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Ebrahimi, M., Schmitz, O., Kerker, S., Liebermann, F., & Czermak, P. (2013). Dynamic cross flow filtration of oil-field produced water by rotating ceramic filter discs. Retrieved from http://krex.ksu.edu

Published Version Information

Citation: Ebrahimi, M., Schmitz, O., Kerker, S., Liebermann, F., & Czermak, P. (2013). Dynamic cross-flow filtration of oilfield produced water by rotating ceramic filter discs. Desalination and Water Treatment, 51(7-9), 1762-1768.

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Digital Object Identifier (DOI): doi:10.1080/19443994.2012.694197

Publisher's Link: http://www.tandfonline.com/doi/abs/10.1080/19443994.2012.694197

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Dynamic Cross Flow Filtration of Oil-Field Produced Water by Rotating Ceramic Filter Discs

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KEY WORDS: Oil-field, Produced Water Treatment, Ceramic Membrane, Rotating Filter, Membrane Fouling

Abstract

Experimental results will be presented for the dynamic cross-flow microfiltration (MF, 0.2 µm pores) and ultrafiltration (UF, 7 nm pores) of oil-field produced water as well as oily model solutions. The purpose of this research paper is to assess the effects of process parameters; membrane rotational speed (1200, 1500 and 1800 rpm), volume concentration factor (VCF) and feed characteristics in terms of oil and TOC separation capability, permeability and permeate quality. For this, a series of membrane filtration experiments were carried out systematically using rotating ceramic filter discs in a fed-batch operating mode. The variation of membrane rotational speed was found to minimize membrane fouling in a significant amount. High oil (>99%) and TOC rejection (>98%) was achieved with both, MF and UF membranes, independent of the rotational speed and the feed concentration.

1. Introduction

The term “produced water” (PW) describes the largest waste water stream that is generated and brought to the surface along with oil or gas during exploration and production operations. On a global spectrum, it is estimated that three barrels of water are produced for every barrel of crude oil [1]. PW is a hazardous waste, difficult to treat and generally contains a mixture of native formation water, dissolved salts, different types of oil (dispersed/dissolved/free), dispersed solid particles (sand and silt), trace heavy metals and any small amount of chemicals added during the production activities [2]. Major pollutant in oilfield wastewater is oil; the concentration may range between 100 and 1000 mg/L or still higher [3].

Due to the increasing volume of waste all over the world in the current decade, the outcome and effect of discharging PW on the environment has lately become a significant issue of environmental concerns [4]. At present, PW is typically disposed in injection wells as waste or for pressure maintenance of the reservoir. Treatment of this wastewater could improve the economic viability of these oil and gas fields and lead to a new source of water for beneficial use [5]. Several techniques to treat PW are already deployed. Current technologies are based on gravity separators, air or gas flotation, chemical flocculation, plate coalescers, hydrocyclones and disposable filters/absorbers [6]. None of these technologies is capable of removing all types of dissolved components within a single process; by now multiple processes must always be run in series [7]. Therefore, there is a need for new PW treatment technologies due to increase focus on water conservation and environmental regulation.

Membrane separation is a useful method for the treatment of oily waste waters. Several studies have shown that cross-flow microfiltration (MF), ultrafiltration (UF) and nanofiltration (NF) membrane processes are suitable and effective techniques in concentrating O/W emulsions and for production of reusable water, respectively [8-12]. Decline of permeate flux with time observed at high oil concentrations is a fundamental problem in classical cross-flow membrane separation processes during the treatment of oily waste waters. This is mostly attributed to the phenomenon of concentration polarization and fouling at high oil concentrations. To overcome these problems different types of high-shear devices have been developed.

Dynamic cross-flow filtration has several essential advantages compared to conventional cross-flow systems. Substantial energy savings, high specific filtrate flux rates and independence of transmembrane pressure (TMP) from cross-flow speed are some of these advantages. With rotating cross-flow systems, over flow – i.e. cake layer control – is generated by filter elements rotating through the feed. Since the entire over flow is realized by rotation, only the permeate outflow is compensated, i.e. pumped through the filter module [13]. Industrial dynamic filtration module generally consist of three types; discs or rotors rotating near fixed membranes or rotating organic/ceramic disc membranes and vibrating systems [14].

