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Performance evaluation of membrane filtration for treatment of H₂S scavenging wastewater from offshore oil and gas production

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ABSTRACT

After removing H₂S from natural gas in offshore oil and gas installations, triazine-based spent scavenger solution remains as a wastewater stream, which in some cases is discharged untreated into the sea. Three nanofiltration (NF) membranes (NF270, NF99HF, and DL), as well as a reverse osmosis (RO) membrane (XLE), were used for TOC reduction from spent and unspent scavenger (SUS) wastewater. The NF270 membrane reduced the TOC of SUS wastewater (61.5 g/L) by 65% while having a superior permeate flux compared to the other studied membranes and thus was subjected to elaborated study. In particular, this membrane showed a separation between the unreacted/unspent scavenger triazine (1,3,5-tris(2-hydroxy-ethyl)hexahydro-s-triazine, HET) and the main spent scavenger reaction product (5-(2-hydroxyethyl)hexahydro-1,3,5-dithiazine, DTZ) as it removed HET by 71% versus zero removal of DTZ. DTZ is a polar and hydrophobic molecule and consequently passes readily through the membrane despite an expected removal value of 50% based on a simple size exclusion pore flow model. The NF270 permeate was then used as a feed for RO filtration by the XLE membrane to further reduce the TOC of effluent stream by 98% (1.1 g/L). Lastly, both membranes did not demonstrate remarkable fouling when they were tested in 24 h preliminary lab-scale fouling experiments with a constant-concentration feed. This paper is the first published study on SUS wastewater treatment using membrane technology proving its applicability for total organic carbon (TOC) reduction and simultaneously provides a promising route for further investigations on recovery of unreacted scavenger from spent scavenger compound.

1. Introduction

Although renewable energy is planned to replace fossil energy, oil and gas remain the primary energy sources worldwide [1]. Therefore, the deployment of advanced technologies is inevitable to decrease the environmental impacts associated with the oil and gas production process. Hydrogen sulfide, H₂S, is one of the main destructive impurities of natural gas causing problems such as corrosion in transport pipelines as well as toxicity [2,3]. Therefore, it is crucial to remove it from the gas (gas sweetening) to maximize the lifetime of facilities and fulfill environmental and safety regulations. Among the methods for removing H₂S from gas streams, the direct injection of chemicals, called H₂S scavengers, reacting with H₂S and converting it into substantially less harmful derivatives is one of the favorite options, especially in offshore production [4,5].

1,3,5-triazines are a category of chemicals frequently employed as H₂S scavengers [6,7]. The most commonly used triazine-based scavenger is 1,3,5-tri(2-hydroxyethyl)hexahydro-S-triazine (HET), which is

injected into the gas stream as a basic aqueous solution, absorbing and converting H₂S into organic species containing nitrogen and sulfur [4,8,9]. As shown in Fig. 1, two nitrogen atoms in HET are substituted in two consecutive steps with the sulfur atom of H₂S resulting in the formation of monoethanolamine (MEA) in each step as well as 5-(2-hydroxyethyl)hexahydro-1,3,5-dithiazine (DTZ) in the second step. There is also an intermediate product, 3,5-bis-(2-hydroxyethyl)hexahydro-1,3,5-thiadiazine (TDZ), formed in the first reaction, which is typically not observed in the final reaction product solution [10–12]. In practice, oil and gas operators inject the HET aqueous solution in excess than the stoichiometric requirement in order to ensure the maximum allowable limits on H₂S concentrations in the gas stream are met. Therefore, the resultant aqueous solution after the scavenging reaction is mainly composed of MEA and DTZ as spent scavenger chemicals as well as unspent (unreacted) HET.

In offshore oil and gas installations, the spent and unspent scavenger solution (simply called SUS solution in this study) is then separated from the gas stream in a gas–liquid separator followed by a further separation

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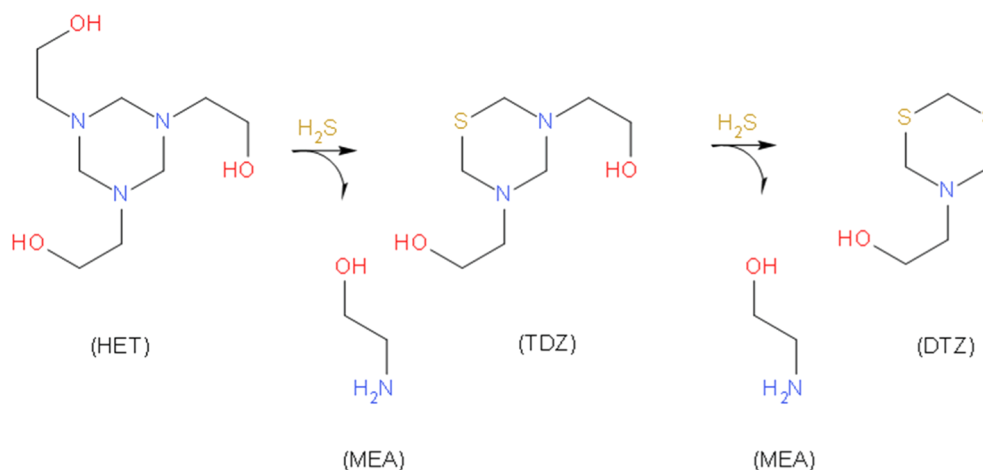


Fig. 1. H_2S scavenging reaction with HET and formation of MEA and DTZ.

step from the gas condensates, eventually leading to a wastewater stream. Due to the fouling and scaling potential of this stream caused by its alkaline pH and possible DTZ polymerization [8], the discharge of this wastewater stream into the sea without any treatment process is sometimes the only feasible solution for oil and gas operators. Although the volumetric flow rate of this wastewater stream is rather small compared to the produced water stream in oil and gas offshore platforms, its discharge may be of environmental concern due to the presence of unreacted triazine and scavenging products [5]. Based on OSPAR, the triazine-based H_2S scavenger solutions are not included in the list of chemicals posing little or no risk to the aquatic environment [13]. Moreover, the European Chemicals Agency (ECHA) defines MEA as a chemical that is “harmful to aquatic life with long-lasting effects” [14]. Therefore, it is essential to endeavor to treat the SUS wastewater before disposal to the aquatic environment in order to reduce its environmental impact factor.

