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Membrane Distillation and Applications for Water Purification in Thermal Cogeneration – Pilot Plant Trials

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Abstract

This research is a continuation of a Värmeforsk prestudy where the performance of membrane distillation (MD) water treatment is the focus of field trials. The report contains details of a test rig deployed at Idbäcken Combined Heat and Power (CHP) Facility (Nyköping) with a five-module MD unit. A long-term performance evaluation including thorough chemical testing of product water quality is presented. District heating supply and return lines were employed for heating and cooling, respectively; feed stocks include municipal water and flue gas condensate.

Sammanfattning

Kostnadseffektiv, pålitlig och energisnål vattenreningsteknik är en viktig del i moderna kraftvärmeverk. Membrandestillering (MD) är en ny, lovande teknik i sammanhanget. Tekniken utnyttjar partialtryckdifferenser för att rena vatten med hjälp av hydrofoba membran och har följande möjliga fördelar gentemot tekniker som omvänd osmos (RO): möjligheten att ta till vara spillvärme; minskad känslighet gentemot variationer i pH eller salthalt; lägre kapital- och driftskostnader. Denna forskning avser en fortsättning av en förstudie (Värmeforsk rapportnr 909) och omfattar fältstudier vid Idbäckens Kraftvärmeverk (Nyköping). Till målgrupperna hör miljötekniker och driftoperatörer med intresse för ny vattenreningsteknik.

Testanläggningen bestod av fem MD-moduler som kunde producera 1-2 m^3/dag ren vatten. Fjärrvärmenätets framledning används för processvärme. Både stadsvatten och rökgaskondensat testades som processvatten. Försöken delades i tre faser: (1) parametrisk analys med hänsyn till utbyte; (2) långtids drift med stadsvatten som processvatten; (3) utvärdering av rökgaskondensat som processvatten. Dessa försök ägde rum mellan april-oktober 2006 med uppehåll under sommaren.

MD:s prestanda vad gäller utbyte beror på processvattnets inloppstemperatur, flödeshastighet samt temperaturskillnaden over membranet. Tidiga resultat från försöken med stadsvatten visade att utbytet var i linje med tidigare experiment, därmed bekräftades förstudiens resultat. En minskad elförbrukning kunde uppnås genom seriekoppling av MD-enheterna, fast utbytet blev lägre, framförallt vid lägre flödeshastigheter. Dessa resultat visar behovet med ordentlig systemdesign för optimeringsändamål.

Vad gäller långtidsprestanda kunde man se en minskning av utbytet först efter 13 dagars kontinuerlig körning. Fällning och igensättning av inloppsmunstycken och delar av membranerna orsakade denna nedgång, dock var permeatets kvalitet oförändrat. Produktvatttnets konduktivitet låg på 1-3 μS/cm för on-line prover och under gränsvärdena för de externa analyserna. Nyckelparametrar såsom kisel och natrium låg i koncentrationer som skulle vara godkända för pannvatten.

Begränsningen att utnyttja provplatsen ledd till bara ett försök med filtrerat rökgaskondensat. Produktvattnet visade relativ hög konduktivitet på grund av ammoniak och kolsyra, men avskiljning av tungmetaller och andra icke-flyktiga ämnen var mycket tillfredsställande.

Det finns ett behov av ytterligare långstidsförsök för att bekräfta resultaten, dessutom är grundläggande forskning av intresse för att optimera MD-enhetens prestanda. Tillämpningar i andra områden som till exempel avsaltning är kanske mer attraktivt just nu.

Nyckelord: membrandestillering; vattenteknik; spädvatten; rökgaskondensat;
spillvärme.

Summary

Water treatment is an important auxiliary process in all thermal cogeneration plants. In this context membrane distillation (MD) is a novel technology that is potentially advantageous to technologies like reverse osmosis in the following ways: ability to utilize low-grade heat; reduced sensitivity to fluctuations in pH or salt concentrations; and lower capital and operation and maintenance costs (assumed in the case of fully-developed technology only). This research is a continuation of a Värmeforsk prestudy (report no. 909) and encompasses field trials at Idbäcken Combined Heat and Power (CHP) Facility (Nyköping). Target groups for this study include environmental engineers with particular interest in emerging water purification technologies.

The test rig consisted of a five-module MD unit capable of producing 1-2 m^3/day purified water. District heating supply was employed for heating; feed stocks include municipal water and flue gas condensate. Field trials can be divided into three phases: (1) parametric study of yield; (2) long term operation with municipal water as feed stock; and (3) evaluation of flue gas condensate as a feed stock. Testing commenced in the beginning of April 2006.

The performance of MD concerning production rate is highly dependent on the feed stock temperature, flow rate and temperature difference across the membrane. Initial results for municipal water feed stocks showed that product water fluxes were in line with previous experiments, thus confirming the findings made in the prestudy. Connecting several MD modules in series has the advantage of reducing the electrical energy consumption needed for recirculation; the penalty comes in less efficient operation from flux point of view. This is more critical in the case of low flow rates, and hence much careful design studies are needed to optimize the system.

Regarding the long term performance, the test period lasted for 13 days on a continuous operation basis before the first flux deterioration was encountered. This was due to scale formation on the module feed inlets, flow distributors inside each cassette, and portions of the membranes. Though the permeate flux was affected, the permeate quality did not deteriorate. Product water conductivity ranged from 1-3 μS/cm for on-site measurements and below detection levels in the sample analyzed in an external lab (for municipal water feedstock). Key parameters like silica and sodium met the requirements for make up water in boilers.

Due to time constraints only one test could be performed with a filtered-only flue gas condensate feedstock. While the product water exhibited high conductivities due to slip of ammonia and carbonates, the performance of the MD unit concerning removal of heavy metals and other non-volatile components was highly encouraging.

Future work concerning additional long-term testing is warranted in order to confirm these findings. There is also a clear need for more fundamental research and development activities in order to optimize the MD unit performance. Applications in

other fields, in particular desalination for drinking water purposes, may be more attractive at this point.

Keywords: membrane distillation; water treatment; make-up water; flue gas condensate; low-grade heat utilization.

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Appendices

1 Introduction

1.1 Background

Water treatment is an important auxiliary process in all thermal cogeneration plants. In this context membrane distillation (MD) is a novel technology that is potentially advantageous to technologies like reverse osmosis in the following ways: ability to utilize low-grade heat instead of electricity; reduced sensitivity to fluctuations in pH or salt concentrations; and lower capital and operation and maintenance costs (assumed in the case of fully-developed technologies only). This research is a continuation of a Värmeforsk prestudy [1] where the performance of MD-based water treatment was explored via laboratory testing, system simulations of thermodynamic performance, and economic evaluations. Part 2, encompassing field trials, contains details of a test rig deployed at Idbäcken CHP Facility (Nyköping) with a five-module MD unit capable of producing 1-2 m^3/day purified water. A long-term performance evaluation including thorough chemical testing of product water quality is presented. District heating supply line was employed for heating while municipal water was used for cooling; feed stocks include municipal water and flue gas condensate.