The main purpose of this study is to investigate the application and the potential of a compact rotating disc device, as it is one of the few dynamic filtration systems available with ceramic membranes, for the efficient treatment of oil-field produced water generated from tank dewatering (TDPW) and different prepared model solutions (MS). The effects of operating variables on permeate flux of the rotating ceramic MF and UF filter discs and rejection efficiency for oil and TOC were investigated.

2. Material and methods

2.1 Rotating membrane device

Compact rotating disc filter (CRD) used in all MF and UF steps, and designed for up to 3 membrane discs with 152 mm diameter, was manufactured by Novoflow GmbH (Rain/Lech, Germany) for test, lab and remote applications. A schematic presentation of the rotating disc system and the experimental apparatus is given in Fig. 1. The system was equipped with a ceramic membrane disc (Tab. 2 and Fig. 2) and two dummies mounted on a hollow rotating shaft. The shaft can rotate at adjustable speeds, ranging from 60 to 1800 rpm. The MF and UF experiments were carried out fed-batch wise. Fresh feed solution is added to the temperature controlled and stirred feed tank at the same rate as permeate is collected. Moreover the feed solution was/will be concentrated over time. All experiments were performed in duplicate. The feed stream entered the vessel housing under pressure and was distributed across the ceramic disc surface by hydraulic action. Permeate was collected in a beaker placed on an electronic scale connected to an automation and process control system in order to measure the permeate flux.

Experiments were performed so that the effects of membrane rotational speed (1200, 1500 and 1800 rpm) on flux, total oil and TOC removal can be properly understood. During MF and UF of oily model solutions and tank dewatering PW, two process parameters were kept constant; the applied TMP was maintained at 1bar and the process temperature at 50°C.

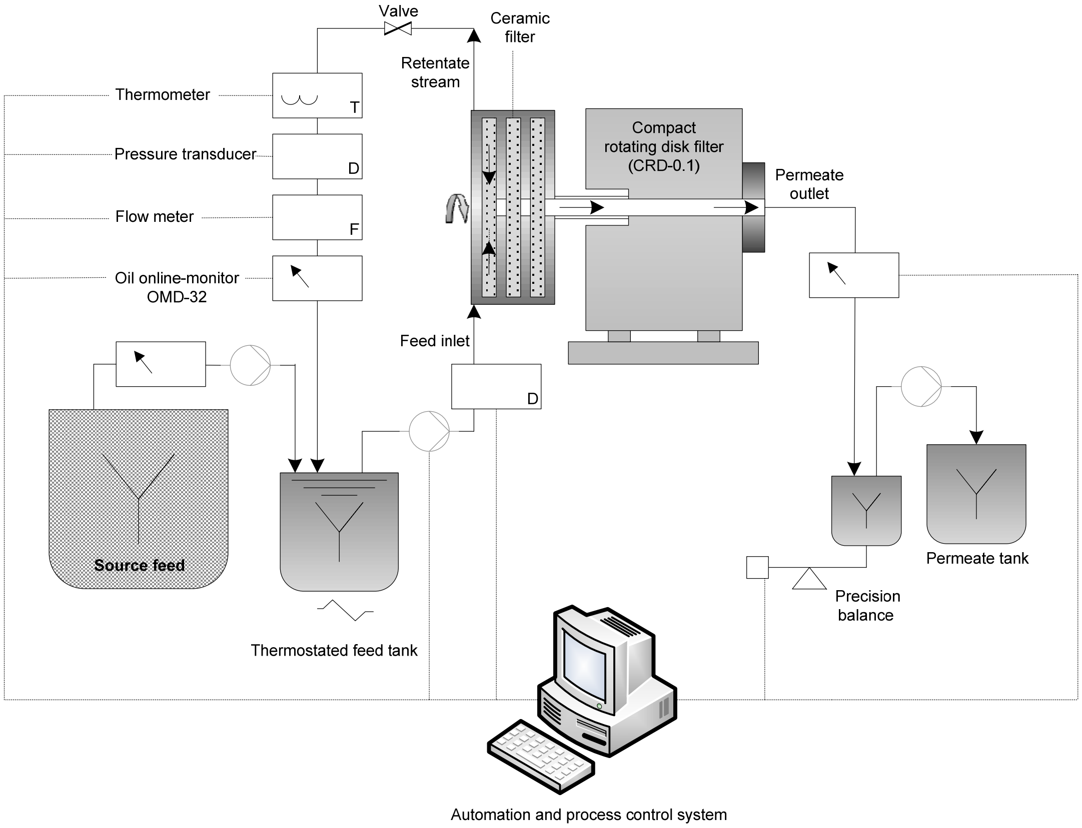

Fig. 1 Schematic diagram of the laboratory scale rotating disc system for the dynamic cross-flow MF- and UF

