupled with traditional RO modules, which reducing fouling allow to get very good performance of the treatment processes.

Eventually, the cost analysis provides further indication for the industrial application, which, we think, is being more and more convenient due to the increasing costs for industrial and potable water.

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