Owing to its modular structure, membrane technology can be considered as a potential candidate for SUS solution treatment allowing retrofitting the existing offshore installation with respect to the fact that the SUS wastewater stream possesses a small size. Furthermore, membrane filtration does not require addition of chemical additives thus affords advantages regarding the quality of treated wastewater and results in a reduced environmental load. In addition, nanofiltration (NF) and reverse osmosis (RO) membrane filtrations have been found to be applicable for the removal of triazine-based pesticides in water treatment [15–19]. For instance, atrazine and prometryn as the most commonly used pesticides from the triazine family were removed from water by 72% and 81% using the Filmtech NF270 NF membrane in a research carried out by Plakas et. al. [17]. They also observed a removal value of 82% and 85% using the Filmtech XLE RO membrane for atrazine and prometryn, respectively [18]. Atrazine and prometryn have molecular weights of 215 Da and 241 Da, respectively, which are in line with the molecular weight of HET (219 Da). Madsen and Sogaard have also reported an atrazine rejection of 92% using the Alfa Laval NF99HF membrane in water treatment [19]. Therefore, a NF and RO membrane-based treatment concept is interesting to study further for management of the SUS wastewater stream and production of a cleaner effluent for discharge, while the generated retentate of reduced volume could be

treated using e.g. oxidation processes. A membrane-based approach may also offer the opportunity for selective separation of unspent HET from the DTZ main scavenging reaction product, as the two molecules are different in terms of molecular size, polarity as well as hydrophobicity. If partial recovery of HET is possible, it may be recycled to the H_2S scavenging injection point thus limiting the chemical consumption for the operation.

To the best of our knowledge, there is no published study on the treatment of SUS wastewater from offshore oil and gas production using membrane technology. Hence, in the present study, the applicability of NF/RO membranes for SUS wastewater treatment was investigated. The performance of four commercial NF and RO membranes was studied for the treatment of a real sample obtained from an offshore oil and gas installation in the North Sea. The best performing membrane in terms of TOC removal and water permeability was further utilized for rejection evaluation of the individual compounds HET, MEA and DTZ, thus the potential of membrane separation for recovery of unspent scavenger chemicals from spent scavenger reaction products was examined. A pore flow model was used for analysis of the governing separation mechanism. In addition, the permeate from the NF treatment was subjected to further treatment using RO and a combined treatment design consisting of both NF and RO processes was proposed to clean the wastewater and partially fractionate unspent and spent scavenger compounds. Finally, the fouling propensity of the SUS wastewater on the NF and RO membranes was studied during long-term experiments.

2. Materials and methods

2.1. Materials

1,3,5-tri(2-hydroxyethyl)hexahydro-S-triazine (HET, $\geq 95\%$, CAS number 4719–04-4) was purchased from Santa Cruz Biotechnology. Monoethanolamine (MEA, $\geq 99\%$) was supplied from Acros Organics and 5-(2-hydroxyethyl)hexahydro-1,3,5-dithiazine (DTZ, $\geq 98\%$, CAS number 88891–55-8) was obtained from Toronto Research Chemicals. Methyl heptadecanoate (MHD, $\geq 99.0\%$) from Sigma-Aldrich was used as internal standard for GC–MS. 2,2,4-trimethylpentane (isooctane, $\geq 99.5\%$) and 1-propanol ($\geq 99.5\%$) from VWR were used as internal standard for GC–FID. Dichloromethane (DCM, $\geq 99.8\%$), also from VWR, was used as liquid–liquid extraction solvent. All the reagents and solvents were of analytical grade and used without any further processing. All solutions were prepared with deionized water.

2.2. Spent and unspent scavenger solution

The spent and unspent scavenger (SUS) solution was obtained from

Table 1
Physical and chemical properties of SUS solution.

Physical properties		Chemical properties	
Density (kg/m^3)	1042	TOC (g/L)	61.5
pH	8.9	Triazine (HET, g/L)	82
Viscosity (cP)	2.9	Monoethanolamine (MEA, g/L)	40
Conductivity (mS/cm)	8.7	Dithiazine (DTZ, g/L)	41

Table 2
Chemical structure and properties of the target compounds.

	Triazine (HET)	Monoethanolamine (MEA)	Dithiazine (DTZ)
Chemical structure			
Molecular weight (Da)	219.28	61.08	165.3
Length (nm)	1.2773	0.7268	1.0338
Height (nm)	0.9589	0.5036	0.7017
Width (nm)	0.6171	0.4543	0.5523
MWd (nm)	0.3846	0.2392	0.3113
pK _a	–	9.45* [20]	–
log K _{ow}	–1.6** [21]	–1.3*** [22]	0.9** [23]
Dipole moment (D)	1.31	1.12	3.66

* Measured at 25 °C; ** Computed by XLogP3 3.0; *** Measured at 25 °C.

an offshore oil and gas production platform in the North Sea. The solution was subjected to in-house physical and chemical characterization as presented in Table 1. The pH of the solution was measured using a PHM210 MeterLab standard pH meter. A Metrohm model 744 pH Meter was used for measurement of conductivity. The analytical methods for chemical characterization are presented in the following section.

The TOC of the SUS wastewater is composed of three major components: HET, MEA and DTZ, which is to say the compounds of interest in this current study. The corresponding carbon content obtained from the concentrations of HET, MEA and DTZ is nearly 70 g/L, which is in line with the measured TOC value of 61.5 g/L. The chemical structure and properties of these molecules are seen in Table 2.

The geometry of the target compounds was estimated using the Gaussian software by a model described elsewhere [19]. In this model, the molecules are considered to have a rectangular parallelepiped shape where the distance between the two most distant atoms (considering Van der Waals radius) is defined as the length of the molecule. The molecule is then projected on a plane perpendicular to the length axis and the sides of an enclosing rectangle with minimum area forms the height and width of the molecule. The molecular width, MWd, is defined as a half of the square root of the area of the enclosing rectangle. The octanol/water partition coefficients (log K_{ow}) and the acid dissociation constant (pK_a) of MEA were found in the literature [20–23]. pK_a of HET and DTZ is not available. The dipole moments were calculated using the Avogadro software.

2.3. Membranes and characterization

Nanofiltration of the SUS wastewater was performed using three commercial flat-sheet thin film composite (TFC) membranes with polyamide as the active layer: Filmtech NF270 from DuPont (Dow), NF99HF from Alfa Laval and DL from SUEZ (GE). The reverse osmosis (RO) experiments were conducted using a flat-sheet low pressure reverse osmosis (LPRO) Filmtech XLE TFC membrane from DuPont (Dow). The membranes were characterized by the determination of pure water permeability, hydrophobicity/hydrophilicity using contact angle measurements, and membrane surface charge using zeta potential measurements.

The molecular weight cut-off (MWCO) of the membranes was reported by the manufacturers. The pure water permeability was determined from the slope of Milli-Q water permeate flux versus applied pressure (2–20 bar) curves. For contact angle measurement, a piece of

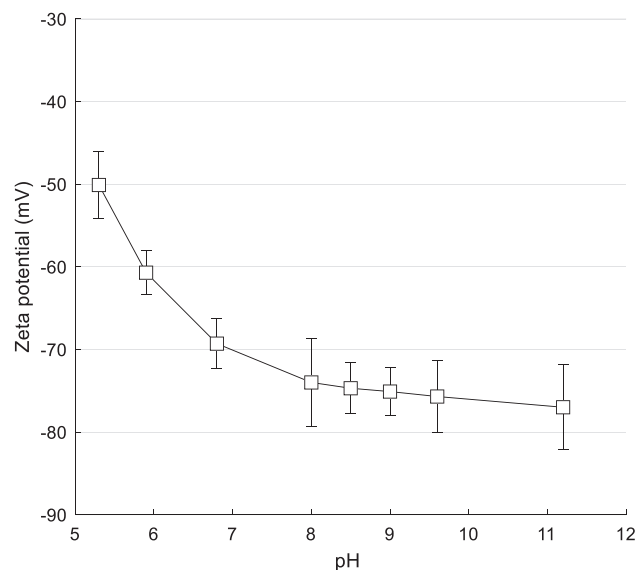


Fig. 2. Zeta potential of NF270 membrane as a function of pH value. Error bars represent the scatter of data within a 95% confidence interval obtained from triplicate measurements.

membrane was immersed in Milli-Q water overnight followed by drying in a desiccator before the measurements that were carried out using a KRUSS DSA100 instrument through the conventional sessile drop method with distilled water. The reported values are the average taken of at least three different measurements on different spots of the membrane piece. Zeta potential was also determined by Anton Paar SURPASS streaming potential analyzer in a 1.0 mM KCl solution at pH 8.5, which is the pH of SUS wastewater. Moreover, the Zeta potential of NF270 was measured for a range of different pH values of 5–11 as depicted in Fig. 2.