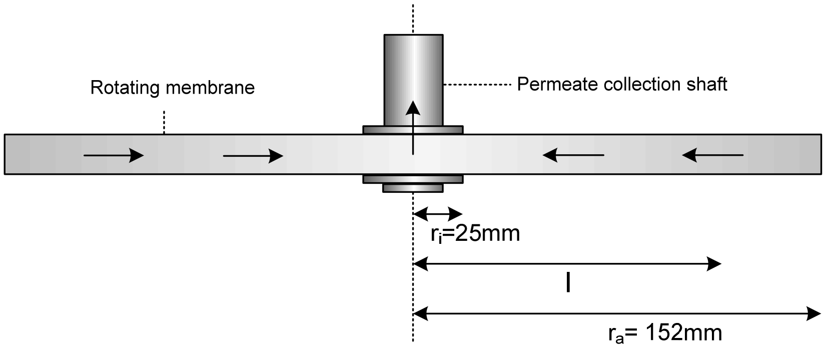

Fig. 2 Cross section of the rotating ceramic membrane disc

2.2. Membranes

Ceramic membranes used in the CRD were micro- and ultrafiltration discs made by KERAFOL Keramische Folien GmbH, Eschenbach, Germany. The membrane characteristics, according to the manufacturer, are summarized in Table 1.

Table 1 Characteristics of ceramic disc membranes used in this investigation

Membrane	MF	UF
Material	Al_2O_3	Al_2O_3 / $MgAl_2O_4$
Pore size	0.2 µm	7 nm
Outer radius	152 mm	152 mm
Inner radius	25 mm	25 mm
Thickness	4.5 mm	4.5 mm
Area	360 cm²	360 cm²
pH	0-14	1-14
Temperature Max.	121°C	121°C

2.3. Membrane and system cleaning procedure

After every experiment, the system was shut down and the membrane housing was drained. In the following the disc membrane and the system were cleaned with lye solutions (1 % (w/w) NaOH solution, alternatively, Ultrasil P3-14 or Ultrasil P3-10 (Henkel, Germany) dissolved in distilled water. Finally the system was drained and flushed again with hot distilled water. After completing the cleaning procedure, the clean water fluxes were measured with distilled water at different temperatures (room temperature and 50°C), varied rotating speed (1200, 1500 and 1800 rpm) and 1 bar TMP.

2.4. Oily wastewaters

In order to investigate the performance of ceramic filter discs, oily model solutions (OMS) and tank dewatering produced water (TDPW) were used. OMS were prepared by premixing the crude oil and water with a rotor stator homogenizer in concentrations of approximately 5000 ppm oil in water, and subsequently passing this emulsion in single pass through a high pressure homogenizer. For adjusting the needed oil concentration, the prepared emulsion was diluted with water and measured with the OMD-32 (explanation, see chapter 2.5), simultaneously. Samples of TDPW were obtained from German BP AG, Oil Refinery Emsland, Lingen, Germany. The general characteristics of TDPW and OMS are presented in Table 2.

Table 2 Characteristics of the OMS and TDPW used in this investigation; N.A.: not available.

Index	Variation range of prepared OMS	Variation range of TDPW
Dispersed oil	30-200 mg/l	200-1000 mg/l
pH value	6.2-6.9	6.0-8.0
Conductivity	8.3-15.7 µS/cm	20,000-80,000 µS/cm
COD	105-169 mg/l	N.A.
TOC	50-658 mg/l	200-2000 mg/l
Iron	N.A.	66 mg/l
Zinc	N.A.	0.55 mg/l

2.5. On-line oil-in-water measurement

The continuous oil in water measurement down to part-per-million (ppm) levels was performed using an innovative prototype “OMD-32”. It is a next-generation on-line monitoring device, developed for industrial applications by DECKMA HAMBURG GmbH, Hamburg, Germany. The measuring principle is based on light scattering. An optical sensor array measures a combination of light scattered and absorbed by oil droplets in the sample stream. The sensor signals are then processed by a microprocessor to produce linearized output [15].

