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Noor, Imtisal-e-; Martin, Andrew; Dahl, Olli

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# District heating driven membrane distillation for advanced flue gas condensate treatment in combined heat and power plants

Imtisal-e – Noor <sup>a, b, \*</sup>, Andrew Martin <sup>a</sup>, Olli Dahl <sup>b</sup>

<sup>a</sup> Department of Energy Technology, KTH Royal Institute of Technology, Stockholm, Sweden
<sup>b</sup> Department of Bioproducts and Biosystems, Aalto University, Espoo, Finland

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# ABSTRACT

This investigation involves recent work on developing membrane distillation (MD) as a novel separation technology for flue gas condensate treatment in combined heat and power (CHP) applications. Flue gas condensate samples were obtained from municipal solid waste and bio-fuel fired CHP facilities and were tested in laboratory-scale air gap MD equipment. Separation efficiencies and other water quality parameters were measured, and the outcomes show that high-quality clean condensate can be recovered, i.e., conductivity <5 mS/m; total organic carbon <2 ppm; total hardness <0.15 °dH; pH ~7.5; and turbidity <1 FNU. Strict discharge limits for cadmium could not be achieved in all trials. This aspect is examined in further detail with respect to potential mechanisms attributed to the non-ideal separation of contaminants below the parts per billion limit. Beyond this, an industrial scale district heating driven membrane distillation system was designed and analyzed. The estimated annual thermal energy demand was 88 GWh for treating 500,000 m<sup>3</sup> of flue gas condensate per year, with an expected clean condensate cost of around 2.6 \$/m<sup>3</sup>.

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# 1. Introduction

The European Union aims to become a frontrunner in the execution of the 2030 Agenda. This framework gives new impetus to member-state environmental regulatory bodies in realizing sustainable development goals. With respect to sustainable development goals 6 (Clean Water and Sanitation), 9 (Industry, Innovation, and Infrastructure) and 12 (Responsible Consumption & Production), the concerned authorities have further tightened up discharge limits of industrial effluent. These actions are starting to have an effect on existing industrial operations as more and more conventional separation techniques cannot achieve these discharge limits. Moreover, industries are encouraged to adopt a zero liquid discharge approach for improved water management, in particular for process water reuse. In combined heat and power (CHP) plants, the emphasis is placed on improved water treatment systems for various effluents to ensure high environmental performance, including recovery for boiler make-up water. Flue gas condensate is a potential source of process water, although the stream is heavily

E-mail address: ieno@kth.se (I.-e. Noor).

contaminated with heavy metals, particulates, salts and solids. Many CHP facilities have considered various membrane separation technologies for flue gas condensate treatment, including reverse osmosis (RO), micro-filtration (MF), ultrafiltration (UF), and nanofiltration (NF) (Kylhammar, 2018; Nobel et al., 2014; Uotila, 2015). These approaches are highly recognized and generally effective, but there is still a need for a cost-efficient system with enhanced water purity. Additionally, there are some technical issues involved in these technologies, including bio-fouling, scaling, disposal of concentrated solutions and upper pressurization limits. CHP facilities have also reported a high concentration of some contaminants in the RO treated flue gas condensate, necessitating the use of alternative technologies for meeting discharge limits.

In this setting, membrane distillation (MD) is one of the promising membrane separation processes which have unique characteristic for water purification (Lawson and Lloyd, 1997). Numerous studies have been published about its successful applications in several industrial applications (excluding desalination) such as pharmaceutical (Woldemariam et al., 2016), nano-electronics (Noor et al, 2019, 2020a), chemicals (Si et al, 2019, 2020), food (Julian et al., 2020) and textile (Leaper et al., 2019). MD uses a microporous hydrophobic membrane that allows only volatiles, i.e., water vapors and lighter alcohols, to permeate through the membrane.

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<sup>\*</sup> Corresponding author. Department of Energy Technology, KTH Royal Institute of Technology, Stockholm, Sweden.

The main driving force is the vapor pressure gradient across the membrane, which is produced due to the temperature difference involved in the process. The MD operation technically occurs in three steps: evaporation, filtration and condensation. The involved phase separation in the MD process is based on the vapor-liquid equilibrium, where the latent heat of evaporation drives the change in phase from liquid to vapor (Alkhudhiri et al., 2012). The methods to condense vapors on the membrane-permeate interface at the cold side divide the MD concept into four primary types of configurations: (1) direct contact membrane distillation (DCMD), where vapors condense at the cold side of the membrane which is in direct contact with the cold stream; (2) air gap membrane distillation (AGMD), where vapors condense at the cold side of the membrane which is in contact with stagnant air-cooled by a cold plate; (3) sweeping gas membrane distillation (SGMD), where an inert gas sweeps vapors at the cold side of the membrane; and (4)vacuum membrane distillation (VMD) where relatively low pressure (vacuum) is maintained at the cold side of the membrane and vapors condense at external condenser (Wang and Chung, 2015). Compared to other membrane and thermal separation methods, the MD process can theoretically reach complete rejection of nonvolatiles, operates at relatively mild operating temperature, i.e., below 100 °C and at atmospheric pressure, remains unaffected by the change in feed concentration, requires less pretreatment and accountable for lower capital costs (Alkhudhiri et al., 2012; Alklaibi and Lior, 2005; Camacho et al., 2013). Since MD is a heat-driven separation process, the required thermal energy to operate the MD system can be achieved from low-grade heat sources, i.e., district heating (DH) (Kullab and Martin, 2011: Woldemariam et al., 2016), solar energy (Asim et al., 2016), geothermal energy (Sarbatly and Chiam, 2013) and waste energy (Noor et al., 2019). To achieve optimum results, the desirable feed to coolant temperature difference may vary between 25 and 60 °C. Therefore, the heat sources and sinks required for the effective operation of MD technology is suitable for CHP integration.