2.4. Membrane filtration setup

A cross-flow filtration FT17-50 unit (Armfield, The UK) was used to conduct membrane filtration tests. A schematic diagram is presented in Fig. 3a. Briefly, the bench-scale membrane unit consists of a flat-sheet membrane cell, a one-liter jacketed feed vessel connected to a chiller unit to adjust the temperature, a feed pump, as well as a make-up vessel and a peristaltic pump for addition of extra feed to the main feed vessel. The membrane cell (See Fig. 3b) consists of two basic parts made from 316 stainless steel. There is an inlet and outlet for the feed in the top part. Machined into the underside of the top part is a spiral-shaped flow channel which is 2.5 mm deep directing the feed flow across the membrane surface below. A flat-sheet membrane coupon is placed on a sintered support disc in the bottom part thereafter the bottom part is screwed to the top part. On the retentate outlet port of the membrane cell, there is a back pressure valve that is used to apply the filtration pressure. The system is controlled by a computer and all data is logged.

For each test, a fresh 90-mm diameter flat-sheet membrane disc (effective surface area of 63.6 cm²) was used. At first, the fresh membrane disc was rinsed with deionized water for 2 min to wash off the preservative layer from the membrane surface. Afterwards, the membrane was placed on a sintered support disc at the bottom part of membrane cell and the bottom part is screwed to the top part of the membrane cell. The feed vessel was filled with Milli-Q water to run the system at 10 bar (adjusted with valve V2) and 25 °C (adjusted with water circulation in the feed vessel jacket) for 1 h for pure water flux determination before filtration test. The permeate is collected in a container placed on a digital balance connected to a computer to monitor and log the changes in the weight of the permeate.

For filtration experiments with the SUS wastewater, 500 mL was transferred to the feed vessel and circulated for 30 min without pressure

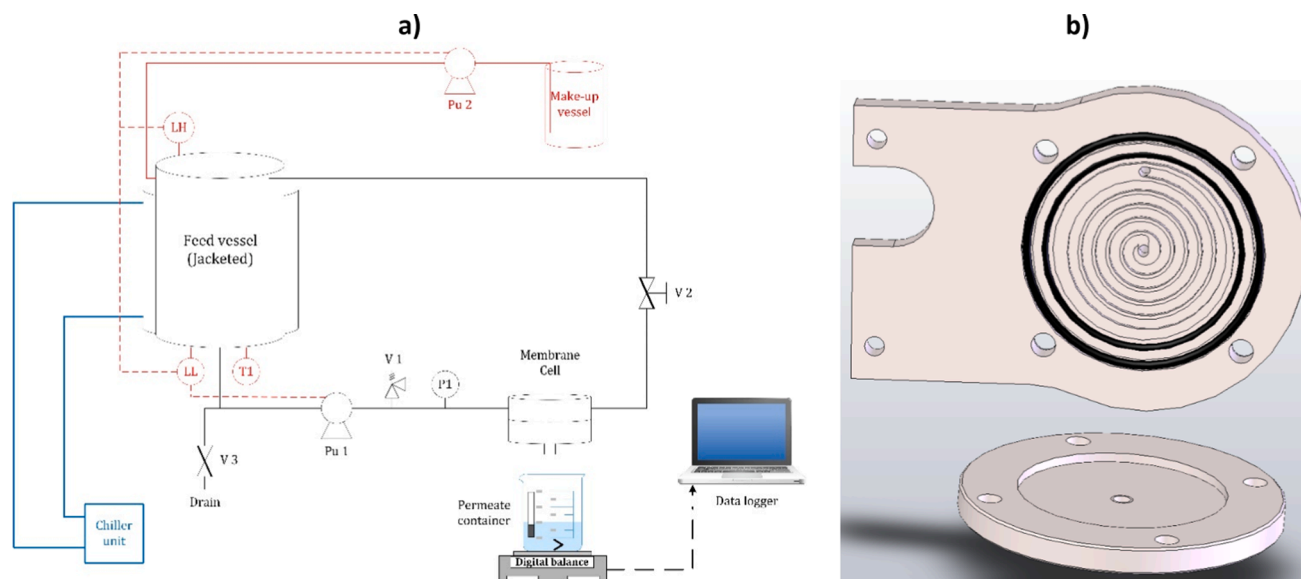


Fig. 3. a) Schematic diagram of FT17-50 cross-flow filtration unit, P1: Pressure sensor; Pu1: Feed pump; T1: Temperature sensor; LL: Low level sensor; LH: High level sensor; Pu2: Peristaltic pump for make-up solution. V1: Safety valve; V2: Back pressure valve; V3: Drain valve. The make-up vessel is used for addition of feed or permeate to the feed vessel using a peristaltic pump i.e. Pu 2. b) Schematic illustration of the flat-sheet membrane cell. Bottom part is shown without a sintered support disc and a flat-sheet membrane coupon. The membrane cell does not contain any spacer.

to allow the membrane surface to equilibrate with the feed solution and heat the system to 40 °C. After preconditioning, a sample was taken from the feed, and filtration was initiated at 30 bar and 40 °C and terminated when 250 mL of permeate was collected (50% recovery). The feed flow rate was adjusted at 100 L/h and the cross-flow velocity was set at 24.5 cm/s. The duration of the filtration experiments was in the range of 2 h to 10 h depending on the permeability of the tested membrane. In order to investigate possible pure water flux decline due to fouling, the pure water flux was determined again at 10 bar and 25 °C after the filtration experiment. All the filtration experiment sets were performed in duplicates.

The performance of the treatment was evaluated on TOC removal using equation (1):

$$TOC_{\text{removal}} = 1 - \frac{TOC_p}{\frac{1}{2} \times (TOC_F + TOC_R)} \quad (1)$$

TOC_p , TOC_F , and TOC_R are TOC of permeate, feed and retentate, respectively.

The same equation was applied for the calculation of the removal of the individual chemical compounds by substituting TOC with the concentrations of the solutes in the permeate, feed and retentate.

