



# Ceramic membrane filtration of produced water: Impact of membrane module



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## ABSTRACT

Produced water (PW) generated from oil exploration requires rigorous removal of suspended matter (free oil and particulate solids) as tertiary treatment (downstream of hydrocyclone and gas flotation) if it is to be re-injected into low-permeability reservoirs. The viability of membrane filtration for this duty is largely dependent on sustaining a high membrane flux to minimise the process footprint.

A pilot-scale study of PW filtration using crossflow multi-channel ceramic membrane technology has been conducted to identify the appropriate membrane characteristics for sustaining the flux whilst maintaining the required treated water quality. Membranes based on two materials (silicon carbide, SiC, and titanium dioxide, TiO<sub>2</sub>) and two different pore sizes were challenged with real PW samples taken from oil platforms operating on the Arabian Gulf. The membranes were characterised according to the overall permeability decline rate and the end permeability.

Results suggest that SiC membranes outperform TiO<sub>2</sub> ones with respect to sustainable permeability under the same operating and maintenance conditions. The SiC microfiltration membrane provided anomalously high permeabilities but also the highest fouling propensity. Results suggest that whilst the high fluxes (1300–1800 L m<sup>-2</sup> h<sup>-1</sup>) are attainable for the technology, this is contingent upon the application of an effective chemical clean.

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## 1. Introduction

Produced water (PW) from oil and gas exploration represents the most significant volume waste product of the petroleum industry, and demands treatment for oil and solids removal prior to discharge. For offshore platforms, produced water reinjection (PWRI) back into the aquifer offers a sustainable disposal route due to the significantly reduced net environmental impact: the PW is employed in place of the seawater to displace the oil, and is reused [4,23]. All additives are also subsequently largely reused, provided they remain in the aqueous phase. However, for “tight” or low permeability reservoirs - associated with carbonate strata - PWRI demands removal of particles down to 3–5 μm in size and <5 mg/L concentration [14,23,12] so as to limit plugging of the reservoir pores and sustain oil displacement. Moreover, the technologies must be robust to fluctuating loads of suspended and sparingly soluble materials, where the latter can form scales both

within the reservoir and the unit process itself [22]. Finally, the removal of microbial species, and specifically sulphate-reducing bacteria, would be expected to mitigate against the build-up of highly insoluble sulphide salts in the reservoir.

Significant research into membrane filtration technologies (ultrafiltration, UF, and microfiltration, MF) for PW tertiary treatment has been reported over the past 20 years, with increasing focus on ceramic membranes (CMs) operated in the crossflow mode. Operation in the crossflow, rather than dead-end, mode permits longer filtration cycles since the shear acting on the membrane as the retentate flows across it serves to suppress membrane fouling. CMs offer the advantage over polymeric materials of increased fouling resistance and operation at elevated temperatures. Their application to PW dates back to trials conducted in the early 1990s [31], with a significant number of studies performed since that time (Table 1).

Recent reviews [2,18] have tended to demonstrate a propensity for bench-scale studies based on synthetic wastewaters, with studies based on real petroleum wastewaters tending to relate to refineries [32,15,27,28]. Demonstration-scale studies, conducted

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**Table 1**  
Studies of MF/UF membrane filtration of PW.

Oil concn., mg/L, water source	Scale	Material	Pore size, $\mu\text{m}$	Init flux, LMH	Fin flux, LMH	TMP, bar	Fin. perm (range), LMH/bar	Time, h	CFV, m/s	T, °C	References
250, synth PW	b(m)	AlO	0.2	200	26	0.7	18	2	0.24–0.91	40	[17]
	b(m)	AlO	0.8	1000	44	0.7	32	2	0.24–0.91	40	
5000, synth PW	b	AlO	0.2	110	22	1.25 ave	18	2	3–5	–	[30]
	b	ZrO	0.2	130	93		74	2	3–5	–	
6000, refinery	b	ZrO	0.2	240	120	1.1	109	1	2.6	25–60	[32]
6000, refinery	b	ZrO	0.2	240	175	1.1	159	1	2.6	25–60	
200–2000, veget. oil	b(m)	AlO	0.05	–	125–165	0.5–3	50–200	–	0.2–1.7	20–21	[11]
50,000, synth cutting oil	b(m)	ZrO	0.02	–	100–200	0.8–4.5	44–125	–	3	50	[26]
	b(m)	PAEK	100 <sup>b</sup>	–	140–170	1–4.5	38–140	–	3	50	
	b(m)	ZrO	0.02	–	35	0.8–4.5	8–88	–	0.6–3	50	
	b(m)	PAEK	100	–	25	0.1–4.5	6–44	–	0.6–3	50	
366, PW	b	PS <sup>a</sup>	0.007	225	128	1–1.7		1	–	–	[5]
	b	PS <sup>a</sup>	0.006	100	70	1–1.7		1	–	–	
200–1000, tank dewatering effl.	b(m)	AlO	0.2	128	28	1	28	2	–	60	[7]
	b(m)	TiO	0.05	80	4	1	<5	2.5	–	60	
	b(m)			120	30	1	120–30	2	–	60	
50–350, synth	p	AlO	0.1–0.5	–	80–175	0.06–0.25	400–800	2	4.5	27	[24]
–, gas field PW	p	Al–ZrO	0.05	–	170–255	–	190–250	600	–	25	[25]
1000, synth C8–C12	b	Mullite <sup>a</sup>	0.29	100	70–75	3	33–23	2	1.5–2	25	[1]
3000, refinery	b	PS	0.1–0.2	145	65	1.5	50–15	11	1.25	27	[15]
–, SAGD effl.	p	AlO	0.05	200	–	1.52	132	–	–	45	[10]
	p	ZrO	20 <sup>b</sup>	200	–	1.52	45	–	–	45	
100, synth	b,p	ZrO	0.1	910	194–240	2	97–120	24	2	25	[27]
~60, refinery effl.	b,p	ZrO	0.1	910	175	2	88	24	2	25	
~250, refinery effl.	b	ZrO	0.1	1000	290	1.5	193	–	3	45	[28]
9/43, PW	p	AlO	0.2	–	295–312	2.5	118–125	–	3	35–60	[21]
	b	PAN	20 <sup>b</sup>	–	104–280	5	20–36	–	1.5	25–55	