Previously some preliminary experimental studies were performed where DCMD and AGMD were employed to explore the potential of this technology in the power industry. Ali et al. (2018) presented the performance of lab-scale DCMD for flue gas desulfurization wastewater treatment in terms of transmembrane flux and specific energy demand. They reported that flat PE membranes outperformed capillary hollow-fiber PP membranes. It was also found that the concentration of all non-volatiles was reduced by 99.5%. Conidi et al. (2018) also investigated a laboratory-scale RO-DCMD integrated process for flue gas desulfurization wastewater treatment in order to minimize the salt content and reuse purified water. In the study, RO-retentate was further concentrated by DCMD (PE membrane-flat module), and the outcomes show that up to 94% of the water was recovered using an integrated approach. Moreover, the non-volatiles were removed successfully, and the conductivity of the MD permeate stream was 80 µS/cm. Yu et al. (2013) evaluated bench-scale DCMD for concentrating cooling tower blowdown (CTBD) in the power industry. The results show that 99.5% of salts were rejected, and the water recovery extent was increased from 68% to 87% when the antiscalant was used. Chuanfeng and Martin (2005) and Kullab and Martin (2011) conducted pre-studies on pilot scale AGMD flat module design for flue gas condensate cleaning in biofuel fired CHP plants. It was observed that particles and heavy metals in flue gas condensate could be separated successfully; however, ammonium could not be removed completely. It is noteworthy that Kullab and Martin (2011) mentioned the district heating driven MD system in the study. Still, the approach to address DH supply – MD demand heat balance and economic analysis of such integrations were not reported. Later, Fortkamp et al. (2015) investigated the flue gas condensate cleaning while using a similar AGMD system and found that ammonium separation issues were still present, which had put the researcher's focus to address this point further.

Recently Noor et al. (2020b) performed an experimental study on MD based process for advanced flue gas condensate treatment in municipal solid waste (MSW) incineration CHP plants. High separation efficiencies were achieved with both laboratory and pilotscale AGMD equipments when acid neutralization was considered for ammonium removal. Preliminary estimates indicated that sufficient thermal energy was available to drive the MD process in a cost-efficient manner. These results suggest the need for follow-on studies to examine MD treatment for other types of flue gas condensate, and to elucidate the practical application of MD technology in this setting. The present investigation focuses on these aspects through experimental trials using flue gas condensate obtained from a CHP facility employing both waste and biofuel. Separation efficiency and quality of treated condensate are considered with particular attention paid to heavy metal separation. Moreover, different thermal integration options are investigated in a technoeconomic analysis.

# 2. Methodology

This section contains the description of the considered CHP facilities, experimental methods for flue gas condensate treatment, and approach for performing techno-economic system analysis of industrial-scale MD system.

# 2.1. Bristaverket CHP plants

The Stockholm Exergi Bristaverket facilities consist of a biofuel fired CHP plant (Brista 1) and waste to energy CHP plant (Brista 2): Brista 1 supplies 760 GWh of heat and 290 GWh of electricity annually while consuming 350000 tons of wood chips, whereas Brista 2 supplies 500 GWh of heat and 140 GWh of electricity annually by combusting 240000 tons of municipal and industrial waste. The process overview of the Bristaverket CHP facility is shown in Fig. 1 (emphasis placed on flue gas condensate treatment system).

The flue gas released from the biofuel-fired boiler (B1) is sent to the electrostatic precipitator (ESP) in order to remove dust and larger particles, prior to its introduction in the acid quench tank for achieving desirable pH of 3.1–3.5. Later, the acidified flue gas is condensed, and the flue gas condensate is then disinfected with sodium hypochlorite (NaOCI) and sent directly to the rotating filter. Here, the clean flue gas is guided to the stack, whereas flue gas condensate is sent for treatment. This stream is first passed through MF shaking screens for removing suspended solids and is then exposed to ultraviolet (UV) light for microorganism control. The UV-filtered flue gas condensate is further sent to UF filters and bag filters for additional treatment and later passes through RO filters. RO permeate is sent to the clean condensate collection tank. The typical volumetric flow rate of flue gas condensate from this line is around 40 m<sup>3</sup>/h.

The flue gas released from waste fired boiler (B2) is treated with a process partially similar to the one installed at Stockholm Exergi's Högdalen facility (Noor et al., 2020b). The flue gas condensate is sent to MF bag filters and follows an analogous route of flue gas condensate treatment as for Brista 1 (exception: UV-filters are not considered in this case). Additionally, RO permeate is passed through an ion exchanger before entering into a condensate collection tank to eliminate mercury (Hg). From Brista 2, around 20 m<sup>3</sup>/h of flue gas condensate is added into the collection tank. Later, based on requirements, the mixed clean condensate is directed towards various avenues: 1) released to the recipient after



Fig. 1. Flow Diagram of Stockholm Exergi Bristaverket CHP facilities.

pH adjustment; 2) sent for further treatment as boiler make-up water; 3) sent to ash slag in B2.

In Brista 1, mercury (Hg) levels after electrostatic precipitators are higher than the release limit (<0.2 ppb). Moreover, the plant clearly has issues with biofouling after MF and UF circulation. Brista 2 also shows a high amount of microorganisms in the raw condensate.

# 2.2. Experimental methods

Fig. 2 shows the schematic diagram of the Xzero lab-scale prototype, which has been considered in this study for performing experimental tests. Flue gas condensate samples totaling 40 L were collected from Bristaverket during 2019. The experiments were



**Fig. 2.** Xzero air gap membrane distillation bench-scale setup with specifications of its main components (Dolfe, 2018).

performed at Xzero AB, Bromma, Stockholm facility in two phases: phase A, when flue gas condensate from MSW CHP plant is tested, and phase B when flue gas condensate from biofuel CHP plant is considered. Keeping in view the limitation of MD for removal of volatile contaminants (ammonia) at elevated temperatures, acid neutralization is necessary (Noor et al, 2019, 2020b); however, the extent of pretreatment was needed to determine. Therefore for each phase, the experiments were performed in two steps: (1) no pretreatment and (2) pretreatment with the addition of 0.5 mL of 40% sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) per L of flue gas condensate samples. Fig. 3 presents the schematic diagram of the proposed flue gas condensate cleaning system. Each test utilized 10 L of feedstock, and experiments were performed at a feed temperature of 70 °C and cold-water temperature of 18 °C. The operating flow rate of the MD feed was 4.9 L/min, and the cold-water flow rate was 6.8 L/min. Each test ran for 3 h, and the initial measurements were taken after achieving a steady state. The feed, concentrate and permeate samples of 1 L each were collected. The collected samples were sent to an external laboratory for physicochemical analyses to determine the water quality. These analyses were performed to measure the following water properties:

• Chemical composition, and metallic and ionic concentration



Fig. 3. Schematic diagram of proposed flue gas condensate cleaning system.