In addition, the permeate flux, J , was determined using equation (2) over the test period:

$$J = \frac{V_p}{A \cdot t} \quad (2)$$

where V_p is the volume of collected permeate, A is active membrane surface area, and t is the time of filtration test.

When a fresh membrane is used for filtration, some solutes tend to adsorb onto the surface of the membrane and this is more pronounced in the case of hydrophobic molecules. Thus, adsorption was also taken into consideration by calculation of the percentage of adsorbed species ($A\%$) using the following mass balance for the solutes:

$$A(\%) = \left(1 - \frac{C_p V_p + C_R V_R}{C_F V_F} \right) \times 100 \quad (3)$$

where, C_F , C_p , and C_R represent the concentration of solutes in feed, permeate and retentate, respectively while V_F , V_p , and V_R represent

volume of feed, permeate and retentate.

2.5. Analytical methods

Total organic carbon (TOC) was determined using a multi N/C 2100 TOC analyzer (Analytikjena multi N/C 2100, Germany). All the measurements were carried out in triplicate and the average values were reported. The relative standard deviations were always lower than 1%.

DTZ was quantitated by GC (PerkinElmer Clarus 680, USA) equipped with an Elite-5 column (30 m, 0.25 mm ID, 0.1 μm) coupled with MS (PerkinElmer Clarus SQ 8T, USA). Sample preparation was carried out by three consecutive liquid–liquid extractions with dichloromethane (DCM) and the reported final value was the summation of extracted DTZ mass in the three extracts divided by the volume of initial aqueous sample. The DTZ concentration was quantitated against an external standard calibration, using methyl heptadecanoate (MHD) as internal standard (IS) to account for inaccuracies between injections. The samples were diluted in pure DCM to be in the range of the calibration standards (i.e. 0.2–1.0 g/L) and were spiked with the IS (0.5 g/L). The R^2 of the calibration curve was always ≥ 0.995 . For all samples, 1 μL was injected. The oven temperature was initially 120 °C and was ramped at 25 °C/min to 175 °C, ramped at 45 °C/min to 225 °C and maintained at 225 °C for 3 min. The injector temperature was maintained at 300 °C and the flow rate of the helium carrier gas at 1.0 mL/min. The concentration of DTZ was determined by calculating the relative response factor (RRF) of DTZ with respect to the IS in the standard solutions as: $(C_{DTZ}/C_{IS}) = RRF \cdot (A_{DTZ}/A_{IS})$ where C_{DTZ} and C_{IS} are the concentrations and A_{DTZ} and A_{IS} are the chromatographic areas of DTZ and IS, respectively. The mass of DTZ in the DCM-extract was then calculated using the density of pure DCM at ambient temperature, which allowed to calculate the mass fraction of DTZ in the SUS feed.

HET and MEA were quantitated on a GC-FID (PerkinElmer Clarus 690, USA) equipped with a PerkinElmer Elite-5 Amine column (30 m, 0.32 mm, 1 μm). The quantitation was performed by an external calibration with five standards in the range of 1.0–7.5 g/L for HET and 0.50–3.75 g/L for MEA. Aqueous samples were diluted with deionized water in such of way that they lie within the abovementioned ranges and were spiked with a certain amount of 1-propanol as the internal standard (1.2 g/L). The injection volume was set at 1 μL, the injector temperature

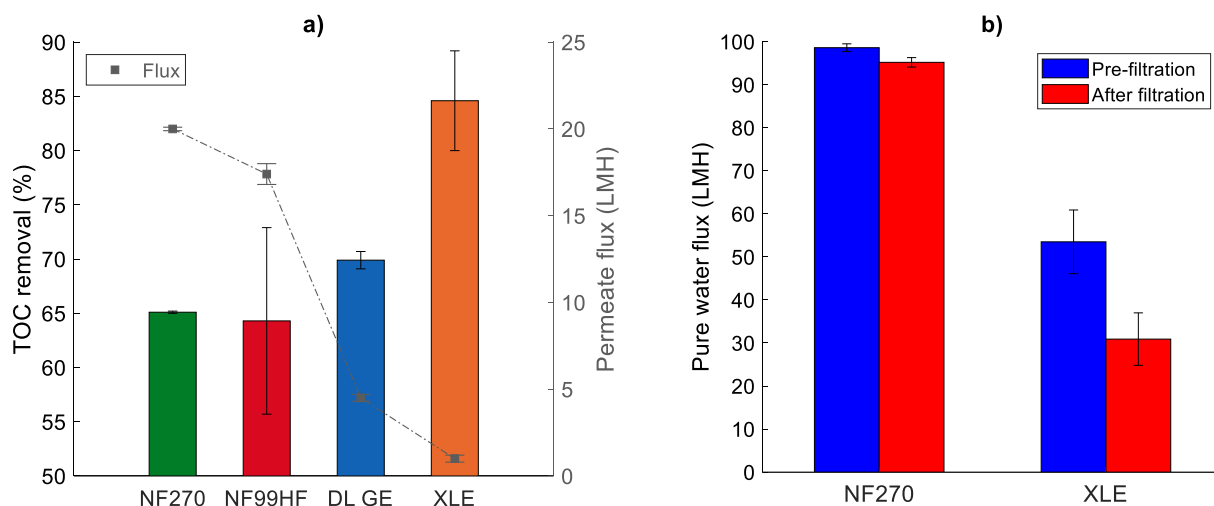


Fig. 4. a) TOC removal of the studied membrane for filtration of SUS wastewater at 30 bar, 40 °C and 50% recovery is presented as a bar chart on the left axis and permeate flux data is presented as a scatter chart on the right axis over the test period. b) pure water permeate flux before and after filtration tests with NF270 and XLE membranes at 10 bar, 25 °C and 50% recovery. E. Error bars represent the scatter of data within a 95% confidence interval obtained from duplicate experiments.

Table 3

Various properties of the commercial membranes used in this study. Variations represent the scatter of data within a 95% confidence interval obtained from triplicate measurements.

	NF270	NF99HF	DL	XLE
Molecular weight cut-off (MWCO) (Da)	~200–400	>300	~150–300	~100
Pure water permeability ($L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$)	15.2 ± 0.7	13.7 ± 0.5	8.2 ± 0.6	5.7 ± 0.6
Contact angle (°)	17.5 ± 3.7	20.7 ± 3.2	39 ± 4.5	75.8 ± 4.5
Zeta potential (mV) at pH 8.5	−75	−78	−21	−52

at 200 °C and the split ratio at 50:1. The oven temperature was kept at 75 °C for 1 min, ramped at 15 °C/min to 120 °C, ramped at 40 °C/min to 200 °C and maintained at 200 °C for 5 min. The temperature of the FID detector was also kept at 300 °C. The concentrations of HET and MEA were calculated by estimating their RRF in the same way as mentioned for the GC–MS quantitation of DTZ.