2.6. Transmembrane pressure (TMP)

The rotation of the membrane causes a backpressure in the membrane housing, which reduces the effective average transmembrane pressure. This pressure depends on the applied rotation speed and the diameter of the membrane. In order to estimate the pressure offset for a certain rotation speed, the permeate and the retentate side of the filtration system were filled with RO-water. When the system was filled, the feed pump was turned off, the inlet of the retentate side was closed and the rotation speed was set. After stabilization of the displayed pressure the value was recorded and used as a TMP-offset in the subsequent filtration experiment.

3. Definitions

Permeate flux (J)

The permeate flux was measured during the filtration process and is defined as:

$$Permeate\ flux\ (\mathrm{J}) = \frac{permeate\ volume\ collected}{disk\ membrane\ area\ \times collection\ time}$$

TOC removal efficiency

The percentage of retention of the TOC at time t was calculated by comparing the concentrations of the TOC in permeate with those in the concentrate as follows:

$$\mathrm{R} = \left(1 - \frac{C_{p(t)}}{C_{c(t)}}\right) * 100\%$$

where R is the retention of TOC in % at time *t,* $\boldsymbol{C_{p(t)}}$ the concentration in the permeate samples in mg/l at time t, and $\boldsymbol{C_{c(t)}}$ the concentration in the concentrate in mg/l at the same time.

Volume concentration factor (VCF)

The volume concentration factor (VCF) of the feed solution in semi-batch operation is given by:

$$VCF = \frac{V_{feed}}{V_{retentate}} = 1 + \frac{V_{permeate}}{V_{retentate}}$$

where VCF is the concentration factor, $V_{permeate}$ is the volume of permeate produced at any given time, V_{feed} is the feed volume and $V_{retentate}$ is the retentate volume.

4. Results and discussion

4.1. Effect of the rotational speed on MF-membrane performance

In Fig. 3 the variation of permeate flux and volume concentration factor (VCF) versus operation time at two different membrane rotational speeds (1200 and 1800 rpm) are presented for a 0.2 µm MF ceramic disc during the treatment of oily model solution. Every run was performed at a constant temperature of 50°C, under a constant TMP of 1 bar and a feed oil concentration of 30 ppm. For both membrane rotational speeds it was shown that permeate flux decay could be divided into three distinctive regions such as rapid flux decay (after 200 min of operation time), slow flux decay (after 900 min) and constant periods (after 1100 min) which corresponds to a balanced mass transfer through the membrane. The stabilized permeate flux at the lower rotational speed (1200 rpm) was 64% lower than that at 1800rpm.

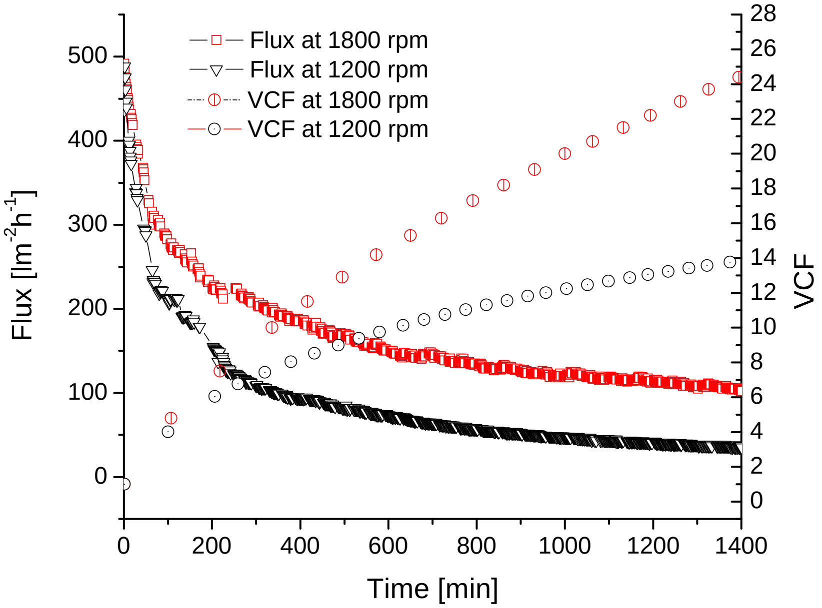

Fig. 3 Variation of permeate flux and volume concentration factor with time for two rotation speeds (1200 and 1800 rpm) using a 0.2 µm MF rotating disc; oily model solution, 30 ppm; TMP, 1 bar; temperature, 50°C.