- Total dissolved solids for quantifying the dissolved combined content of all organic and inorganic substances in water
- Total hardness for measuring the concentration of dissolved earth metals, calcium and magnesium
- Total organic carbon (TOC) for estimating the concentration of organic carbon
- Chemical oxygen demand (COD) for evaluating oxygen concentration required for chemical oxidation of all organic compounds into their inorganic end products
- Turbidity for measuring the relative clarity of the water that is caused by the presence of organic/inorganic suspended particles
- Electrical conductivity for accessing the concentration of charged ionic species in the water
- pH for determining the acidity or basicity of water and alkalinity for defining the buffer capacity of the water to resist pH changes

Table 1 presents the selected methods and their limits of quantitation (LOQ) for the considered analyses.

# 2.3. Techno-economic assessment

Concerning the technical evaluation, the main challenge in operating an industrial scale MD system is addressing its relatively high thermal energy demand. For CHP plants, one option is to utilize district heating lines for operating the MD system to perform the advanced flue gas condensate treatment. For exploring different approaches of DH-MD integration, the critical parameter is DH temperature. Based on seasonal variance, DH temperature levels and flue gas condensate produced in CHP plants differ considerably. During summer times, DH temperatures drop, and flue gas condensate flow rate decreases accordingly and vice versa in winters. Usually, the temperatures of DH supply lines differ from 70 °C in the summer to 100 °C in the winter, whereas the DH return line temperature ranges from 35 to 65 °C correspondingly (Gadd and Werner, 2014; Werner, 2017). Similarly, the amount of flue gas condensate produced varies from 10  $m^3/h - 25 m^3/h$  for small CHP plants up to 60  $\text{m}^3/\text{h}$  –125  $\text{m}^3/\text{h}$  for large CHP plants due to seasonal variance. Previously, Woldemariam et al. (2016) presented different approaches for integrating DH-MD to treat pharmaceutical residues. These approaches were mainly based on 1) integration of DH supply line or return line as a heat source and cooling water from an external source as a heat sink and 2) consideration of the DH supply line as a heat source and DH return line as a heat sink. In this study, we have adopted similar but modified approaches to make them well-fitted for flue gas condensate treatment in CHP plants. In case 1, the DH supply line of temperature 95 °C is used as a heat source, while the DH return line of 40 °C is

#### Table 1

Physico-chemical analysis methods for water quality determination.

considered as a heat sink for the MD system. In case 2, the DH return line of 60 °C is used for heating the MD feed whereas, an external cooling source of 10 °C, i.e., municipal water or water from the cooling tower, is chosen as a heat sink. Fig. 4 shows the proposed semi-batch process configurations to integrate the DH-MD system for cleaning flue gas condensate in CHP facilities while employing acid neutralization as the only prospective pretreatment technique. The capacity of proposed industrial-scale DH driven MD system was considered 500,000 m<sup>3</sup>/year (matching the yearly maximum amount released from Bristaverket CHP facility), as a base scenario.

Depending upon MD feed flow rate and specific thermal energy demand (Noor et al., 2020b), the required membrane area along with the total number of MD modules were estimated. For enhanced energy efficiency, every two MD modules (a cascade) were configured in series where the hot feed outlet (retentate from the earlier module) was used as feed for the second MD module. Heat exchanger areas were calculated based on heat source/sink flow rates and temperatures. The number of pumps was also estimated accordingly. In order to increase the extent of water recovery, the design allowed the concentrate to recycle back to the MD system until the concentration limit was reached then the



Fig. 4. Flow diagram of the proposed industrial-scale DH-MD integrated system; (a) scheme for Case 1 (DH supply line as a heat source and DH return line as a heat sink), (b) scheme for Case 2 (DH return line as a heat source and other cooling sources as a heat sink).

Methods	Measurement (LOQ)
ICP-MS	Na (0.1 ppm), K (0.1 ppm), Ca (0.05 ppm), Fe (0.005 ppm), Mg (0.1 ppm), Mn (0.0005 ppm), Al (0.01 ppm), As (0.0002 ppm), Ba (0.001 ppm), Pb
	(0.0005 ppm), Cd (0.0001 ppm), Co (0.00005 ppm), Cu (0.0005 ppm), Cr (0.0005 ppm), Mo (0.0002 ppm), Ni (0.0005 ppm), V (0.0002 ppm), Zn
	(0.002 ppm)
CV-AFS	Hg (0.0001 ppm)
IC-EC	Cl <sup>-</sup> (0.1 ppm)
Nefelometry	SO <sub>4</sub> <sup>-2</sup> (1 ppm), turbidity (0.1 FNU)
Spectrophotometry	NH <sup>+</sup> <sub>4</sub> (0.01 ppm), NH <sub>3</sub> –N (0.01 ppm), COD <sub>cr</sub> (20 ppm), TOC (2 ppm)
Gravimetry	TDS
By analysis of	Total Hardness
Ca + Mg	
Potentiometry	pH (2)
Conductometry	Conductivity (2 mS/m)
Titration	Alkalinity (2 ppm HCO <sub>3</sub> )

concentrate was purged from the MD system to fulfill the safety regulations. The purged concentrate was considered to be further directed to the acid quench tank/scrubber (depending upon the facilities), in line with procedures for the existing MF, UF and RO systems.