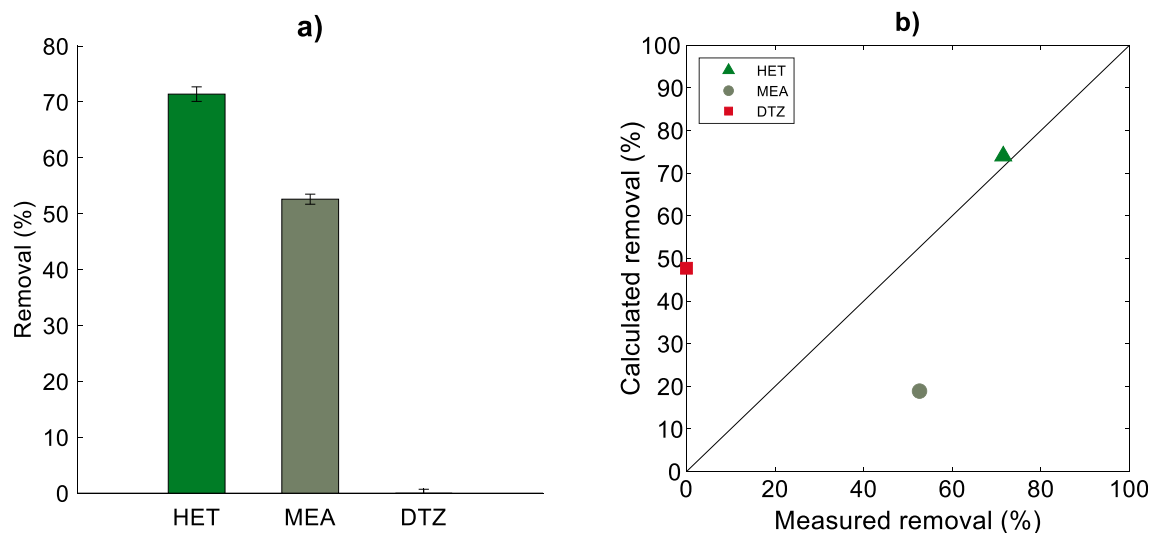


Fig. 5. a) Removal values of major compounds of SUS solution using NF270 membrane at 30 bar, 40 °C and 50% recovery. b) Fit for measured removal values modelled by purely steric pore flow model. Error bars represent the scatter of data within a 95% confidence interval obtained from duplicate experiments.

Table 4

The concentration data (g/L) of the NF tests with the NF270 membrane.

	HET			MEA			DTZ		
	C _F	C _P	C _R	C _F	C _P	C _R	C _F	C _P	C _R
NF270 #1	79.1	30.3	129.6	38.2	21.9	53.0	38.2	36.8	35.2
NF270 #2	85.2	30.6	132.1	39.1	22.3	53.6	43.9	43.2	39.6
Overall	82.2	30.4	130.9	38.6	22.1	53.3	41.1	40.0	37.4
Removal (%)	51.9			71.4			0		
Adsorption (%)	1.8			2.5			5.7		

Fig. 4a shows the TOC removal and permeate flux data obtained from duplicate membrane filtration tests. The data shows a reduction of TOC of the SUS wastewater by >60% by all tested membranes. Amongst the NF membranes, DL GE exhibited the highest TOC removal of 69.9% compared to 65.1% and 64.3% for NF270 and NF99HF, respectively. The TOC removals are negatively correlated to the molecular weight cut-off (MWCO) of the studied NF membranes (Table 3). The MWCO determines the distinction in the average pore size of the membranes, and the DL membrane from GE has an MWCO of 150–300 Dalton, which is lower than that of both NF99HF and NF270 membranes. This result is in line with the literature where the MWCO of membranes has been found to negatively correlate with the rejection of compounds by the membrane [26,27]. Considering the average permeate flux over the filtration period, the 20.1 LMH of NF270 outperforms the 17.4 LMH of NF99HF and 4.5 LMH of DL, in line with the characterized permeability data for the membranes as listed in Table 3.

The XLE LPRO membrane showed a considerably higher TOC removal of 84.6% in line with the lower 100 Da MWCO. As the XLE membrane has an MWCO < 200 Da, it is considered as a “tight” or “dense” membrane so that the passage of molecules across the membrane is impeded by steric hindrance and diffusion restriction [28]. The cost of the higher removal is the significantly lower permeate flux of 1.1 LMH over the filtration time, indicating that the permeate flux of this membrane might be profoundly influenced by fouling. This was investigated by comparing the pure water flux of the NF270 and XLE membrane before and after filtration (Fig. 4b). The plot shows that the pure water permeate flux declined nearly 45% after the filtration test with the XLE membrane whereas the NF270 membrane did not encounter a considerable flux decline after treatment, suggesting it is less affected by fouling. Although the XLE membrane showed superior performance in terms of TOC removal, the disposition to fouling and low permeate flux prevents it to be a candidate for stand-alone SUS wastewater treatment. Considering the overall performance i.e. both TOC removal and permeate flux, the NF270 membrane was selected as the best performing membrane and was subjected to further investigations.

Fig. 5a shows the removal of the HET, MEA and DTZ, i.e. the major components of the SUS wastewater, using the NF270 membrane. HET was removed by 71%, while the removal of the associated component MEA was found to be nearly 52%. Interestingly, DTZ, i.e. the sulfur-containing reaction product of the H₂S scavenging reaction, was not rejected from the SUS feed by the NF270 membrane. This suggests that NF270 is selective towards the retention of HET and MEA compared to DTZ. Therefore, the NF with this membrane can be used for separating unreacted scavenging chemicals (HET, MEA) from the spent scavenger (DTZ). The high removal of HET can be explained taking into account the molecular size of this molecule compared to the other two molecules as listed in Table 2. The HET molecule is the largest and, as a result, the rejection of the membrane is higher compared to MEA and DTZ. The markedly different rejection of MEA and DTZ cannot however be explained by the sizes of the molecules, since DTZ is larger than MEA but was not observed to be rejected at all by the NF270 membrane, whereas MEA was rejected by 50%. The degree to which the rejection can be explained by steric hindrance was investigated by using the pore flow model.

As presented in Fig. 5b, the steric hindrance mechanism explains

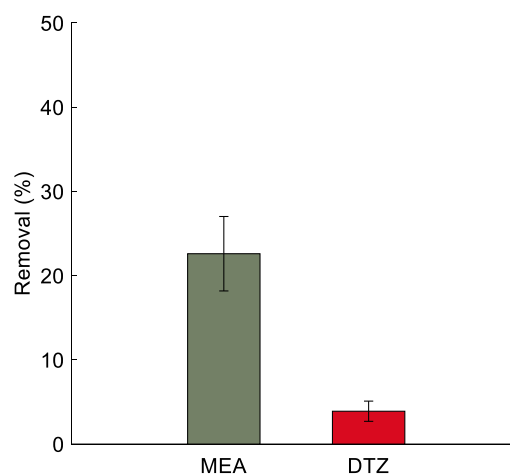


Fig. 6. Removal of MEA and DTZ by NF270 in single-solute solutions prepared with 10 g/L of MEA and 50 mg/L of DTZ in Milli-Q water. Error bars represent the scatter of data within a 95% confidence interval obtained from duplicate experiments.