It is well known that high shear rates at the membrane, caused by high rotation speeds, could reduce concentration polarization and cake formation during the dynamic cross-flow filtration [16].

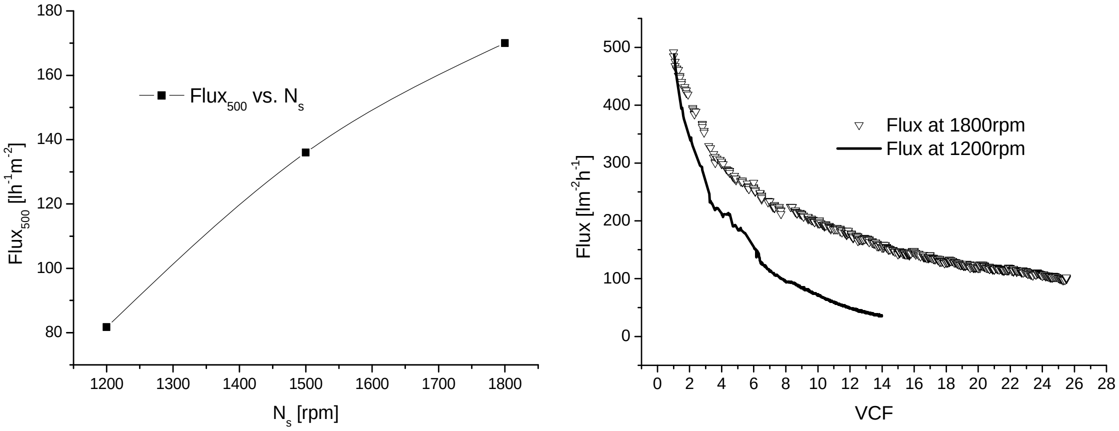

Fig. 4, left) Effect of three rotational speeds (1200, 1500 and 1800 rpm) on flux at a particular instant of time (t=500 min) using a 0.2 µm MF rotating disc; oily model solution, 30 ppm; TMP, 1 bar; temperature, 50°C, **right)** Variation of permeate flux versus volume concentration factor for two rotation speeds (1200 and 1800 rpm) using a 0.2 µm MF rotating disc as well.

Fig. 4, left, shows the effects of the variation of rotational speed (1200, 1500, and 1800 rpm) on the permeate flux obtained with a 0.2 µm MF ceramic membrane. The permeate flux, however, increased from 82 l h^{-1} m^{-2} to 170 l h^{-1} m^{-2} by increasing rotational speed from 1200 rpm up to 1800 rpm. The permeate flux modifications plotted against rotating speed at VCR 1 to 14 are demonstrated in Fig. 4, right. Based on these data, it could be observed that an increase in volume concentration factor from 1 to 10 results in a rapid decrease in the membrane filtrate flow at both rotational speeds considering the same oil concentration of 30 ppm of the oily model solution. The reason is the high concentration polarization for concentrated feeds. For a given VFC higher than 1, however, the increase in the disc rotational speed caused an increase in the filtrate flow.

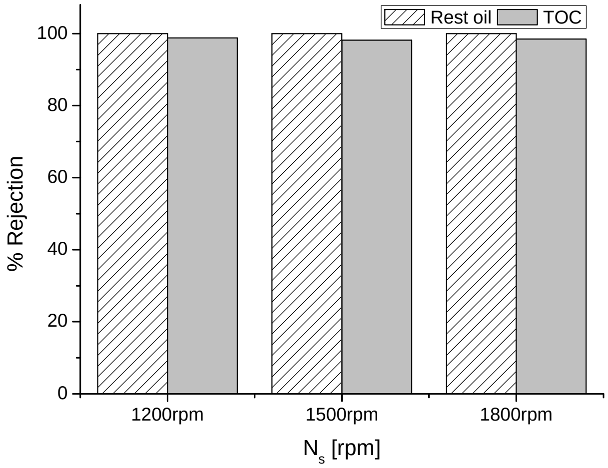

Fig. 5 Oil and TOC rejection for three rotation speeds using a 0.2 µm MF rotating disc; oily model solution, 30 ppm; TMP, 1 bar; temperature, 50°C.

The data illustrated in the Fig. 5 showed that the 0.2 µm ceramic membrane disc gave an excellent oil and TOC rejection of more than 99% and 98%, respectively during the treatment of oily model solution. It was also remarkable, that the removal efficiency of oil and TOC was independent of membrane rotational speed.