Based on the technical requirements, the process economy was also estimated where the ultimate criterion is unit clean condensate cost. The unit clean condensate cost was calculated considering the annual capital and operational expenses of the DH-MD integrated system, with data obtained from the literature and communication with manufacturers. Key equipment that were responsible for the bulk of the total capital investment includes MD modules, heat exchangers, pumps, tanks, sensors, etc. When calculating annual capital cost, it was assumed that plant life would be 20 years; the membrane should be replaced after five years; interest rate remains 5%; and retrofitting and insurance accounts for 4% and 5% of the purchased equipment cost, respectively. The OPMEX contributors are thermal energy cost (77 \$/MWh) (Werner, 2016), cooling water cost (0.02 \$/m<sup>3</sup> of total cooling water) (Turton et al., 2009), labor cost (0.03 \$/m<sup>3</sup>) (Kesieme et al., 2013), electricity price (0.09 \$/kWh) (Eurostat, 2019), service and maintenance cost (0.033 \$/m<sup>3</sup>) (Drioli et al., 2006) and annual membrane replacement cost (15% of total membrane cost per year) (Medesol Report, 2010).

# 3. Results and discussion

This section presents the performance of MD based process for treating the flue gas condensate samples released from MSW and bio-fuel fired CHP facilities. This is followed by process design and techno-economic assessment of district heating integrated industrial-scale MD system.

## 3.1. Separation efficiency

#### 3.1.1. MSW CHP facility

Table 2 presents the concentration analysis of different contaminants in flue gas condensate (with/without pretreatment) samples from MSW CHP facility, MD purified permeates and resulting concentrates. Moreover, the concentration of reference permeate (RO and ion exchange-treated permeate/condensate from the particular CHP facility) sample is also mentioned and compared. In phase A, for step 1 (no pretreatment): the raw flue gas condensate samples and resulting concentrate and permeate samples are symbolized with S1-A, C1-A and D1-A, respectively and for step 2 (with pretreatment): the neutralized flue gas condensate samples and associated concentrate and permeate samples are shown as S2-A\*, C2-A\* and D2-A\*, respectively.

The outcomes of the chemical analysis of D1-A show that most of the metals have concentrations below detection limits, except for Fe, Pb and Cd, but their concentration is very low with a maximum of 8 ppb. The ionic concentration of D1-A was guite low for ammonium (1.9 ppm) and ammoniacal-nitrogen (1.5 ppm), and Cl<sup>-</sup>,  $F^-$  and  $SO_4^{-2}$  were not even detectable in permeate. The conductivity was found <2 mS/m, and the pH was 7.2. The sample D1-A was also compared with reference permeate, and the outcomes show that D1-A has a higher concentration of Cd and Pb while reference permeate has a higher concentration of Na and As; however, the values of metallic concentration of these contaminants are quite low in both permeate samples. The analysis of D2-A\* presents that ammonium and ammoniacal-nitrogen were not detectable. The concentration of most metals in D2-A\* were below detection limits and comparable to values in reference permeate, except for Cd, Pb and Cu.

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acceptable quality, and neutralization is not required in this specific case, contrary to what has been demonstrated in previous studies (Noor et al., 2020b). The reason might be the better quality of Brista MSW plant flue gas condensate compared to Högdalen MSW-CHP plant flue gas condensate. It is noteworthy that both plants follow partially different flue gas cleaning procedures, and both have similar (MSW waste) but variable boiler fuel. Concerning their corresponding permeates, it was found that Cd and Pb among 20 analyzed metals are detected in both MD permeates and have a comparable concentration when no pretreatment was considered. Other quality parameters show the better quality of MD permeate of Brista flue gas condensate samples might be due to relatively cleaner feed quality. Similarly, when neutralization was performed, the MD permeate of Brista pretreated flue gas condensate samples has lower contamination compared to MD permeate of Högdalen pretreated flue gas condensate samples. It was also noticed that the reference permeate of the Brista MSW-CHP plant has a higher quality standard, and the release limits are also considerably tighter, especially for Cd.

# 3.1.2. Biofuel CHP facility

Table 3 summarizes the chemical analysis of test samples, which include flue gas condensate (untreated/treated) samples from biofuel-fired CHP facility, and associated MD concentrate and MD treated permeates. Moreover, the quality of reference permeate (RO-treated permeate from the respective CHP facility) is presented. In phase B, for step 1 (no pretreatment): the raw flue gas condensate samples and related concentrate and permeate samples are denoted with S1–B, C1–B and D1-B, respectively. However, for step 2 (with pretreatment): the neutralized flue gas condensate sample and related concentrate and permeate samples are presented with notations S2–B\*, C2–B\* and D2-B\*, respectively.

Comparing samples S1-A and S1–B, it was found that the metallic concentration of S1-A was reasonably higher than S1–B, especially for Na, K, Ca, Mg, Ba, Pb, Cu and Hg. Similarly, sample S1-A has a higher concentration of  $Cl^-$  and  $SO_4^{-2}$ ; however, S1–B has a considerably higher level of ammonium and ammoniacal-nitrogen compared to S1-A. The other quality parameters, including TOC, TDS, total hardness, conductivity, alkalinity and pH, are also determined. It was found out sample S1-A has higher values of these parameters; therefore, the quality of S1–B seems improved. The observed difference in the quality of both samples is certainly associated with boiler feed and age, and flue gas cleaning processing.

The chemical analysis of permeate D1-B shows that among 20 metals, only Cd was detectable; however, the concentration was sufficiently low. The ionic concentration of Cl<sup>-</sup>, F<sup>-</sup> and SO<sub>4</sub><sup>-2</sup> in D1-B was below detection limits; however, removal of ammonium and ammoniacal-nitrogen was not satisfactory (35%), and the concentrations were quite high in D1-B, i.e., 12 and 9 ppm respectively. The pH was 8.8, and the conductivity was 6.1 mS/m, which is considered acceptable. The sample D1-B was also compared with reference permeate. It can be observed that Fe and Hg concentration in reference permeate is higher than D1-B, whereas Cd concentration in D1-B is slightly higher compared to reference permeate. Moreover, the concentration of ammonium and ammoniacal-nitrogen is quite high in D1-B as compared to reference permeate. Therefore, pretreatment was needed to be considered in terms of neutralization, which helped to reduce ammonium and ammoniacal-nitrogen concentration up to 0.045 and 0.035 ppm, respectively, in D2-B\*. However, it leads to other difficulties such as the higher concentration of Pb, Cd and Hg compared to D1-B and reference permeate.