1.2 Description of the research area

Thermal cogeneration plants require purified or treated water for a number of processes, i.e. boiler/district heat make-up water systems and flue gas condensate treatment. The selection of the exact water treatment process is of course dependent upon the final water quality along with the volume of water to be treated. Membrane-based technologies like reverse osmosis (RO) are an important component in such systems as they have been developed to meet the needs of the power generation industry. While RO and other methods are well established and generally effective, there is still room for improvement regarding economy and enhanced water purity. In this context membrane distillation (MD) is a unit operation that deserves more attention as a promising alternative or complementary technology for water treatment in cogeneration facilities.

1.3 Brief Description of Membrane Distillation (MD)

Membrane distillation (MD) is a thermally driven process that utilizes a hydrophobic micro-porous membrane to support a vapor-liquid interface. If a temperature difference is maintained across the membrane, a vapor pressure difference occurs. As a result, liquid (usually water) evaporates at the hot interface, crosses the membrane in the vapor phase and condenses at the cold side, giving rise to a net transmembrane water flux. The technology was introduced in the late 1960s but initially did not receive significant interest due to several reasons, e.g. the observed lower production compared to reverse osmosis, and unavailability of suitable membranes for the process [2-4]. MD received renewed interest within the academic communities in the early of 1980s when novel membranes and modules with better characteristics became available [4]. Moreover, the ability of MD to utilize low grade heat in a form of waste heat/renewable energy sources had boosted the interest and research in order to find suitable application areas as well as improving the merits of the technology. Nonetheless, MD is not implemented yet in industry for water purification or desalination. A thorough historical perspective of MD development can be found in the review articles by Lawson and Lloyd [2], Alklaibi and Lior [3], and Bourawi et al.[4].

In general MD has a several advantages and disadvantage including [1,2]:

Advantages of MD:

- 100% (theoretical) rejection of ions, macromolecules, colloids, cells, and other non-volatiles
- Lower operating temperatures than conventional distillation
- Lower operating pressures than conventional pressure-driven membrane separation processes
- Low sensitivity to variations in process variables (e.g. pH and salts)
- Good to excellent mechanical properties and chemical resistance
- Reduced vapor spaces compared to conventional distillation processes

Disadvantages of MD:

- High energy intensity (although energy, i.e. heat, is usually low grade)
- Low yield in non-batch mode; high recirculation rates in batch mode
- Sensitive to surfactants
- Undesirable volatiles such as ammonia or carbonates must be treated separately (degassing, pH control, or other methods required)

1.4 Summary of Pre-study results

In 2004 Värmeforsk commissioned a pre-study in order to examine the potential of MD water purification for cogeneration applications [1]. Specific elements of this work included a literature survey, theoretical considerations of heat and mass transfer, and scale-up of experimental results for a case study involving a 10 m^3/h water treatment system. Results show that MD is a promising alternative to RO in existing or new treatment facilities. The most favorable results were obtained for alternatives where either the district heat supply line or low-grade steam is available. Specific energy consumption ranges were reported as follows: 4-5 kWh/m^3 thermal; and 1.5-4.0 kWh/m^3 electrical. The relatively high electricity consumption was linked primarily to high recirculation rates versus relative low water production in batch mode. Although the combined energy consumption is higher than RO, future process improvements can be employed to offset or eliminate this disadvantage. MD thus demonstrates satisfactory energy performance compared to existing technologies. Specific costs lie in the range of 10-14 SEK/m^3 for the most likely MD system scenarios. These results indicate that MD is presently more expensive than RO, although this comparison should be weighed against the level of development for each respective technology.

1.5 Research Partners

Since 2003 the Department of Energy Technology at KTH has collaborated with XZero AB on membrane distillation. XZero and its parent company Scarab Development AB has spent several years developing MD technology for applications in the semiconductor industry, process industries, desalination, and drinking water purification. Previous cooperating agencies included Statsföretag and ABB. The technology has been independently evaluated by Sandia National Laboratory in the US in the context of ultrapure water production for the semiconductor industry. Tests have also been performed in Greece for desalination for an 11-month period. XZero has commercialized its technology for small-scale drinking water production and is interested in finding other application areas.

During 2005 Xzero ordered the construction of a five-module MD pilot plant. This facility was assembled at Uddevalla Finmekanik AB and is capable of producing 1-2 m^3/day purified water, depending upon the heat source; necessary auxiliary equipment such as a tank, controls, and measurement systems are included (see Figure 1). Parallel to this work discussions were initiated with CHP facility owners in order to locate a suitable test site. Vattenfall Utveckling AB showed keen interest and helped to open up Idbäcken CHP Facility for testing. The MD pilot plant was finally delivered to Idbäcken in April 2006.

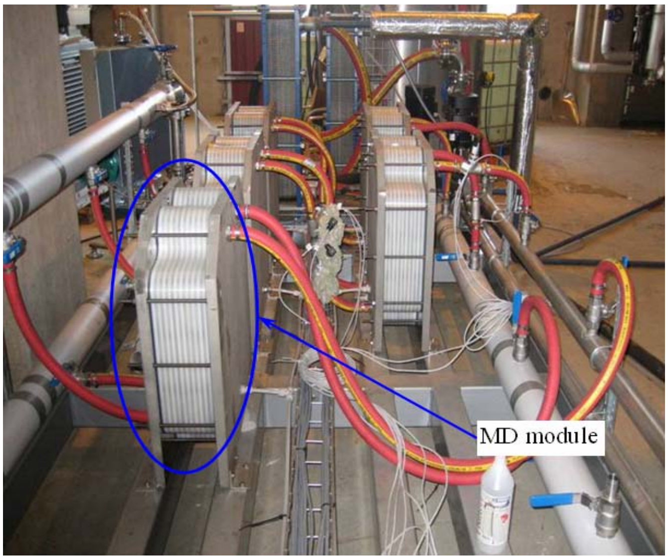

Figure 1. Test facility at Idbäcken Cogeneration Facility (Nyköping)

Figur 1. Testanläggning vid Idbäckens Kraftvärmeverk (Nyköping)

1.6 The purpose of the research assignment and its role within the research area

The purpose of this research is to compliment the work performed in the previous Värmeforsk study with pilot-plant trials at Idbäcken CHP Facility. Specific goals include the following:

- Obtain long-term experimental performance data for:
  - Water production rates as a function of process variables (hot/cold side temperatures, recirculation rates, feedstock type, etc.)
  - Product water quality for both municipal water and flue gas condensate feedstocks
- Observe any possible tendencies for performance deterioration (like leakage or fouling) under realistic operating conditions
- Check previous findings in relation to yield, thermodynamic performance, and economics
- Provide recommendations with respect to commercialization

2 Test Plan and Test Facility

2.1 Test Facility

The test facility (Figure 1) comprises five Air Gap Membrane Distillation (AGMD) modules connected in three parallel cascades. Each cascade (except the third set) consists of two modules connected in series. Each module consists of 10 cassettes and the total membrane area is 2.3 m^2. The membrane material is PTFE with a porosity of 80% and thickness of 0.2 mm. The width of air gap of AGMD is 2 mm. The membrane module is made up of nine feed channels and nine permeate channels, and the size of module is 63 cm wide and 73 cm high with a stack thickness of 17.5 cm. Figure 2 illustrates the test layout. System control and primary data acquisition were handled via PLC connections to Citect Runtime software installed on a standard PC. A separate data logger recorded interstage inlet/outlet temperatures and inlet/outlet pressures. On site conductivity measurements were also introduced to check the instant bulk quality of the product water. Product water yield was determined manually. A de-gassing unit is connected to the facility but failed to function properly during the test period.