4.2. Effect of the rotational speed on UF-membrane performance

In order to investigate the effect of membrane rotational speed on the performance of ultrafiltration during the treatment of oily model solution, the permeate flux profile for two membrane speeds (1200 and 1800 rpm) operated over an applied TMP of 1 bar and feed concentration of 30 ppm has been shown in Fig. 6. It can be observed that under fixed operation conditions, membrane rotation plays an important role in determining the permeate flux. The permeate flow rate rises from 198 l h^{-1} m^{-2} at membrane rotational speed of 1200 rpm to 250 l h^{-1} m^{-2} at a speed of 1800 rpm. High UF membrane rotation speed creates substantial turbulence near the membrane surface, which minimizes the extent of concentration polarization and may reduce fouling at the membrane surface.

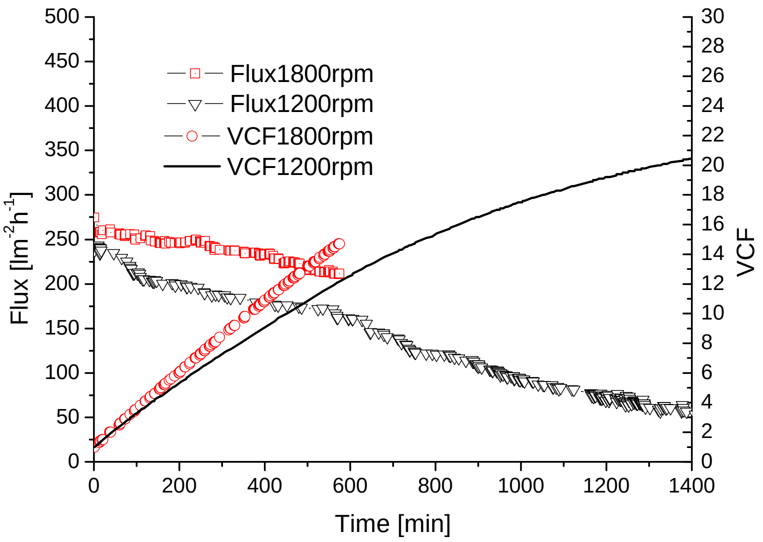

Fig. 6 Variation of permeate flux and oil content in concentrate stream with time for three rotation speeds (1200 and 1800 rpm) using a 7nm UF rotating disc; oily model solution, 30 ppm; TMP, 1 bar; temperature, 50°C.

Fig. 7, left, shows, that the permeate flux at any fixed VFC was found to be higher at a rotational speed of 1800 rpm in comparison to 1200 rpm.

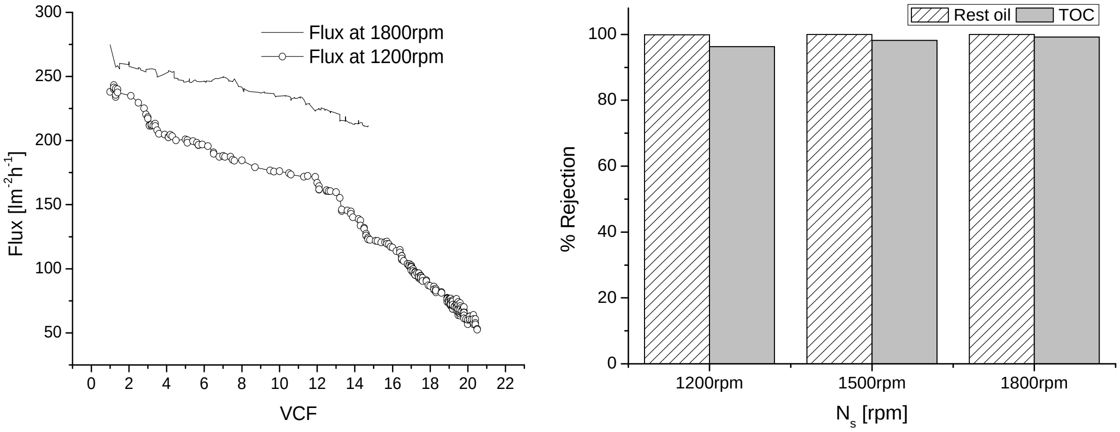

Fig. 7 left) Variation of permeate flux versus volume concentration factor for two rotation speeds (1200 and 1800 rpm) using a 7 nm UF rotating disc, **right)** Rest oil and TOC rejection for three rotation speeds (1200, 1500 and 1800 rpm) using a 7 nm UF rotating disc as well; oily model solution, 30 ppm; TMP, 1 bar; temperature, 50°C.