Since the separation is not fully completed, it is possible that some contaminants (i.e., elemental Hg, Pb and Cd and/or their associated compounds) became adsorbed and/or permeated

The comparison of D1-A and D2-A\* shows that D1-A has

#### Table 2

Separation efficiency of the MD module in case of MSW CHP facility.

	Linite	C1 A	C1 A	D1 4	C2 A*	C2 A*	D2 4*	Defense es norma este
	Units	51-A	CI-A	DI-A	52-A*	CZ-A*	DZ-A*	Reference permeate
Na	ppm	27	23	<0.1	23	25	0.33	3.37
K	ppm	2.1	1.9	<0.1	2	2.2	<0.1	<0.4
Ca	ppm	25	23	<0.05	25	27	0.13	<0.2
Fe	ppm	< 0.005	0.0082	0.0055	0.014	0.014	< 0.005	<0.01
Mg	ppm	3.7	3.3	<0.1	3.4	3.7	<0.1	<0.2
Mn	ppm	< 0.0005	0.0013	< 0.0005	0.0024	0.0019	< 0.0005	<0.0009
Al	ppm	< 0.01	0.021	<0.01	0.17	0.14	< 0.01	<0.01
As	ppm	< 0.0002	< 0.0002	< 0.0002	0.00021	0.00021	< 0.0002	0.000507
Ba	ppm	0.0052	0.0068	< 0.001	0.0077	0.0081	< 0.001	<0.001
Pb	ppm	0.0034	0.0019	0.00082	0.0079	0.0076	0.0017	<0.0005
Cd	ppm	0.00054	0.00029	0.00037	0.00061	0.0004	0.00032	<0.00005
Со	ppm	< 0.00005	0.000063	< 0.00005	0.000093	0.000055	< 0.00005	<0.0002
Cu	ppm	0.0033	0.033	< 0.0005	0.14	0.12	0.0017	<0.001
Cr	ppm	< 0.0005	< 0.0005	< 0.0005	0.00096	0.00073	< 0.0005	<0.0009
Hg	ppm	0.00059	0.00039	< 0.0001	0.00029	0.00026	<0.0001	<0.00002
Mo	ppm	0.0006	0.00069	< 0.0002	0.00038	0.00039	< 0.0002	<0.0005
Ni	ppm	0.0012	0.018	< 0.0005	0.058	0.051	< 0.0005	<0.0006
Tl	ppm	<0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001	<0.0001	<0.0001
V	ppm	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	<0.0002
Zn	ppm	0.0089	0.044	< 0.002	0.28	0.23	< 0.002	<0.004
Cl <sup>-</sup>	ppm	14	13	<0.1	15	16	<0.1	-
$SO_4^{-2}$	ppm	37	36	<1.0	360	400	<1.0	-
F <sup></sup>	ppm	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	-
$NH_4^+$	ppm	1.1	0.57	1.9	0.46	0.64	<0.01	1.21
NH <sub>3</sub> -N	ppm	0.84	0.44	1.5	0.36	0.5	<0.01	0.943
NO <sub>3</sub> —N	ppm	0.22	0.21	<0.1	0.24	0.26	<0.1	-
TOC	ppm	2.7	3.6	<2.0	4.5	4.3	<2.0	-
COD <sub>Cr</sub>	ppm	<20	<20	<20	<20	<20	<20	-
TDS	g/l	0.14	0.1	<0.02	0.45	0.39	< 0.02	-
Total Hardness	°dH	4.4	3.9	<0.15	4.2	4.7	<0.15	-
Turbidity	FNU	<0.1	1.9	0.13	1.2	0.63	<0.1	-
pH		7.9	8.3	7.2	2.4	2.4	4.8	5.5
Conductivity	mS/m	28	25	<2.0	210	240	<2.0	22
Alkalinity	ppm HCO <sub>3</sub>	89	77	7.1	<2.0	<2.0	<2.0	-

'<' indicates a value below the respective detection limit of the measuring equipment. Note that samples of MD treated permeate and RO treated permeates are sent to different external laboratories. Therefore, different detection limits can be observed. '-' indicates the unavailable values.

through the membrane. (Penetration of contaminants in the liquid phase has not been considered.) The key responsible parameters are temperature and pH levels, which have affected the concentration of contaminants in feed, concentrate and permeate samples. Some of the potential mechanisms attributed to the non-ideal separation of contaminants are mentioned below.

- Higher temperature corresponds to enhanced solubility of many salts as well as leads to an increase in the vapor pressure of some elements or their compounds. For example, Hg in elemental form can be volatile (high vapor pressure) at higher temperatures; however, the solubility of HgCl<sub>2</sub> (abundant in flue gas condensate) increases with temperature, which might be the reason for the presence of Hg both in concentrate and permeate (Gaffney and Marley, 2014). The solubility of CdCl<sub>2</sub> also increases with elevated temperature (Lerner, 1996). The other contaminants, for instance, PbCl<sub>2</sub> is also highly volatile and sparingly soluble; however, its solubility also increases with temperature.
- The pH of the solution affects the surface charge of the polymeric (PTFE) membrane, as well as the speciation of elements in the solution.
  - o At low pH, electrostatic interactions are introduced among charged solutes and membrane surface, which defines the retention/permeation of the contaminants; co-ions are repelled while counter-ions are attracted to the membrane surface (An et al., 2016; Kheriji et al., 2015; López-García et al., 2018). That might be another reason for the presence of contaminants in permeates.

- o Speciation of metals based on the chemical/thermodynamic stability of those elements and their corresponding compound in the solution at certain pH can be described by the Pourbaix diagram (Pourbaix, 1974). For example, oxides of Hg, Cd and Pb can only be present at least when pH is above 4 (Lyon, 2010; Pourbaix, 1974; Spyropoulou et al., 2018). Therefore, the neutralized flue gas condensate sample might not include the aforementioned oxides. Moreover, a higher concentration of NH<sub>3</sub> is present at elevated pH levels, whereas lower pH directs the equilibrium towards NH<sup>+</sup><sub>4</sub> in aqueous solution (Emerson et al., 1975).
- Additionally, Le Chatelier's principle is accountable for the solubility of different salts based on pH and their associated anions (Le Chatelier, 1884). This means acid neutralized flue gas condensate dramatically increases the solubility of essentially all sparingly soluble salts whose anion is the conjugate base of a weak acid. In contrast, variation in pH level has little to no effect on the solubility of salts whose anion is the conjugate base of a stronger weak acid or a strong acid, respectively (e.g., chlorides and sulfates).