<sup>a</sup> Bespoke membranes.

<sup>b</sup> Molecular weight cut-off.

under conditions fully representative of those prevailing at full scale [13,19,20], are limited in experimental detail and are not necessarily subject to rigorous control of the filtration operating conditions. Against this, trials conducted on both real PW and synthetic effluents have tended to demonstrate the significantly more challenging nature of PW than synthetic waters [7,5,27]. Evidence suggests that both the dissolved and suspended content of PW impacts on flux [1,11].

The filtration profile of crossflow MF/UF CMs challenged with PW and operated under constant conditions of transmembrane pressure (TMP) is typified by a rapid decline in flux to a neo-steady-state value. The decline appears to be dependent on a number of different system parameters, including feedwater composition, hydraulics (primarily crossflow velocity, CFV), and temperature, as well as the characteristics of the membrane itself. The rapid flux decline demands both physical cleaning, i.e. back-flushing or backpulsing, and chemical cleaning in place (CIP) to sustain the flux, with physical cleaning efficacy demonstrated in a number of studies [14,7,8,24,21,27,28]. As a consequence of the large number of variables impacting on operation, reported permeability values for CMs challenged with PW or its analogues have been very wide-ranging, from below 20 to ~800 LMH/bar (Table 1).

Whilst the studies of CMs challenged with oil-laden waters have been extensive, they have tended to be constrained either by feedstock availability, limiting the filtration time (to 1–2 h for many of the studies listed in Table 1), and/or by recourse to analogues. On the other hand, evidence from demonstration trials [13,19] and recent extended trials [28] indicate very significant permeability loss over a period of several days. There have also been few studies comparing candidate CM materials; comparative studies have tended to focus on the relative performance of ceramic and polymeric membranes [26,21]. Studies which include CM chemical cleaning efficacy [7,28] have not encompassed long-term use of the cleaned membrane. Moreover, the CMs investigated have been largely limited to oxides of aluminium,

zirconium and titanium, with few controlled studies of silicon carbide (SiC).

This study aims to address the above using real PW samples taken from oil platforms operating on the Arabian Gulf and a multi-stream pilot plant fitted single and multi-channel membranes. Two different CM materials of two different pore sizes have been used. An operating protocol was developed which allowed the installed membranes to be challenged with a range of feedwater suspended oil concentrations over an extended time period. The programme aimed to permit a comprehensive and quantitative appraisal of the relative efficacy of the different membranes for sustaining both treated water quality and membrane permeability.

## 2. Materials and methods

### 2.1. Materials

The pilot plant (Fig. 1) comprised two streams of tubular membrane modules, of varying characteristics (Table 2), operated in crossflow mode. The plant, supplied by Liqtech International (Ballerup, Denmark), allowed operation at a fixed conversion with automated physical cleaning provided by backflushing and backpulsing and manually-applied chemical cleaning in place (CIP). Recirculation of both retentate and permeate streams was possible, with retentate recirculation used to increase the feedwater suspended oil concentration.

All silicon carbide (SiC) membranes were provided by Liqtech, and titanium dioxide (TiO<sub>2</sub>) ones by TAMI Industries (Nyons, France). Four membranes were studied, the elements all being ~305 mm in length and 25 mm diameter and with various specifications of material type, channel characteristics (number, dimensions and geometry) and pore size (Table 2).

The PW was sampled downstream of the induced gas flotation (IGF) stage of a classical hydrocyclone-IGF PW treatment train. It was shipped in three lots of 5-tonne batches from an oil platform

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