nicely the measured removal of HET, but cannot alone explain the measured removal of MEA and DTZ. Based on the size parameters, the model predicts a higher removal of DTZ (approximately 50%) and simultaneously a lower removal of MEA (nearly 20%). This is because DTZ has a higher MWd value in comparison to MEA (Table 2), which has a more linear structure that more easily can pass through a cylindrical pore. Therefore, the model suggests that steric hindrance is not solely responsible for the rejection. A possible explanation and contributing factor to the undetectable removal of DTZ is the higher dipole moment of DTZ (3.66 D) compared to the other two molecules (See Table 2). It is previously discussed by other researchers that organic compounds with high dipole moment (>3 Debye) exhibit significantly lower retention than expected based on a purely size exclusion mechanism, where the molecule approaches the membrane pore randomly and Stokes radius represents the size of the solute [16,29–32]. When the polarity of a molecule is high, it can be directed to approach the membrane pores head-on due to attractive interaction between the molecule polar centers and fixed charged groups on the membrane surface [29]. This effect is more pronounced in the case of molecules with cylindrical shapes (large ratio of length to width [33]) [29]. The molecule with a dipole moment ends up in orienting towards the membrane pore and thus enters more easily into the pore and permeates across the membrane structure. As a result, the solute retention would be lower [29,30]. The effect of the dipole moment may be considerable for DTZ as this molecule has a large ratio of height to width and can be considered as a cylindrical molecule (See Table 2).

In addition, in contrast to HET and MEA, DTZ is a more hydrophobic compound, as indicated by the value of log K_{ow} (i.e. 0.9) (Table 2). It is well known that hydrophobic species tend to adsorb on the membrane surface and consequently diffuse through the membrane [34–36]. This is defined in the literature as the breakthrough effect where an adsorbed component after saturation of membrane can pass through the

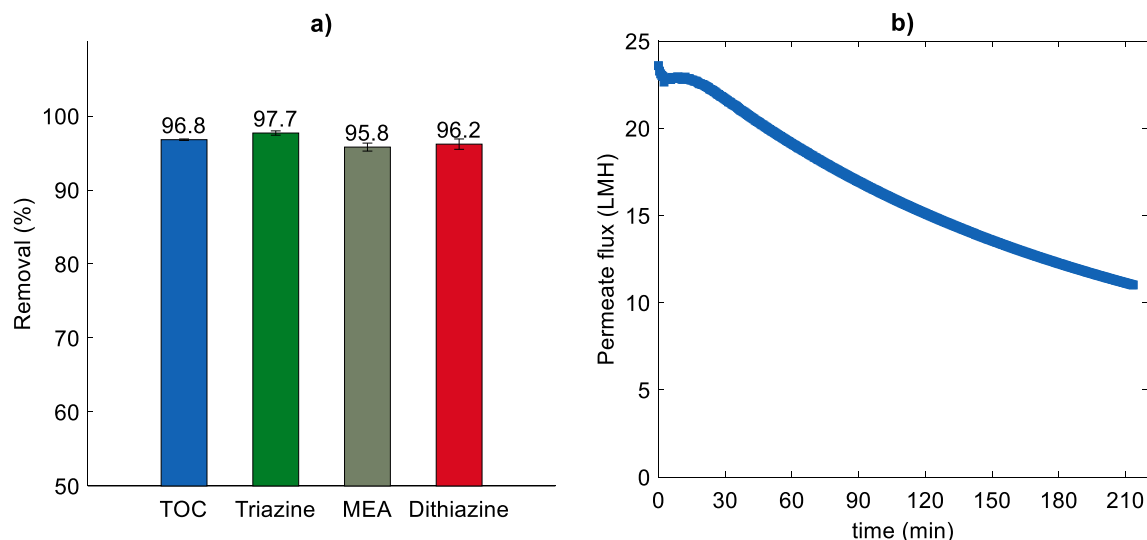


Fig. 7. a) TOC Removal and rejection values of major compounds for RO filtration tests using XLE membrane on the permeate collected from NF tests using NF270 membrane at 30 bar, 40 °C and 50% recovery. b) Permeate flux data of RO filtration using the XLE membrane. Error bars represent the scatter of data within a 95% confidence interval obtained from duplicate experiments.

membrane to the permeate side by diffusion and/or convection [26,37]. The adsorption data for the three solutes is presented in Table 4 where adsorption is more significant in the case of DTZ (i.e. 5.7 %) as a result of hydrophobicity. Therefore, the hydrophobicity of DTZ may also contribute to the permeance of this molecule through the membrane resulting in its zero removal.

In an attempt to elucidate the removal of MEA and DTZ from the SUS wastewater, two single-solute solutions with 10 g/L of MEA and 50 mg/L of DTZ were prepared in Milli-Q water and filtrated by NF270

membrane in duplicate. According to the obtained results shown in Fig. 6, DTZ was removed by 3.9% when a synthetic DTZ solution was used. This value is in line with the DTZ removal of nearly zero from the SUS wastewater suggesting that, regardless of solution matrix, DTZ molecules readily pass through the membrane due to the polarity and hydrophobicity of this compound. In contrast, the MEA removal was found to be 22.6% in a synthetic single-solute MEA solution, which is interestingly equal to the predicted value acquired by the pore flow model (See Fig. 5b). This similarity suggests that in the filtration of pure