Thus higher temperatures and lower pH affected the performance of the MD systems considerably due to the change of characteristics of the contaminants as well as of the MD membrane.

# 3.1.3. Combined scenario

At the Bristaverket plant, flue gas condensates from both facilities are mixed; therefore, the final reference permeate can be the

#### Table 3

Separation efficiency of the MD module in case of bio-fired CHP facility.

Fille gas condensa	te from bioluei fire	d boller line						
	Units	S1-B	C1–B	D1-B	S2-B*	C2-B*	D2-B*	Reference permeate
Na	ppm	0.18	4.4	<0.1	1.2	0.96	0.22	<0.5
K	ppm	0.15	0.12	<0.1	0.24	0.31	<0.1	<0.4
Ca	ppm	0.25	0.19	< 0.05	0.73	0.77	0.085	<0.2
Fe	ppm	< 0.005	0.005	< 0.005	0.012	0.018	< 0.005	0.0119
Mg	ppm	<0.1	<0.1	<0.1	0.12	<0.1	<0.10	<0.2
Mn	ppm	0.008	0.0051	< 0.0005	0.013	0.019	< 0.0005	< 0.0009
Al	ppm	<0.01	<0.01	<0.01	0.027	0.065	< 0.010	<0.01
As	ppm	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0005
Ba	ppm	0.0015	0.0011	< 0.001	0.0025	0.0031	< 0.001	< 0.001
Pb	ppm	0.001	0.0013	< 0.0005	0.0042	0.0062	0.00095	< 0.0005
Cd	ppm	0.00039	0.00025	0.00011	0.00053	0.00068	0.00018	< 0.00005
Со	ppm	< 0.00005	< 0.00005	< 0.00005	< 0.00005	< 0.00005	< 0.00005	< 0.0002
Cu	ppm	< 0.0005	0.0017	< 0.0005	0.011	0.025	0.00078	< 0.001
Cr	ppm	< 0.0005	< 0.0005	< 0.0005	0.00073	0.0009	< 0.0005	<0.0009
Hg	ppm	0.0016	0.0013	< 0.0001	0.0025	0.0028	0.00099	0.00018
Mo	ppm	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0005
Ni	ppm	< 0.0005	0.0012	< 0.0005	0.015	0.023	< 0.0005	< 0.0006
Tl	ppm	< 0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001	< 0.0001
V	ppm	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002	< 0.0002
Zn	ppm	< 0.002	0.024	< 0.002	0.022	0.03	< 0.002	< 0.004
Cl-	ppm	7.5	6.3	<0.10	8.6	11	0.14	-
$SO_{4}^{-2}$	ppm	<1.0	3.1	<1.0	430	510	<1.0	-
F <sup></sup>	ppm	0.32	0.33	<0.2	<0.2	<0.2	<0.20	-
$NH_4^+$	ppm	36	35	12	37	45	0.045	1.59
NH <sub>3</sub> -N	ppm	28	27	9.5	29	35	0.035	1.24
NO <sub>3</sub> -N	ppm	1.3	<0.10	<0.1	7.4	2.8	<0.1	-
TOC	ppm	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0	-
COD <sub>Cr</sub>	ppm	<20	<20	<20	<20	<20	<20	-
TDS	g/l	0.02	0.13	< 0.02	0.45	0.4	< 0.02	-
Total Hardness	°dH	<0.15	<0.15	<0.15	<0.15	<0.15	<0.15	-
Turbidity	FNU	0.13	0.35	<0.1	0.71	0.16	<0.1	-
pH		7.6	8.6	8.8	2.3	2.2	3.7	5.2
Conductivity	mS/m	26	23	6.1	290	330	10	19
Alkalinity	ppm HCO <sub>3</sub>	77	69	41	<2.0	<2.0	<2.0	-

'<' indicates a value below the respective detection limit of the measuring equipment. Note that samples of MD treated permeate and RO treated permeates are sent to different external laboratories. Therefore, different detection limits can be observed. '-' indicates the unavailable values.

mixture and denoted as HR, whereas release limits for the recipient can be stated as RL. The comparison of D1-A and D2-B\* with mixture HR tells that Na, Hg (in case of D1-A), ammonium and ammoniacal-nitrogen have a higher concentration in RO treated condensate; however, MD treated permeates have a relatively higher concentration of Pb, Hg (in case of D2-B\*) and Cd. The final check of comparing the values of MD permeates with RL tells that D1-A and D2-B\* have partially fulfilled the RL requirement.

In order to improve MD-treated permeates quality, the mixing of permeates is considered as one of the options. The outcomes of D1-B and D2-B\* shows that when they are mixed in the ratio of 4 to 1, respectively, the quality of the mixed permeate (M1) is much closer to the desired outcome. Moreover, when M1 is mixed with D1-A (this mixture denoted as M12), the MD-treated mixed permeate fulfills the RL requirement, except for Cd. Usage of cation resin in the existing ion-exchanger can be a potential method to handle Cd ions concentration before disposal or reuse of the clean condensate (Pyrzynska, 2019). Table 4 shows the comparison of RO and MD treated permeates with the release limits of the Brista plant. (Since the metallic concentration of K, Mg, Mn, Al, As, Ba, Co, Cr, Mo, Ni, Tl, V, Zn was below the detection limits in phase A and B; therefore, mixing follows that their concentration in M12 is also below detection limits as well as below release limits. For that reason, these species are not shown in Table 4.)

# 3.2. Industrial scale integrated MD system

Based on the reported performance of the semi-commercial

pilot plant (Noor et al., 2020b), the outlet temperatures of MD feed and coolant streams, average transmembrane flux and specific thermal energy demand were estimated and shown in Table 5. Using the mentioned information, the total required and net required thermal power was calculated. The results show that for case 1, the total thermal power demand was approx. 64 MW, however, 83% of heat can be recovered by the district heating return line, and resultantly the net thermal power consumption was only 11 MW. In contrast, in case 2, net thermal power consumption was similar to total thermal power demand, i.e., 29 MW since the heat recovery is not considered when the heat sink is municipal water or cooling water.