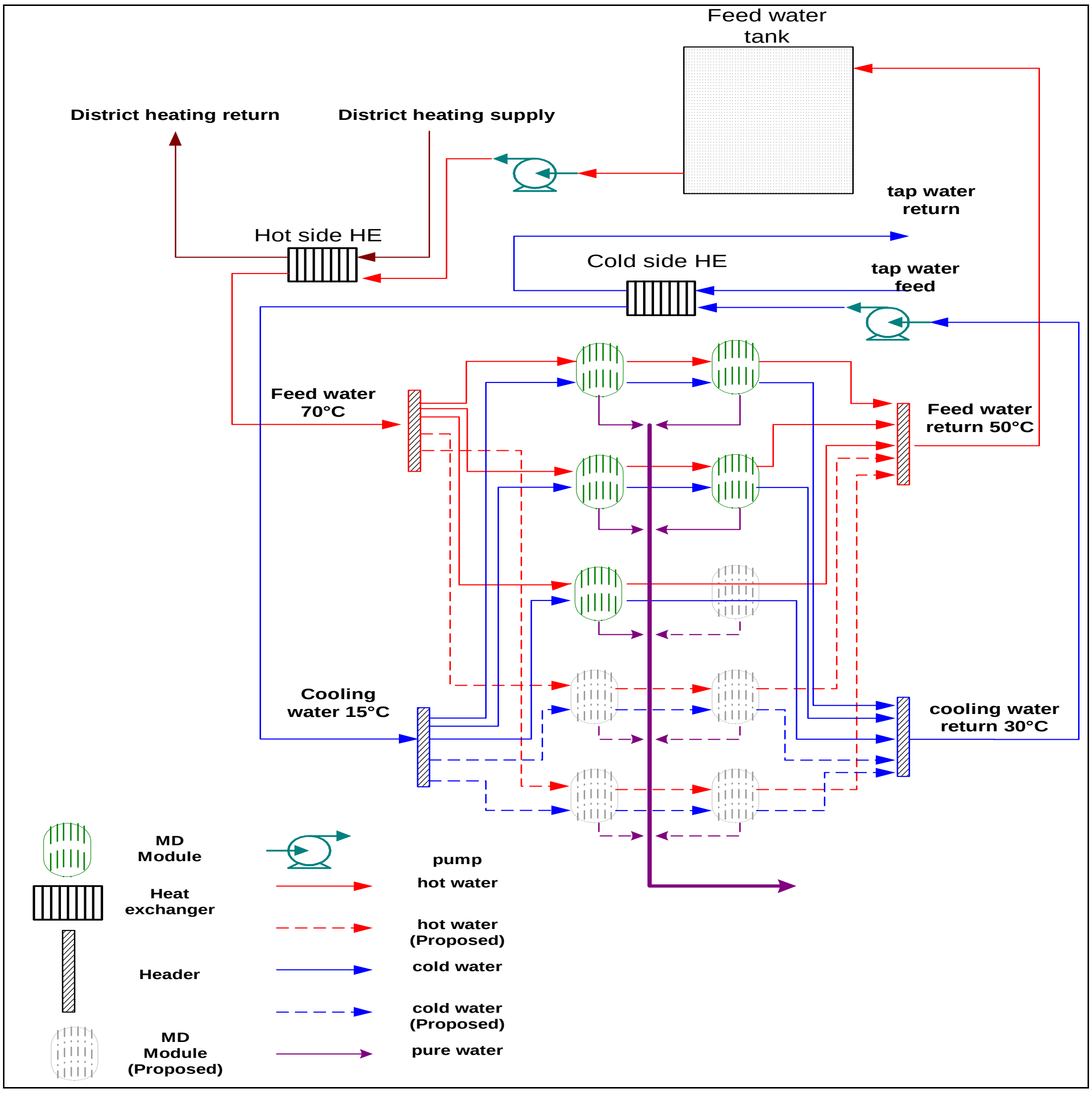

Figure 2. Test facility layout. Modules 1, 3, and 5 shown at left, while modules 2 and 4 shown on right.

Figur 2. Flödeschema over testanläggningen. Modul 1, 3 och 5 till vänster, samt modul 2 och 4 till höger.

2.2 Test Plan and Actual Outcome

Testing was divided into three phases: (1) parametric study of yield with municipal water as feedstock; (2) long term operation with municipal water as feedstock; and (3) evaluation of flue gas condensate as feedstock. Originally tests were planned to start up in February 2006, but various delays meant that testing commenced first in April. Some problems were encountered initially, some of which were attributed to limitations in the rig design. For example the facility lacked an air bleed valve, so eliminating air pockets or bubbles was handled manually. The installed degasser malfunctioned, despite repair attempts by the supplier. This had a negative impact on operation and also contributed to vapor carryover in subsequent flue gas condensate trials. The combination of these factors caused air to accumulate in the hot-side primary loop heat exchanger, which in turn caused a large pressure drop, reduction in flow rate, and reduction in yield. The line pressure was also affected and in several cases the high pressure alarm was activated and the system shut down. Nonetheless some data was gathered, and water samples were taken for later analysis. Experiments were suspended on 24 May 2006 in conjunction with Idbäcken's summer maintenance period.

The second phase aimed at testing the facility for long term performance (continuous operation) for around one month. This phase started on 13 September 2006 and lasted until 13 October 2006. During the first ten days of operation, the MD facility was operated continuously and no major disturbance was noticed. However, by the 13th day a 20% reduction in flux was observed coupled with an increase of pressure at the feed inlet along with a lower flow rate. The first speculation that such a resistance to flow was due to blockage of flow channels in the MD modules by accumulation of particulates. Shortly before this point in time a light brownish color of the feedwater in the tank was observed and it was attributed to contamination by dust and dirt from the open environment (the site where the facility was installed contained many airborne contaminants). However, a further investigation supported the assumption that the modules were subjected to scaling which caused a reduction in the flow passages and/or reduction on the membrane area available for evaporation. By the end of the test period, the flux reduction reached a high level of 32%. During this phase, a sample of the product water was taken and analyzed.

The third phase was conducted by testing one of the modules with flue gas condensate as a feed stock. This phase was done on two separate days. Samples of the product water and the raw flue gas condensate were taken and analyzed. Operations ceased at the end of October 2006 in conjunction with the start of the heating season. The total accumulated run time for facility was around 500 h.

