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Investigations on the use of different ceramic membranes for efficient oil-field produced water treatment☆

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ABSTRACT

Efficient performance of the combination of treatment processes for oilfield produced water generated from oil tank dewatering was investigated in the study presented below. By-produced wastewater is generated in significant quantity during exploitation of crude oil and gas from onshore and offshore production operations. This wastewater, commonly referred to as “produced water”, has distinctive characteristics, due to their organic and inorganic compounds. However, these characteristics change from well to well. The treatment process investigated here consists of a pre-treatment step utilizing microfiltration (0.1 and 0.2 µm pore size filters) and/or a simulated batch dissolved air flotation (DAF), and a multistage post-treatment step utilizing cross-flow ultra- (0.05 µm pore size and 20 kDa molecular weight cut-off filters), and nanofiltration (1 and 0.75 kDa MWCO filters). Filters used were ceramic membranes. To determine the separation capability of the processes described, various parameters, such as trans-membrane pressure varying from 0.5 to 2 bar, cross-flow velocity in the range of 0.6 to 1.3 m/s, influent oil concentration ranging from 32 to 5420 parts per million (ppm) and different membrane cleaning methods used were investigated. The average permeate flux varied from 3.4 to 3300 l/h m^2 bar, total oil removal was up to 99.5% and total organic carbon removal reached 49%.

1. Introduction

The onshore and offshore conveyance of crude oil and natural gas is associated with the co-production of significant quantities of wastewater, referred to as “produced water”. Produced water is considered the largest volume waste stream in the exploration and production process of oil and gas [1]. Oilfield produced water has distinctive characteristics due to organic and inorganic matter. Mainly, it includes salts and oil hydrocarbons, which may be toxic to the environment. However, its characteristics and volume vary greatly from well to well and depend on the lifetime of a reservoir [4]. Over time, the percentage of water increases and the percentage of product decline. Hence, produced water is difficult to treat. Disposal, re-injection and reuse are the available handling options of produced water [2,3]. Disposal of produced water requires imperative environmental regulations and produced water re-injection (PWRI) requires skilful planning and treatment to meet the quality needed for re-injection water to avoid formation damage.

In general produced water treatment is approached through de-oiling and de-mineralizing before its disposal or utilization. Various technologies and methods exist for treatment of oil field produced water. Successful treatment generally requires a series of pre-, and post-treatment operations to remove various contaminants. Traditional technologies such as clarifiers, dissolved air flotation, hydrocyclones and disposable filters and absorbers respectively [*e.g.*, 5] do not achieve the separation efficiency required for beneficial use of produced water by meeting potable and irrigation water quality standards [6]. The practicality of using treated produced water for beneficial purposes depends on a number of factors, including the volume of water available, the existence of a local need for water, and the amount of treatment required to meet government or industry-use standards [7].

Membrane technology is used in industrial processes, in industrial wastewater treatment, and is utilized currently for oil field produced water treatment [4,8,9]. Ceramic (or inorganic) membranes have attracted interest due to their superior mechanical, thermal, and chemical stability. The primary advantage of using ceramic membranes is the ability to accomplish the current and pending regulatory treatment objectives with no chemical pre-treatment. The study

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Table 1
Material and properties of the ceramic membranes used in this investigation.

Membrane	MF — Al_2O_3	MF — Al_2O_3	UF — TiO_2	NF — TiO_2	NF — TiO_2
Membrane material	Al_2O_3	Al_2O_3	TiO_2 / Al_2O_3	TiO_2 / TiO_2	TiO_2 / Al_2O_3
Cut-off	0.1 μm[a]	0.2 μm[a]	0.05 μm[a] and 20 kDa[a]	1000 Da[a]	750 D[a]
External diameter	25.4 mm	25.4 mm	25.4 mm	10 mm	10 mm
Internal diameter	10 mm	10 mm	10 mm	6 mm	6 mm
Length	450 mm	450 mm/ 70 mm	450 mm/ 70 mm	250 mm	250 mm
pH	0–14	0–14	0–14	0–14	0–14
Temp. max. [°C]	121	121	121	150	120

[a] As indicated by the manufacturer.

presented here focuses on the efficient development of single and combined treatment processes for oilfield produced water and different prepared feed solutions. The process consists of a pre-treatment step utilizing cross-flow microfiltration (MF) (0.1 and 0.2 μm pore size filters) and/or a simulated batch dissolved air flotation (DAF), and a multistage post-treatment step utilizing cross-flow ultra- (UF) (0.05 μm pore size and 20 kDa molecular weight cut-off filters), and nanofiltration (NF) (1 and 0.75 kDa molecular weight cut-off filters). Filters used were ceramic membranes. Various parameters potentially affecting the permeation and separation behaviour of the purification process such as trans-membrane pressure (TMP), cross-flow velocity (CFV), oil concentration in the feed and different membrane cleaning methods used were investigated through the measurement of the average permeate flux, the oil removal efficiency and the total concentration of organic compounds (measured as TOC).