Based on the information and outcomes mentioned above, the number of MD modules and required membrane area was calculated. It was found that since the transmembrane flux is higher at elevated feed temperature and higher temperature difference between feed and coolant inlet temperatures, in case 1, the required number of MD modules is lower than the required amount in case 2. Similarly, 1.6 times more heat exchanger area is needed in case 2, where a higher total MD feed flow rate is required to achieve the annual target capacity of the MD system. The number of pumps and their capacities are also estimated according to the flow rates and configuration of the system. Keeping in view the experimental results, approx. 15% of the total flue gas condensate needs to be pretreated; therefore, the amount of pretreatment chemicals is calculated accordingly. The required cooling water amount was based on the considered configuration and the total MD flow rate. In case 1, when district heating return is considered as a heat sink,

#### Table 4

Characteristics of RO/MD treated permeates and release limits for Bristaverket CHP plant.

		Mixed reference permeate	Release limits	MD treated pe	rmeates	Mixture of M1 and D1-B
		HR	RL	M1 (4:1)	D1-A	M12 (1:1)
Na	ppm	8.01	-	0.124	<0.1	0.112
Ca	ppm	<0.4	-	0.057	<0.05	0.0535
Fe	ppm	<0.01	-	< 0.005	0.0055	0.00525
Pb	ppm	<0.0005	0.0015	0.00059	0.00082	0.000705
Cd	ppm	<0.00005	0.00008	0.000124	0.00037	0.000247
Cu	ppm	<0.001	0.002	0.000556	< 0.0005	0.000528
Hg	ppm	0.00021	0.0002	0.000278	< 0.0001	0.000189
Cl-	ppm	-	-	0.108	<0.1	0.104
$SO_4^{-2}$	ppm	<1.0	-	<1.0	<1.0	<1.0
F <sup></sup>	ppm	<0.2	-	<0.2	<0.2	<0.2
$NH_4^+$	ppm	4.75	-	9.609	1.9	5.7545
NH <sub>3</sub> -N	ppm	3.69	5	7.607	1.5	4.5535
NO <sub>3</sub> -N	ppm	<0.1	-	<0.1	<0.1	<0.1
TOC	ppm	-	-	<2.0	<2.0	<2.0
COD <sub>Cr</sub>	ppm	-	-	<20	<20	<20
TDS	g/l	-	-	< 0.02	< 0.02	<0.02
Total Hardness	°dH	-	-	<0.15	<0.15	<0.15
Turbidity	FNU	-	-	<0.1	0.13	0.115
рН		-	-	7.78	7.2	7.49
Conductivity	mS/m	-	-	6.88	<2.0	4.44
Alkalinity	ppm HCO <sub>3</sub>	-	-	33.2	7.1	20.15

'<' indicates a value below the respective detection limit of the measuring equipment. Note that samples of MD treated permeate and RO treated permeates are sent to different external laboratories. Therefore, different detection limits can be observed. '-' indicates the unavailable values.

#### Table 5

Estimated parameters for considered cases of integrated MD system of capacity  $500,000 \text{ m}^3/\text{year}$ .

Parameters	Case 1		Case 2	
	MD1	MD2	MD1	MD2
MD feed inlet temperature	90 °C	80.9 °C	55 °C	52.75 °C
Feed outlet/concentrate temperature	80.9 °C	74.8 °C	52.75 °C	50.81 °C
Coolant inlet temperature	45 °C	52.8 °C	15 °C	17.9 °C
Coolant outlet temperature	52.8 °C	58.3 °C	17.29 °C	19.12 °C
Average transmembrane Flux	4.78 kg/	m²/h	2.91 kg/m	1²/h
Total specific heat demand	1020 kWh/m <sup>3</sup>		464 kWh/m <sup>3</sup>	
Total thermal power demand	63.75 MW		29 MW	
Net thermal power demand	11 MW		29 MW	

cooling water for condensation purposes was not required. However, in case 2, an enormous amount of cooling water is needed to satisfy the operation's requirement. The outcomes of the equipment design are presented in Table 6.

# 3.3. Process economy of anticipated industrial-scale system

Fig. 5 presents a summary of the process economy of the proposed DH driven industrial-scale MD systems. Using the design outcomes, the total equipment cost was estimated. The comparison of capital investment in both cases shows that choosing the



Fig. 5. Process economy for proposed DH-MD integrated industrial-scale systems.

configuration of case 1 over case 2 can result in a reduction of more than 4 M\$. It was found that in both cases, MD modules (~70% of the total equipment cost) and heat exchangers (~17% of the total equipment cost) are the leading components of the capital investment. This cost can be reduced mainly by introducing more efficient MD modules in terms of heat and mass transfer, which would indirectly affect the heat exchanger area and pump capacity,

#### Table 6

Design specification of industrial-scale integrated MD system for both cases.

Components	Specification				
	Case 1	Case 2			
MD Modules	N = 5678	N = 9326			
Membranes	$Area = 13058 m^2$	$Area = 21450 m^2$			
Heat Exchangers	$N = 2$ , Heat Transfer area $= 15821 m^2$	N = 2, Heat Transfer area = 25988 m <sup>2</sup>			
Pumps	N = 4, Capacity 3500 m <sup>3</sup> /h	N = 4, Capacity 5595 m <sup>3</sup> /h			
	N = 2, Capacity 65 m <sup>3</sup> /h	N = 2, Capacity 65 m <sup>3</sup> /h			
Raw material	Amount	Amount			
40% concentrated sulfuric acid	37.5 m <sup>3</sup> /year	37.5 m <sup>3</sup> /year			
Cooling Water	-	45 Mm <sup>3</sup> /year			