3 Test Results and Discussion

3.1 Production Flux, Municipal Water Feedstock

The parametric study included mainly running tests with variable feed flow rates ranging from 300 to 1050 l/h for one cascade (i.e. two modules connected in series). Figure 3 shows the relation between the flow rate and flux. Different points must be stated here. First, the curve is drawn for two units connected together in which the feed outlet for the first is used as a feed in the second (see Figure 2). At high flow rates (1050 l/h) the second stage produces about 35% of the total product output, while at low flow rates this stage is virtually non-functioning. It is well known that low flow rates results in larger thermal boundary layer, i.e. larger temperature polarization effect [4]. Such effects result in a lower membrane surface temperature compared to the bulk temperature in the flow channels within the module, and hence lower trans-membrane temperature difference and lower flux. Moreover, the large temperature drop exhibited at low flow rates causes the flux in subsequent stages to drop significantly. Previous findings available in the literature reported a linear increase in flux enhancement with higher flow rate [3,4], and the present results largely follow this trend.

The direction of coolant flow was reversed to yield a counterflow-type arrangement (i.e. lowest temperature stream introduced in the second stage). Here the flux was found to be enhanced slightly. This point is more important in the case of optimizing the operational design in the case of designing an industrial MD rig, and not much focus was put on this at present.

A noticeable decay in flux was noted during the second phase of testing, around the 13th consecutive day of operation (corresponding to around 370 cumulative hours of operation). Approximately 20% reduction in product water flux was recorded, coupled with an increase of pressure at the feed head along with a lower flow rate. By the end of the test period, the flux reduction reached a high level, over 30%. By that time the first stage (modules 1, 3 and 5) had a very low product water flux, roughly half that measured in the beginning of the test. The flux decay was a result of scaling problem that caused clogging of the feed inlets and feed channel distributors and also reduced the membrane surface area available for evaporation. First stage modules were more adversely affected by scaling as compared to second stage modules owing to higher operational temperatures.

The pressure drop over one module ranged from 0.02 to 0.04 bar, corresponding to a flow rate range of 300 to 1200 l/h. This is much lower than the estimated pressure drop of 0.1 bar stated in the previous study [1], as a higher flow rate was considered here (2400 l/h).

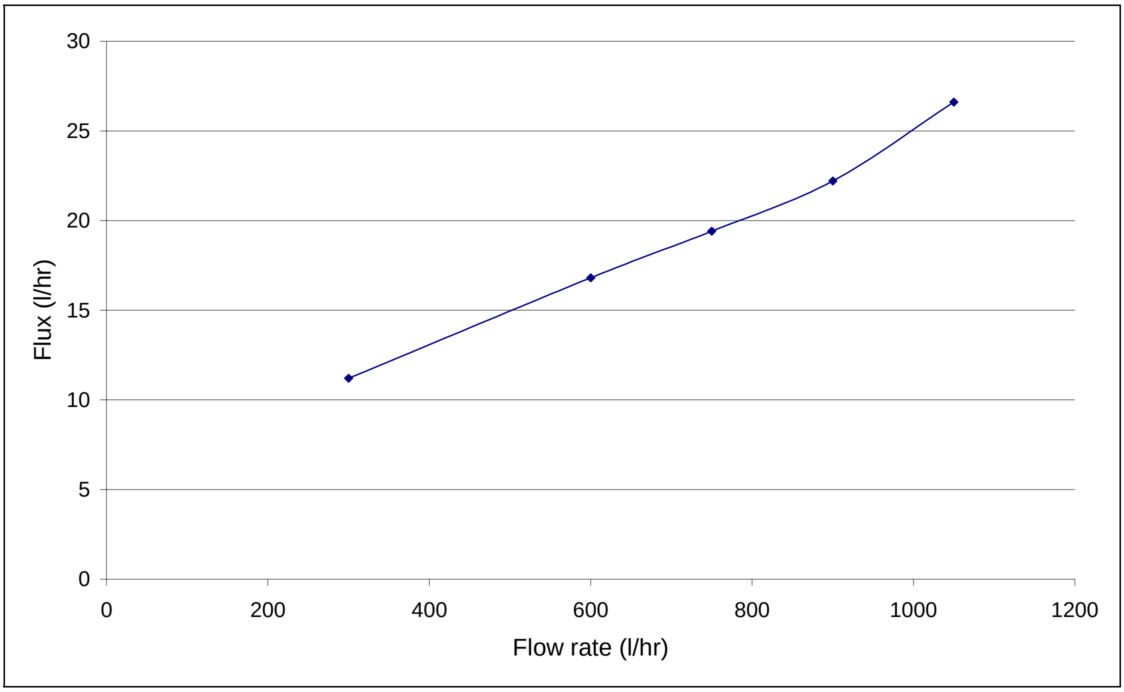

Figure 3. Water production for one cascade at various flow rates (70 ºC feed temperature and 15 ºC cold water supply temperature; cold water flow rate approximately the same as the feed flow rate)

Figur 3. Renvattenproduktion vid kaskadkoppling av två moduler vid olika flöden (70 ºC matarvattentemperatur och 15 ºC kallvattentemperatur; kallvattenflöde ungefär detsamma för matarvattenflöde)

3.2 Water Quality, Municipal Water Feedstock

In addition to on-site water quality checks, several water samples of the product water were taken from each module. Table 1 contains water analysis results obtained in the early phase of testing. Here SiO_2, Na and conductivity were selected as quality indicators, which is typically the design basis for a water treatment plant in cogeneration facilities [5]. Comparisons are also made with the feedwater as a base line. The bulk quality indication, represented by conductivity, shows a high separation efficiency. Product water conductivities ranged from 1.5 to 2.0 µS/cm for most of the samples. There were some cases were conductivity levels exceeded 2.0 µS/cm, most probably due to contamination of the collecting flask. Most of SiO_2 and Na levels were below the detection limit in the product water obtained from module 1, where a slightly higher level is presented. However, on-line conductivity checks did not reveal a different behavior for this module compared to the others, and the possible explanation for this irregularity is probably due to contamination during sampling. Overall these results are directly competitive with those that could be derived from other technologies like RO permeate (e.g. see Hellman [6]). These results are also favorable in respect towards meeting water quality guidelines for cogeneration facilities. For instance the following limits have been recommended for condensate and steam in steam turbine cycles [6]: SiO_2 < 5 µg/kg (≈ 5 µg/l); Na < 3-5 µg/kg (≈ 3-5 µg/l); and conductivity < 3-6 µS/cm (ammonia dosing for pH control).

Table 1. Water analysis for each module's product water

Tabell 1. Vattenanalys för de olika modulerna

Parameter	SiO_2		Na		Conductivity	
unit	µg/l		µg/l		µS/cm	
Feed Water	5000-10000		17500		467	
Module 1	6	13	14	60	1.6	2.5
Module 2	≤ 5	≤ 5	≤10	≤10	1.5	2.0
Module 3	≤ 5	12	≤10	18	1.6	1.7
Module 4	≤ 5	≤ 5	≤10	≤10	1.4	1.9

Table 2 represents a more comprehensive analysis for the cations and anions in the MD product water sample collected. Analyses were performed by an accredited laboratory (see Appendix A). As stated previously, flux decay was observed during this phase. The results shown below are for one sample collected after the flux deterioration. Most of the elements are below the detection levels and others are slightly above, and comparable to results in Table 1. Sodium (Na) is considerably high compared with the cut-off represented in the previous analysis (Table 1). There is no solid explanation that can be given in this case.