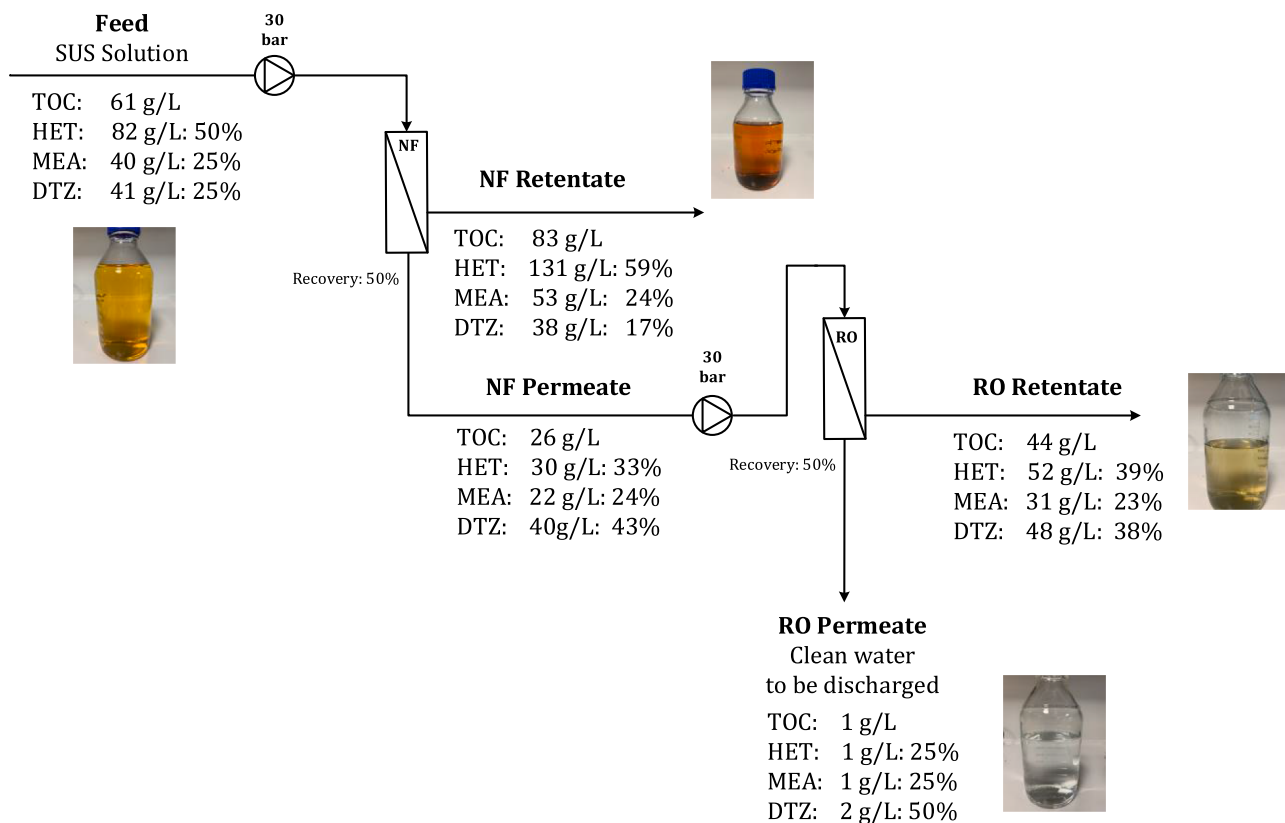


Fig. 8. Schematic illustration of combined NF and RO processes for treatment of SUS wastewater. The percentages indicated are the mass fraction of the specific compound in each particular stream on a water-free basis.

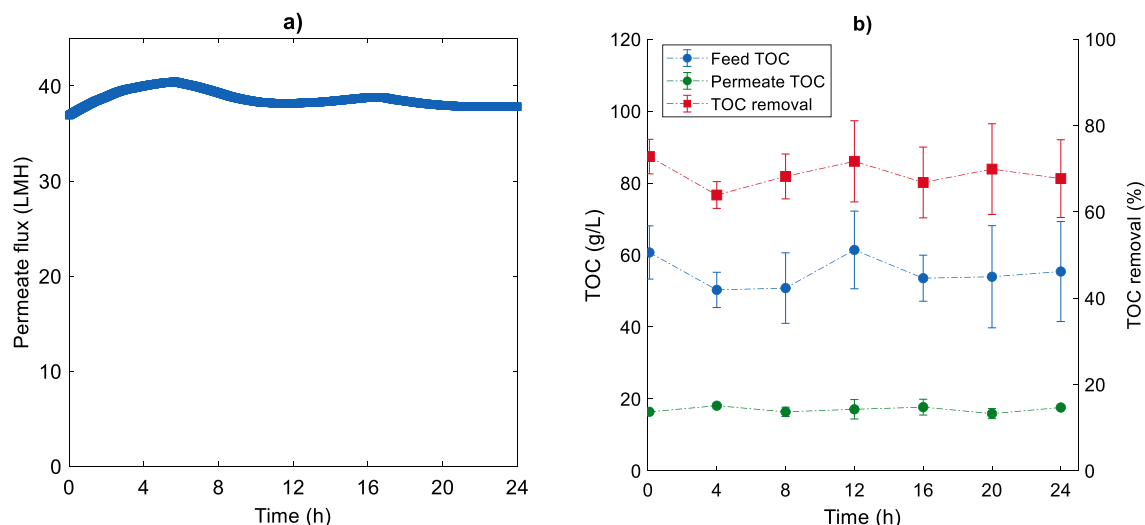


Fig. 9. a) Permeate flux measurement data for NF270 membrane at 30 bar and 40 °C. The permeate was recirculated back to the feed tank to keep the feed concentration at a constant level for 24 h. b) TOC of feed and permeate as well as TOC removal of samples taken in 4 h intervals during the fouling study test. Error bars represent the scatter of data within a 95% confidence interval obtained from duplicate experiments.

MEA solution with NF270 the governing separation mechanism is size exclusion. One possible explanation for the significant difference of MEA removal in SUS solution compared to pure MEA solution might be a possible interaction of MEA and HET in the SUS wastewater sample through intermolecular hydrogen bonding and formation of complexes of higher molecular sizes justifying higher measured removal values in SUS solution. It is shown in the literature that there is theoretically a potential for formation of $OH\cdots N$ and $NH\cdots O$ strong hydrogen bonds [38] and the presence of these functional groups in both HET and MEA molecules strengthens this hypothesis. However, such a complex formation merits further investigation in future studies.

3.2. Complete treatment of SUS wastewater

As shown in Fig. 4, the NF270 membrane was found to reduce the TOC of the SUS wastewater by nearly 65% meaning that the TOC of the solution was reduced from 61.5 g/L to 26 g/L in the obtained permeate. In this section, the possibility of further TOC reduction to lower TOC values is investigated. In order to further treat the SUS wastewater, the LPRO XLE membrane was used for filtration of the NF270 permeate.

Fig. 7a presents the data for filtration tests using the XLE membrane on the NF270 permeate. The results demonstrate that the XLE membrane was capable of reducing the TOC by approximately 97% generating a permeated effluent of 1.1 g/L TOC. The three major compounds of SUS wastewater were rejected > 96% (97.7% for HET, 96.2% for DTZ and 95.8% for MEA).

The permeate flux of XLE membrane throughout the filtration test is seen in Fig. 7b. The initial flux of 24 LMH declined to 11 LMH, when 50% of permeate was obtained from the feed solution and the feed was concentrated. The flux decline is not necessarily a result of fouling. The feed concentration increases in a batch filtration mode, which leads to increased osmotic pressure of the feed and consequently a decreased driving force provided by the constant TMP of 30 bar. A detailed investigation of the membrane behavior in terms of permeate flux during longer time operation was also carried out, as discussed in Section 3.3.

The performance of the LPRO XLE membrane suggests that a combination of NF and RO processes can effectively reduce the content of chemicals in the SUS wastewater to be discharged. An overview of the combined processes is illustrated in Fig. 8. The produced retentate and permeate samples of the membrane filtration steps are also visually distinct. The collected permeate from the RO process with a TOC of 1.1

g/L is transparent, while the NF retentate with a TOC level of 83 g/L is even darker than the initial SUS feed solution reflecting the higher concentration of chemicals in this stream.