and associated costs owing to lower MD feed flow rate for achieving the target product yield. In these cases, the other equipment including water tanks, pumps, process control equipment and membranes were accountable for ~12-13% of the total equipment cost. The annual normalized capital cost was also calculated while considering the assumptions mentioned in section 2.3 and found that 1.53  $m^3$  is accountable in case 1. whereas it is 2.27  $m^3$  in case 2. The share of utilities (thermal energy and/or cooling water) is 98% of annual operating and maintenance costs in both cases. However, the normalized thermal energy cost is higher  $(35.8 \text{ }/\text{m}^3)$ for case 2 when the heat is not recovered, and additional expenses of cooling water are needed, whereas, in case 1, the normalized thermal energy cost is 13.66  $/m^3$ . This represents the importance of choosing different heat source and sink options. The other operating expenses include electricity cost, membrane replacement cost, maintenance and service cost, labor cost, chemicals cost and concentrate management cost. For case 1, the annual operating cost sums up to ~7 M\$ whereas, this number increases to ~19 M\$ for case 2. The results show the economic superiority of case 1, where the estimated unit clean condensate cost can be approx. 15  $/m^3$ . In waste-fired CHP plants, case 1 configuration would incur an added fuel (MSW) volume, which in turn leads to the economic benefit of MSW power plant and, at a larger scale, to environmental benefit. Since MD is not fully adequate in handling Cd, therefore, the additional cost for adsorption of Cd can vary from 0.5 \$/m<sup>3</sup> to 4 \$/m<sup>3</sup> (depending upon considered adsorbent) when adsorbent cost, including sludge treatment and the electricity cost, is considered (Kyzas and Kostoglou, 2014).

Fig. 6 presents the sensitivity of clean condensate cost while varying different techno-economic parameters. For the purpose, plant capacity (500,000 m<sup>3</sup>/year), plant life (20 years), interest rate (5%), membrane price (90  $\text{m}^2$ ), thermal energy cost (77 MWh), MD module price (6100  $\text{m}^2$ ), thermal energy cost (0.02  $\text{m}^3$ ) were varied  $\pm$ 50% under the techno-economic limitations. The outcomes show that among the analyzed technical parameters, plant life takes the lead where reducing the plant life by half (10 years) results in a 6% increase of clean condensate cost for case 1 and a 3.5% increase for case 2. In contrast, considering plant capacity, when it becomes 0.75 Mm<sup>3</sup>/year, the clean condensate cost can only be reduced by 1% for case 1 and 4% for case 2. It is note-worthy that the clean condensate cost is highly sensitive to thermal

energy costs. When thermal energy cost reduces by 50%, the clean condensate cost decreases by 45% for both cases. Additionally, MD modules price can also affect the clean condensate cost by a maximum of 3-4% when the price becomes half (3000/unit). However, when the interest rate is considered 7.5%, the treatment cost increases by 2.5% for case 1 and 1.1% for case 2. Furthermore, the prices of heat exchangers, membrane replacement frequency, and pretreatment extent do not significantly impact the clean condensate cost. It was found that the clean condensate cost can be varied maximum up to ~45% when various techno-economic parameters are altered  $\pm$ 50%.

As mentioned above, plant capacity merely affects the unit clean condensate cost. However, it significantly influences the total thermal power demand of the MD system, as shown in Fig. 7. Due to the limitation of practical allowable use of locally produced district heating, a limit is necessary to set, i.e., 10% of total locally produced district heating for operating the MD system. This limit can define the reasonable MD plant capacity in order to match the corresponding thermal power demand with the amount of available district heating for both cases. For example, Bristaverket produces 1253 GWh district heating; therefore, case 1 is feasible for up to 80  $m^3/h$ ; however, for case 2, the functional MD capacity is only  $35 \text{ m}^3$ /h. It is noteworthy that this study has taken the district heating retail price (77 \$/MWh); however, the internal DH production cost ranges from 5 to 15 \$/MWh. In the following scenario, the clean condensate cost can be reduced to  $2.58 \text{ }/\text{m}^3$ , in case 1. with the normalized operational cost of  $\sim 0.8 \text{ s/m}^3$ . The estimated operating cost for the MD system is comparable to the cost for the existing RO system for flue gas condensate cleaning (Lundgren, 2018) and lower than that of other studied methods (i.e., physicochemical method cost ~1 \$/m<sup>3</sup> (Chen et al., 2018)).

In summation, the proposed approach (district heating driven membrane distillation for flue gas condensate cleaning in CHP plants) can contribute towards achieving sustainable development goals in terms of clean and responsible energy production by introducing a novel method to recycle water (using sustainable district heating) as boiler feed in a cost-efficient manner. Moreover, this study represents a model for future innovative applications of membrane distillation.

#### 4. Conclusion



The present work is focused on experimental investigation and



Fig. 6. Sensitivity analysis of clean condensate cost for different techno-economic parameters.

Fig. 7. Annual district heating energy consumption for flue gas condensate cleaning using the MD system.

techno-economic evaluation of the membrane distillation process for advanced treatment of flue gas condensate in cogeneration plants. Considering the quality of permeate, the findings reveal that the MD system (without any pretreatment) is well suited for the waste-fired CHP facilities. However, due to high ammonia content, part of the flue gas condensate released from bio-fuel fired CHP facility was neutralized using sulfuric acid prior to its introduction to the MD system. (The other part was added to the MD system without considering any pretreatment.) Mixing of resulting clean condensates can provide the desired quality according to strict environmental regulations, i.e., conductivity <5 mS/m; total organic carbon <2 ppm; total hardness <0.15 °dH; pH ~7.5; total dissolved solids <20 ppm; chemical oxygen demand <20 ppm; turbidity <1 FNU; total hardness <0.15 °dH; and alkalinity ~20 ppm HCO<sub>3</sub>. The technical analysis of the industrial scale MD system of capacity 500,000 m<sup>3</sup>/year tells that annual thermal energy consumption can be approx. 88 GWh, when district heating supply and return lines are used to operate the MD system as the heat source and sink, respectively. Process economy evaluation shows that the unit clean condensate cost can be as low as 2.6  $/m^3$  for the aforementioned system, and varies considerably depending upon thermal energy cost.

# **Credit author statement**

**Imtisal-e- Noor:** Conceptualization; Data curation; Experimentation; Formal analysis; Investigation; Methodology; Validation; Visualization; Writing - original draft; Writing- review & editing.

Andrew Martin: Supervision and Project administration. Olli Dahl: Supervision.

# **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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