Table 2. MD product water analysis, municipal feedwater

Tabell 2. MD produktvattenanalys, med stadsvatten som matarvatten

Parameter	Result	Error (±)	Unit
Ca	0.297		mg/l
Fe	<0.004		mg/l
K	<0.5		mg/l
Mg	<0.09		mg/l
Na	0.175	0.24	mg/l
S	0.337	0.064	mg/l
Al	4.67	5.63	µg/l
As	<1		µg/l
Ba	<0.2		µg/l
Cd	<0.05		µg/l
Co	0.105	0.105	µg/l
Cr	<0.5		µg/l
Cu	<1		µg/l
Hg	<0.02		µg/l
Mn	<0.2		µg/l
Ni	2.55	0.53	µg/l
Pb	<0.2		µg/l
Zn	<2		µg/l
Chloride	<0.60		mg/l
Sulfate	<0.50		mg/l
Ammonia	<0.025		mg/l
Alkalinity	1.4		mg HCO_3/l
pH	5.9		

3.3 Test With Flue Gas Condensate as Feed stock

The test was done on two separate days mainly to confirm the results. The bulk quality of the product was checked via the on-site conductivity meter. The raw flue gas condensate was filtered with a 5-micron filter to remove larger particles. Product water fluxes recorded in this phase was similar to the one recorded at the end of phase two, when fouling effects were noted. It was not possible to obtain more detailed measurements as the main aim was to evaluate the product water quality.

A chemical analysis of the flue gas condensate is presented in Table 3. It can be seen that Na level is considerably high in the flue gas condensate, along with heavy metals (mainly Pb and Zn). In the anions table, the condensate is characterized by high presence of chlorides, sulfate, ammonia and alkalinity. It must be noted that the flue gas condensate used in the test was untreated and did not pass any kind of treatment other than cartridge filtering. Typical steps for an industrial application would include sand filtration, volatile stripping, and pH adjustment.

Table 3. Analysis of untreated flue gas condensate

Tabell 3. Analys över obehandlat rökgaskondensatet

Parameter	Result	Error (±)	Unit
Ca	35.0	4.2	mg/l
Fe	0.0268	0.0069	mg/l
K	3.59	0.46	mg/l
Mg	4.28	0.53	mg/l
Na	902	109	mg/l
S	60.6,	8.9	mg/l
Al	89.4	16.5	µg/l
As	25.7	7.0	µg/l
Ba	55.3	9.5	µg/l
Cd	0.910	0.155	µg/l
Co	0.422	0.183	µg/l
Cr	3.21	1.07	µg/l
Cu	32.7	5.6	µg/l
Hg	5.64	0.37	µg/l
Mn	28.6	3.6	µg/l
Ni	7.20	0.35	µg/l
Pb	46.0	7.8	µg/l
Zn	387	61	µg/l
Chloride	1490		mg/l
Sulfate	168		mg/l
Ammonia	115		mg/l
Alkalinity	225		mg HCO3/l
pH	8.4		

After treating the flue gas condensate in the MD facility, a quick check with the on-line conductivity meter showed high readings, up to 28 mS/m (280 µS/cm). The analysis (Table 4) indicates that such high conductivity was a result of volatile ammonia passing through the membrane and presence of alkalinity (HCO_3). Most problematic metals were successfully removed and their concentrations were below the detection limits. However, sodium and chloride ions were quite high and exceeded the recommended values for boiler feedwater.

Table 4: Analysis of MD product water, flue gas condensate feedstock

Tabell 4. MD produktvattenanalys med rökgaskondensat som matarvatten

Parameter	Result	Error (±)	Reduction (%)	Unit
Ca	<0.2		>99%	mg/l
Fe	<0.004		>85%	mg/l
K	<0.5		>86%	mg/l
Mg	<0.09		>98%	mg/l
Na	1.27	0.24	99,90%	mg/l
S	0.274	0.064	99,50%	mg/l
Al	5.98	5.63	93,30%	µg/l
As	<1		>96%	µg/l
Ba	<0.2		>99,6%	µg/l
Cd	<0.05		>94,5%	µg/l
Co	0.105	0.105	75,10%	µg/l
Cr	<0.5		>84%	µg/l
Cu	<1		>96,9%	µg/l
Hg	<0.02		>99,6%	µg/l
Mn	<0.2		>99,9%	µg/l
Ni	1.62	0.53	77,50%	µg/l
Pb	<0.2		>99,6%	µg/l
Zn	<2		>99,5%	µg/l
Chloride	1.5		99,90%	mg/l
Sulfate	<0.50		>99,7%	mg/l
Ammonia	49		57,40%	mg/l
Alkalinity	180		20%	mg HCO_3/l
pH	8.7			

4 Results: Scale-up and Discussion

In order to understand the results in a large-scale production scheme, the MD facility is assumed to be connected to the district heating network, where the heating is supplied by the district heating supply line, and the cooling is provided by the district heating return line. The thermal energy consumption is thus calculated based in the difference between the supplied heat and the heat added to the sink. Figure 2 illustrates a simplified scheme of the layout (without the temperature defined in the actual testing). The feed water temperature (assumed to be supplied by district heating supply) was chosen to be either 70 or 90 ºC based on different possible district heating operation conditions in different cogeneration plants. The district heating return line temperature is assumed to be 40 ºC. The assumed flow rate is chosen to be 1200 l/h per cascade, and the targeted production rate is 10 m^3/h. Aspen Utilities was employed as the simulation software tool, and the other major assumptions are included in the prestudy [1]. Table 5 present the simulation results for hot side feed temperatures of 70 to 90 ºC.

Table 5. Simulation results, scaled-up performance

Tabell 5. Simuleringsresultat, uppskalade prestandan

Parameter	Case I	Case II	Alternative 1, Prestudy [1]
Cascade flow rate (l/h)	1200	1200	2300
MD hot side temperature (°C)	70	90	90
MD cold side temperature (°C)	40	40	55
Pure water output (m^3/h)	10	10	10
Specific thermal energy consumption (kWh/m^3)	5.5	12.1	6.0*
Specific electricity consumption (kWh/m^3)	1.3-1.5	0.6-0.7	2.0
Membrane area (m^2)	2392	1141	828
No of MD Modules	1040	496	360

*original value adjusted to account for heat losses as discovered in recent field trials

It is clear that the best results in terms of efficient membrane use are obtained with higher feedwater temperatures. Reducing the area of the membrane and subsequent number of modules used is crucial since they comprise the most expensive part of the investment (around 40% of the total cost of the MD facility [1]). This is also true for the operational cost, since less MD modules means less maintenance cost. As shown in comparing Case I and II, the thermal energy consumption is almost doubled if the operation temperature is raised from 70 to 90 ºC, while the specific electrical consumption is halved. The reason for this increase in the specific thermal energy consumption is due to the relatively high level of heat lost to the environment.

Alternative 1, which was analyzed in the prestudy, has the best performance in terms of membrane use and thermal energy consumption for two reasons: (a) much higher flow rates were considered; (b) a cascading concept was not employed, i.e. heat exchange was utilized between stages in order to maintain high inlet temperatures. However this arrangement has relatively large electricity consumption due to the high flow rate. For comparison typical reverse osmosis (RO) facilities for CHP applications have a specific electricity consumption around 1.5 kWh/m^3 (with no thermal energy consumption).