2. Materials and methods

2.1. Ceramic membranes

Ceramic membranes used are tubular and consist of a porous support material (generally α-alumina), a minimum of one layer of decreasing pore diameter and a separating layer (α-alumina, zirconia, etc.) covering the internal surface of the tube [5]. Asymmetric multilayer Al_2O_3 and TiO_2 ceramic MF, UF and NF membranes in different stainless steel housing (Table 1 and Fig. 1) were used.

2.2. Fouled membranes and cleaning methods

Fouling through suspended oil and grease, particles and colloids, salts and various other trace metals is one of the most common problems and a major operational factor encountered in produced water treatment applications of membranes [10]. To reduce membrane fouling, the effect of chemical cleaning and back flushing on ceramic membranes was investigated.

Table 2
Characteristics of the model solutions (a–c) and tank dewatering produced water used.

Parameter	Model solution (a)	Model solution (b)	Model solution (c)	Variation range of tank dewatering produced water
Dispersed oil	113 mg/l	5420 mg/l	148.6 mg/l	200–1000 mg/l
pH value	7.5	6	7.3	6.0–8.0
Conductivity	213 μS/cm	162 μS/cm	168 μS/cm	20,000–80,000 μS/cm
TOC	94 mg/l	41.1 mg/l	23 mg/l	200–2000 mg/l
wt.%	5	20	10	–
Waste oil type-number	1	2	3	1

2.2.1. Chemical cleaning

Chemicals used for membrane cleaning were lye solutions (1% (w/w) NaOH solution, Ultrasil P3-14, Ultrasil P3-10 for 30 to 60 min), dissolved in distilled water. Cleaning efficiency was evaluated determining the water flux after cleaning relative to the initial water flux.

2.2.2. Back flushing

Back flushing is a method applied commonly to remove a layer of retained material [11]. Here, the flow was reversed to flush the membrane pores from the permeate and, thus, to release material retained in the membrane pores.

2.3. Preparation of model solution

Three different model oily wastewaters (model solutions a–c) were prepared in a heated stirred tank through mixing waste oil (5%, 10%, 20% (w/w)) with distilled water for 30 min at 60 °C (Table 2). To simulate a primary process of separation from the oil, the mixture was left for 30 min to clarify. The free oil was recovered and pumped back to the waste oil tank. The model oily wastewater showed a uniform yellowish colour.

2.4. Oilfield produced water characterization

Characteristics of produced water from oil and gas fields, mainly containing salts and oil hydrocarbons, vary and may differ significantly from well to well [12]. Samples of produced water from tank dewatering were obtained from German BP AG, Oil Refinery Emsland, Lingen. The concentration range of components in tank dewatering produced water (Table 2) used in this study is given.

2.5. Membrane-assisted continuous reactor

The cross-flow membrane filtration equipment (MF, UF, NF) was conducted using a continuous stirred tank reactor (CSTR) with a

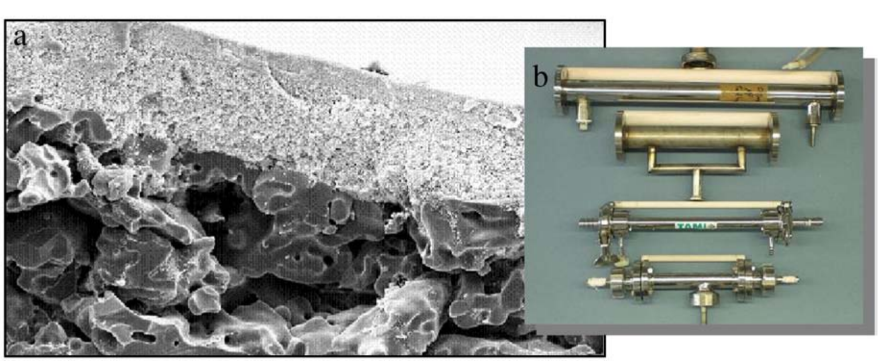

Fig. 1. a) SEM-micrograph of a ceramic UF-membrane, b) Different ceramic membranes and housing.