Fig. 7, right, represents the measured rejection efficiency of oil and TOC for the investigated 7 nm UF membrane at three rotating speeds (1200, 1500 and 1800 rpm) under fixed operating conditions (TMP=1 bar). For oil and TOC rejection values > 99% and > 98%, respectively, could be reached.

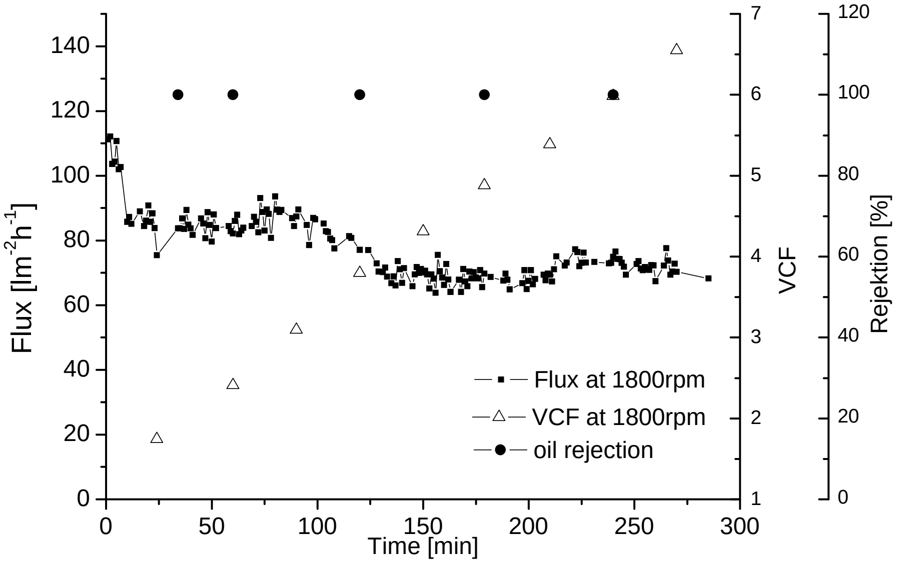

Fig. 8 Variation of permeate flux and oil content in concentrate stream with time for three rotation speeds (1200 and 1800 rpm) using a 7 nm UF rotating disc; tank dewatering PW, 40 ppm; TMP, 1 bar; temperature, 50°C.

Fig. 8 shows the measured filtrate flow rate and the fouling behavior versus time for a 7 nm ceramic UF membrane disc during the treatment of tank dewatering produced water with an initial oil concentration of 40 ppm in a dynamic filtration experiment. Although real PW had higher oil and TOC concentration, compared to oily model solutions used (Tab. 2), the rejection efficiency for oil was more than 99 %, indicating that UF treatment of tank dewatering PW by CRD module could obtain very high permeate quality for reuse, and VCF could be raised to a level between 2 to 6. The stability of the permeate flow after 150 min of operation time represents the stabilization of the filling of the membrane due to shear rates caused by the rotation speed of the membrane disc.

Conclusions

The main purpose of this study was to investigate the application of rotating ceramic filter discs and the potential of the CRD, as it is one of the few dynamic filtration systems available with ceramic membranes, for the treatment of oily model solutions and tank dewatering PW. On the basis of Figs. 3-8, it follows that for both, MF and UF membranes, the permeate flux was dependent on the rotational speed. The overall results indicate that ceramic MF and UF disc membranes used appear to be very effective in removing oil (> 99 %) and TOC (> 98 %) from tank dewatering produced water and model solutions. Additionally it could be observed that the rejection efficiency was independent of membrane rotational speed. Furthermore it could be shown that the increase of membrane rotational speed minimizes membrane fouling in a significant amount.

For studying the impact of the TMP and the feed characteristics on the fouling behavior of the membranes further investigations are already in progress.

Acknowledgement

The authors gratefully thank the Federal Ministry of Economics and Technology Germany for the financial support (FKZ: KF2829802RH1). Furthermore the authors would like to thank Dipl.-Ing. Martin Müller, German BP AG, Oil Refinery Emsland, Lingen, Germany for providing us with the tank dewatering produced water.

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