The cost of water for the system studied in Case II is expected to be about 40% higher than the figures reported the Prestudy [1] – 14-20 SEK/m^3 (check Appendix C for detailed cost estimation) as compared to 10-14 SEK/m^3. This expectation assumes direct proportionality to the additional number of modules that are estimated in the updated simulations as compared to the earlier analysis. A two-stage system would be preferable if electricity consumption is to be reduced, otherwise the layout proposed in the prestudy is favorable.

5 Fouling and Scaling

5.1 Background

Fouling and scaling are two important mechanisms that affect stability of the MD process and lead to reduce the overall efficiency. Despite reports that membrane fouling in MD is less problematic than in other processes due to larger pore size [2], the phenomena is not well studied, either experimentally or analytically [4]. Fouling and scaling can cause pore clogging in MD membranes which lead to reduce the membrane area available for water vaporization and hence reduce the flux. In addition, such a build-up of fouling and scaling surfaces reduces the flow channel area which causes a pressure drop and lower flow rates, leading to higher temperature polarization effects and reductions in flux [8-11]. Moreover, fouling and scaling may cause membrane partial wetting or severe membrane damage. There are several types of fouling and scaling phenomena: biological fouling; particulate fouling; and crystallization fouling or scaling. A brief description of these phenomena is contained below.

Biological fouling is caused by microorganisms' growth on the membrane surface and is mainly dominant where operational conditions are similar to the natural aqueous environment in which the microorganisms thrive [8]. Concerning MD, typical operational conditions featuring a high temperature, (possible) high salt concentration, and low pH due to acids (for scaling control) all lead to a low biofouling potential. However it is well known that bacteria are able to grow under extreme conditions such as high temperatures as 110 °C and pH values as low as 0.5 [8,12].

Scale formation fouling results from deposition and formation of crystals on membrane surfaces during the treatment of salt concentrated feed solutions [4]. Scale formation causes clogging of membrane pores and reduces its surface area. The build-up of scale layer contributes to the temperature polarization effect and causes a pressure drop, and hence reduces the flux. Moreover, such deposition may deteriorate the membrane hydrophobicity and cause membrane wetting which influence the product quality. Scaling accrues when the solubility of such compound is exceeded. As solubility depend on the degree of salinity and temperature, scaling is more likely to occur in MD operation due to high temperature operation level.

Gryta et al. [13] reported experimental results with tap water as a feed for the Direct Contact Membrane Distillation (DCMD) process, which showed a rapid decline of the permeate flux. Heating of the feed in the MD process caused decomposition of HCO_3^- ions present in the feed water and precipitation of $CaCO_3$ on the membrane surface. Cleaning the membrane with HCl resulted in the dissolution of the deposit on the membrane surface, and the initial membrane permeability was restored. Acidification of the feed water to pH of 4 was found to be efficient in controlling scaling formation caused by precipitation of $CaCO_3$.

5.2 Scaling Observed in Tested MD Modules

As mentioned previously, scaling was encountered during the test period. The effect of scaling on the flux quantity was noticed clearly after 400 h of operation and had led to significant flux decay. However, one can say that scale was building up during the whole process. Factors like several shut downs and interruptions would be assumed to boost the process. No pretreatment or scale control measures were taken during the test period. Moreover, no rinsing was conducted after the first test break (24^{th} of May).

In order to understand the mechanism of scaling in this particular module type, Module 1 was de-assembled and inspected. Figures 4 and 5 show the scale formation at the feed inlets of each cassette of the module and at the distribution channels inside one of the cassettes, respectively. The scale formation at the feed inlets was building up as finger-like crystals on the inlet openings causing a partial or total clogging. Inside the cassettes, the scale formed as loose flakes which accumulated and clogged the distribution channels. On the membrane surface, the scale layer was gradually decreasing with the length of the membrane flow direction (corresponding to change in tangential temperature decrease). The scaling pattern on the membranes gives an assumption of how the mass transfer takes place. Since the scale is dominant on the upper part where the highest temperature is expected, it is most probably in this region major part of the mass transfer of vapor takes place.

Figure 4. Scale formation at the cassette entrances

Figur 4. Utfällning vid kassetinloppen.

Figure 5. Scale formation blocking the distribution channels

Figur 5. Igensättning av inloppsmunstycken.

The content of the scale flakes was analyzed by scanning electron microscopy (SEM) coupled with the energy dispersion spectrometry (EDS) (See Appendix B). The major scale content was $CaCO_3$, which is the most experienced one in running with tap water. Other elements such as Fe, Mg and S were also presented with a very low level though it is suggested that the light brown color of scale was due to Mg and Fe_2O_3.

6 Long-Term Performance in Other Investigations

Since MD technology is still at the research stage, there is a lack of operation experience and long-term behavior. The shortage in time and several delays and interruptions due to technical problems occurred during the test phases had a significant effect on performing a long test operation. In the following sections, a brief literature review of long term operation recorded in MD technology is presented.

The longest series of trials has been reported by Gryta [10] who conducted a three-year test program on a lab-scale DCMD module utilizing hydrophobic capillary polypropylene membranes. The MD installation was operated in a continuous mode in several measurement series. The installation was shutdown between the measurement series for a period ranging from a few weeks to several months. A total of 3500 hours of operation had been conducted over the entire period; with two types of feed water: around 2300 hours using RO permeate and 1200 hours using tap water. In the operation with RO permeate; the flux was almost stable for the first series which lasted for 400 hours with slight decrease at the period end. The product quality did not change and the conductivity was stable at the level of 1.6 μS/cm. However, the encountered flux decay was found to be a result of feed flow rate reduction caused by formation of a deposits layer on filter net mounted at the inlet of MD module. By switching to tap water as a feed, a rapid decline of the process efficiency was observed. Considerable amounts of CaCO3 precipitates were found on the membrane surface which caused a reduction in the surface area available for vaporization. A cleaning of such deposits was needed every 40-80 hours in order to recover the initial flux. The cleaning was achieved by rinsing the module with 2-5 wt. % HCl. However, a decline of product quality was observed due to possible membrane partial wetting. Over the three-year operational period it was found that capillary hydrophobic membranes were suitable for solute separation without suspended solids, and membranes exhibited a stable thermal stability.

A few other tests on DCMD were reviewed by El-Bourawi et al.[4] who reported more or less similar results. A four-month period experiment was conducted by Schneider and Wollbeck [14], aimed at investigating the scale and fouling formation applying normal tap water with a preliminary conductivity of 350 μS/cm as feed for the DCMD module, reaching a feed concentration with a conductivity up to 3500 μS/cm. A flux decline was observed during the first few weeks of the test. Treatment with HCl had resulted in a complete restoration of the original flux levels. However, although the original flux levels were restored after each HCl treatment process, flux decline seemed to be dominant and was reduced by approximately 20% lower than the original one by the last month. A similar flux decline was reported during 5-day continuous DCMD run carried out with brine as feed solution [4].

As a conclusion, it is clear that this aspect of MD technology is still far from being completely understood due to lack of data. Moreover, testing with seawater is even less studied.