Another promising aspect of treating the SUS wastewater by membrane separation is the potential for recovery of scavenger chemicals (HET and MEA) from the spent scavenger compound (DTZ). As seen from the composition of the NF retentate, the fraction of DTZ declined from 25% in the SUS feed to 17%, whereas the fraction of HET in this stream increased to 59%, from 50% in the initial feed. This means that the chemicals with scavenging properties are more concentrated in the produced retentate compared to DTZ and it has been partially separated from HET and MEA. In order to recycle this stream, to benefit from the scavenging potential of the separated HET, research in membranes with a higher selectivity in the separation of HET and DTZ is required, but the results shown in this paper are promising. DTZ is known to be a cause of fouling due to polymerization [8], so to prevent problems due to the build up of this reaction product, it needs to be reduced further in the recycle stream. The produced retentate from the RO unit is enriched in terms of DTZ and can be handled through a secondary treatment method, e.g. hydrothermal oxidation [39]. In this way, the capacity need of the hydrothermal oxidation reaction is reduced resulting in a smaller reactor size matching offshore requirements. Therefore, membrane filtration can be considered as a promising option for SUS wastewater treatment and simultaneously recovery of unreacted chemicals from the waste stream, which is in favor of the H_2S scavenging economy.

3.3. Fouling performance of membrane filtration

A grave concern associated with membrane technology is fouling. In this study, a set of experiments were designed to investigate whether the used NF and RO membranes encountered fouling during a longer time operation (24 h). Similar to the previous tests, the cross-flow velocity was set at 24.5 cm/s while the feed flow rate was 100 L/h. In these experiments, the permeate was recycled back to the feed vessel with an initial feed volume of 2 L to resemble a continuous filtration process where the concentration of the feed contacting the membrane remains constant. Samples were then taken from both feed and permeate in 4 h intervals to track their TOC as well as the TOC removal throughout the duration of the test.

The flux measurement data for the NF270 membrane is shown in Fig. 9a and shows that the permeate flux maintained at a constant level

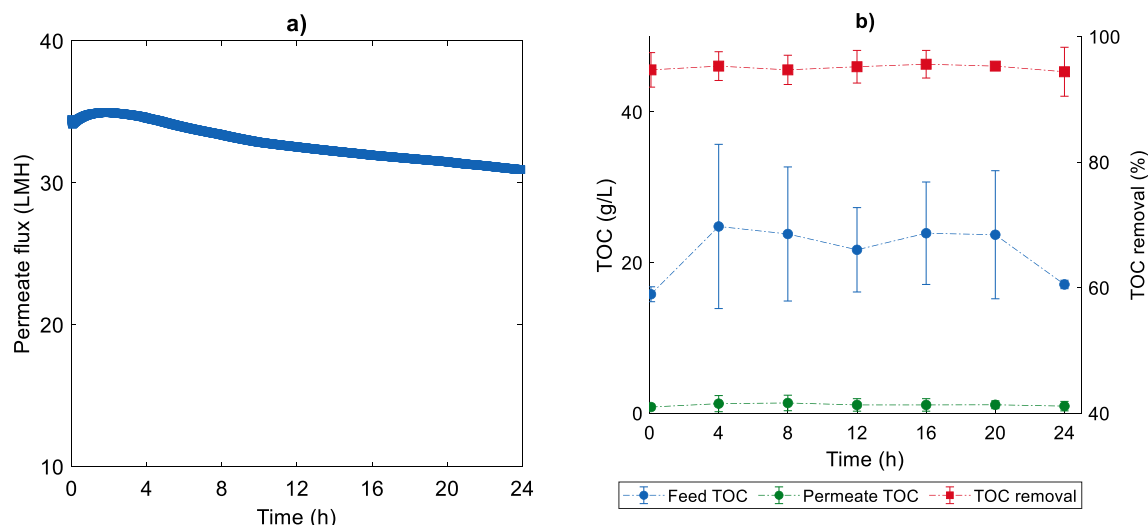


Fig. 10. a) Permeate flux measurement data for XLE RO membrane at 30 bar and 40 °C. The permeate was recirculated back to the feed tank to keep the feed concentration at a constant level for 24 h. b) TOC of feed and permeate as well as TOC removal of samples taken in 4 h intervals during the fouling study test. Error bars represent the scatter of data within a 95% confidence interval obtained from duplicate experiments.

of 37 to 40 LMH during 24 h. No notable flux decline was observed, suggesting that in a bench-scale 24 h test the flat-sheet NF270 membrane did not foul with the SUS wastewater components. Using the data presented in Fig. 9b, one can control the possibility of maintaining the TOC values constant through permeate recirculation. It demonstrates that the TOC of the feed remained within the range of 50–60 g/L as a result of permeate recirculation, as intended in this experiment. The permeate TOC was also maintained at a constant level of 16.5–18.1 g/L showing that the filtration proceeded continuously with a sustained quality. Therefore, the resultant TOC removal also remained within the range of 64% to 72%. This data demonstrates that the permeate recirculation was successfully performed resulting in a constant level of feed TOC and TOC removal.

As shown in Fig. 10a, the permeate flux of the XLE flat-sheet RO membrane (the feed was the permeate from the NF270 filtration) declined slightly from approximately 34 to 31 LMH during the 24 h. Compared to the initial flux, this permeate flux decline is still below the threshold value of 20%, which is set to determine if fouling is significant for an RO membrane [40,41]. Comparing to NF270, this data reveals that the XLE membrane is more prone to fouling, maybe due to a higher fraction of DTZ in the NF permeate than the initial SUS feed solution (See Fig. 8). Therefore, as DTZ polymerization is the main cause of fouling in oil and gas installation facilities [8] its concentration is a limiting factor that must be taken into account before designing the membrane unit. Similar to the NF, the TOC of the feed and permeate and the TOC removal were maintained in narrow intervals throughout the experiments showing that the permeate recirculation was capable of adjusting the feed TOC to a constant level (See Fig. 10b).

It is worthwhile to mention that this was an introductory bench-scale fouling study performed for the SUS wastewater. For a solid conclusion regarding the fouling propensity of NF/RO membranes for this specific matrix, a comprehensive fouling study requires to be carried out where the membrane configuration and flow regime is more representative of the real-world industrial conditions.

4. Conclusion

The paper presents a proof-of-concept study of using membrane filtration for treatment of a particular spent and unspent H₂S scavenger wastewater generated during offshore oil and gas production, when triazine-based chemicals are used for removal of H₂S from the natural gas stream. TOC removal and permeate flux was determined for three

commercial NF membranes and one LPRO membrane on a real wastewater sample. From a high initial feed TOC of 61 g/L, the NF270 membrane showed a TOC removal of 65% at a permeate recovery of 50% with a permeate flux of 20.1 LMH and was selected as the best performing NF membrane. The XLE LPRO membrane tested under the same conditions showed a TOC removal of 84.6% while showing a poor permeate flux of 1.1 LMH. Using NF270 permeate as feed for the XLE membrane (sequential treatment), TOC in the RO permeate was reduced to 1.1 g/L. Long-term (24 h) preliminary lab-scale experiments using flux measurements did not show signs of fouling, but further investigation in larger scale (e.g. pilot-scale) is required to study this further in more realistic conditions. With respect to individual compound rejection, the NF270 membrane showed the capability of separating the excess unspent scavenger compound, HET (rejection 71%), from the spent scavenging reaction product, DTZ (zero rejection). On top of the environmental benefits of reducing the discharge of environmentally harmful chemicals to marine recipients, this finding demonstrates a potential recovery of the unspent scavenger chemical for re-injection in the H₂S scavenging process and can serve as a baseline for future studies.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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