7 Conclusions and Future Work

The performance of MD concerning production rate is highly dependent on the feed stock temperature, flow rate and temperature difference across the membrane. Although connecting several MD modules in series has the advantage of reducing the electrical energy consumption needed for recirculation, the penalty comes in less efficient operation from flux point of view. This is more critical in the case of low flow rates, and hence much careful design needed to optimize the system.

Regarding the long-term performance, the test period lasted for 13 days on continuous operation basis before the first flux deterioration was encountered. The reason for this was attributed to scale formation on the membrane cassettes feed inlets, flow distributors inside each cassette and the membranes upper half. Such scale caused a total or partial clogging for these channels/distributors/membrane pores, reducing the feed flow and hence the permeate flux. It must be mentioned that the MD modules were previously subjected to several discrete test periods which must be added to the operation time. The main problem encountered during the long-time test was scaling due to the high content of hardness in the feedstock (Ca and Mg). This problem can only be overcome with some kind of pre-treatment. The most common way to deal with hardness is by using softeners (ion exchangers or antiscale chemicals), which can be implemented readily in practice.

Though the permeate flux was affected by scaling problems, the permeate quality was stable over time and did not deteriorate. For municipal water feedwater (467 µS/cm) the product water conductivity was 1-3 µS/cm as measured on-site and lay below the detection levels in the sample analyzed at the external lab. Important parameters like silica and sodium levels met the requirements for make up water in boilers when running with municipal feedwater. However, when running with the flue gas condensate feedstock, high conductivity level was recorded in the product water due to the presence of ammonia and alkalinity (measured as HCO_3). Also, some ions like Na^+ and Cl^- were present in the product water at relatively high levels. There is no specific known reason for that taking in consideration that the purification is evaporative process. However, since treating flue gas condensate took place after the scale formation, it is possible that such scale caused partial damage for the membrane's hydrophobic character. The performance of the MD unit concerning removal of heavy metals is encouraging.

Possible implementation of MD technology in the near term would first require the resolution of a few issues. The one obvious hurdle is the high estimated product water cost of 10-20 SEK/m^3 (depending upon the system configuration). This value is higher than current technologies like RO and clearly points to the need for developing more efficient and inexpensive MD modules. Some more specific research and development needs include the following:

- The permeate flux is relatively low comparing with the recirculation rate, which has a negative impact on the electricity consumption. To be able to overcome this problem, appropriate redesign of the module that takes in account enhancing the mass transfer and increasing the membrane area per module volume is needed. Other possible MD configurations such as spiral-wound configuration [15] could be of interest in terms of reducing the foot print of the technology. However, such alternative must be judged by means of other factors such as energy efficiency, cost of production and handling simplicity.

- A rough investigation over the module internal design showed several weaknesses in hydrodynamic conditions. These weaknesses caused a high pressure drop which directly affecting the electrical energy consumption. More severely, possible flow maldistribution could be seen via membrane scaling, indicating the inefficient use of the membrane area and hence a relatively significant flux reduction.
- Measures to reduce the heat lost to the surrounding and lost heat by conduction through the plastic frames of the module cassettes must be taken, in the case of adopting the current module as it is today. A further fundamental study of the module design will eventually have a significant effect on flux enhancement and potentially, increase the technology efficiency.
- More attention should be paid to appropriate pre-treatment strategies in order to ensure reliable operation. Continued improvements in MD technology will most certainly enhance its attractiveness in the near future.

8 Acknowledgement

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Appendices

A Chemical Analysis

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Sida 1 (3)

Projekt

Vattenfall Utveckling AB
Anders Wik

162 87 Stockholm

Registrerad 2006-11-08
Utfärdad 2006-11-10

Analys: V3A

Er beteckning: **MD Product**
Labnummer: U10284767

Parameter	Resultat	Mätosäkerhet (±)	Enhet	Metod	Utf
Filtrerad	NEJ			1	V
Ca	<0.2		mg/l	1	E
Fe	<0.004		mg/l	1	H
K	<0.5		mg/l	1	E
Mg	<0.09		mg/l	1	E
Na	1.27	0.24	mg/l	1	E
S	0.274	0.064	mg/l	1	E
Al	5.98	5.63	µg/l	1	H
As	<1		µg/l	1	H
Ba	<0.2		µg/l	1	H
Cd	<0.05		µg/l	1	H
Co	0.105	0.105	µg/l	1	H
Cr	<0.5		µg/l	1	H
Cu	<1		µg/l	1	H
Hg	<0.02		µg/l	1	F
Mn	<0.2		µg/l	1	H
Ni	1.62	0.53	µg/l	1	H
Pb	<0.2		µg/l	1	H
Zn	<2		µg/l	1	H

Er beteckning: **Flue Gas**
Labnummer: U10284768

Parameter	Resultat	Mätosäkerhet (±)	Enhet	Metod	Utf
Filtrerad	NEJ			1	V
Ca	35.0	4.2	mg/l	1	E
Fe	0.0268	0.0069	mg/l	1	H
K	3.59	0.46	mg/l	1	E
Mg	4.28	0.53	mg/l	1	E
Na	902	109	mg/l	1	E
S	60.6	8.9	mg/l	1	E
Al	89.4	16.5	µg/l	1	H
As	25.7	7.0	µg/l	1	H
Ba	55.3	9.5	µg/l	1	H
Cd	0.910	0.155	µg/l	1	H
Co	0.422	0.183	µg/l	1	H
Cr	3.21	1.07	µg/l	1	H
Cu	32.7	5.6	µg/l	1	E
Hg	5.64	0.37	µg/l	1	F
Mn	28.6	3.6	µg/l	1	E
Ni	7.20	1.35	µg/l	1	H
Pb	46.0	7.8	µg/l	1	H
Zn	387	61	µg/l	1	E

Figure A-1: Scan of chemical analysis results for:
-Content of MD-Treated Flue Gas Condensate (mainly cations except for S)-*First table in the figure*
- Content of Raw Flue Gas Condensate (mainly cations except for S)-*Second table in the figure*

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Er beteckning **Product Water**

Labnummer U10284769

Parameter	Resultat	Mätosäkerhet (±)	Enhet	Metod	Utf
Filtrerad	NEJ			1	V
Ca	0.297	0.040	mg/l	1	E
Fe	<0.004		mg/l	1	H
K	<0.5		mg/l	1	E
Mg	<0.09		mg/l	1	E
Na	0.175	0.182	mg/l	1	E
S	0.337	0.066	mg/l	1	E
Al	4.67	5.55	µg/l	1	H
As	<1		µg/l	1	H
Ba	<0.2		µg/l	1	H
Cd	<0.05		µg/l	1	H
Co	0.105	0.103	µg/l	1	H
Cr	<0.5		µg/l	1	H
Cu	<1		µg/l	1	H
Hg	<0.02		µg/l	1	F
Mn	<0.2		µg/l	1	H
Ni	2.55	0.66	µg/l	1	H
Pb	<0.2		µg/l	1	H
Zn	<2		µg/l	1	H

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Figure A-2: Scan of chemical analysis results for:
- MD product water content (mainly cations except for S)-Municipal feed water

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Projekt
Bestnr
Registrerad 2006-11-06
Utfärdad 2006-11-20

Vattenfall Utveckling AB
Anders Wik

162 87 Stockholm

Analys av vatten

Er beteckning: **MD product**

Labnummer: O10127016

Parameter	Resultat	Enhet	Metod	Utf
klorid	1.5	mg/l	1	1
sulfat	<0.50	mg/l	2	1
ammonium	49	mg/l	3	1
alkalinitet	180	mg HCO3/l	4	2
pH	8.7		5	2
konduktivitet	23.3	mS/m	6	2

Er beteckning: **Flue gas**

Labnummer: O10127017

Parameter	Resultat	Enhet	Metod	Utf
klorid	1490	mg/l	1	1
sulfat	168	mg/l	2	1
ammonium	115	mg/l	3	1
alkalinitet	220	mg HCO3/l	4	2
pH	8.4		5	2
konduktivitet	525	mS/m	6	2

Er beteckning: **Product water**

Labnummer: O10127018

Parameter	Resultat	Enhet	Metod	Utf
klorid	<0.60	mg/l	1	1
sulfat	<0.50	mg/l	2	1
ammonium	<0.025	mg/l	3	1
alkalinitet	1.4	mg HCO3/l	4	2
pH	5.9		5	2
konduktivitet	<1.0	mS/m	6	2

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Figure A-3: Scan of chemical analysis results for:
- Content of MD-Treated Flue Gas Condensate (mainly anions except for Ammonium)-*First table in the figure*
- Content of Raw Flue Gas Condensate (mainly anions except for Ammonium)-*Second table in the figure*
- MD product water content (mainly anions except for Ammonium)-Municipal feed water-*Third table in the figure*

B Scale Composition; SEM-EDS Analysis

The following figure shows the chemical composition of the scale formed at the membrane surface, feed inlets and feed distribution channels inside the MD module

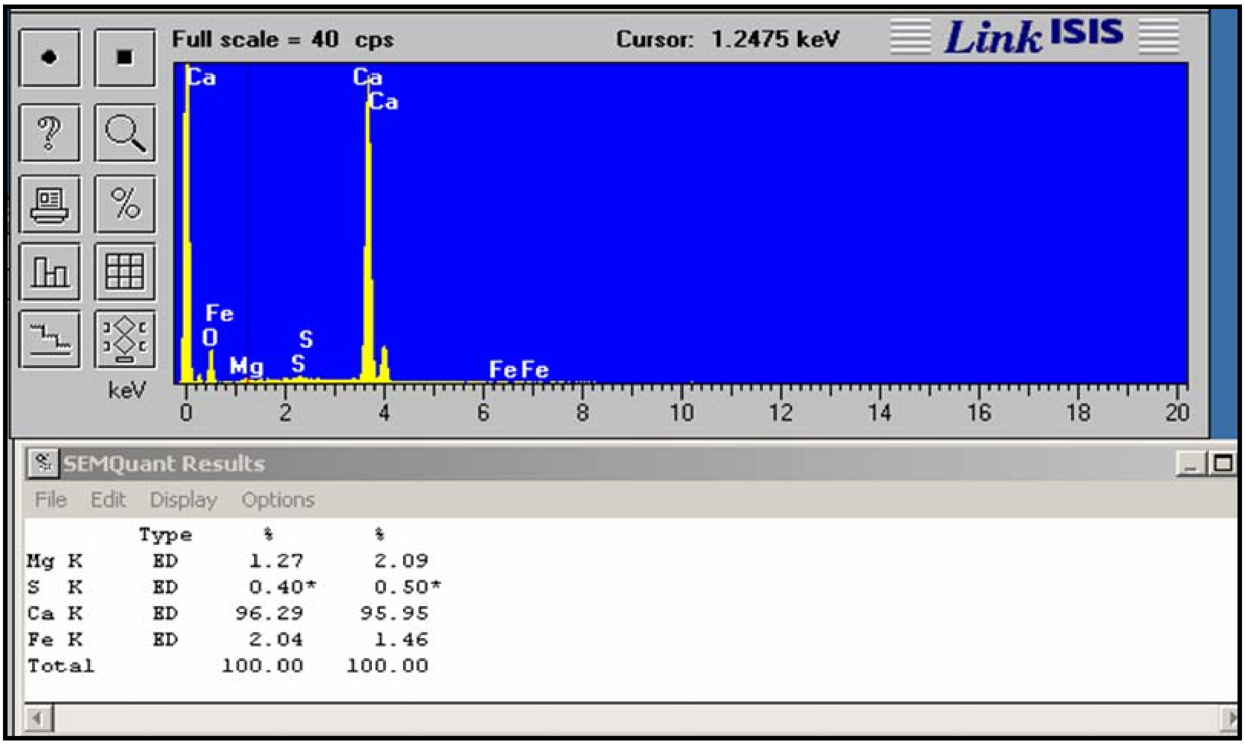

Figure B-1: SEM-EDS analysis of the scale content

C Cost Estimation

Cost estimation was based on the same calculation figure used in the prestudy [1]. The assumptions made for the economical analysis are shown below (comments in brackets are added):

- Cogeneration facility operated 8000 h/yr.
- Demineralized water production is 10 m^3/h (nominally).
- Heat sources/sinks are described in previous chapter.
- The net present value and internal rate of return is 7 %.
- Membrane lifetime is 5 yr, other equipment lifetime is 15 yr.
- Combined unforeseen expenses (i.e. margin of safety) and gross profit is 50%.
- The annual maintenance cost is 5% of the installed cost.
- Membrane price is 290 SEK/m^2.
- Electricity price is 0.5 SEK/kWh, and heat energy price is 0.4 SEK/kWh (taxes excluded).
- Construction price is 350 SEK/hour.
- Cost of control system for MD unit is 90 000 SEK
- The lifetime of steam line in Alternative 3 is 20 years.
- System heat losses are 5%. (Update: Heat losses in this case was included in module simulation)
- Required steam flow rate (5-10 kg/s) does not significantly impact plant operations.
- Costs related to increased space demands not included.
- Variable cost includes membrane removal and replacement at five year intervals.

In the current study, the cost estimates are presented below:

Case I

Item	Quantit y	Unit Cost SEK	Net Cost (MSEK)
Membrane	2392 m^2	290 SEK/m^2	0,69
MD modules	1040	5000	5,20
Heat Exchangers PH1	10	40000	0,40
Heat Exchangers PH2	10	30000	0,30
Pump	20	30000	0,60
Water tank	1	20000	0,02
Piping	1400 m	600 SEK/m	0,84
Valves	200	2000	0,40
Temperature Indicators	50	2300	0,12
Pressure indicators	50	2800	0,14
Security system	4	5000	0,02
Control system			0,02
Construction	2240 h	350 SEK/h	0,78
Others			0,30
Subtotal			9,83
50% margin of error/profit			2,50
Total investment			**12,33**

Table C-1: Investment cost of Case I

Item	Quantity	Unit Cost SEK	Net Cost (MSEK)
Pump electricity	120 MWh	500	0,06
Heat energy	440 MWh	400	0,18
Maintenance cost			0,10
Variable cost			0,10
Total annual cost			**0,44**

Table C-2: Annual cost of Case I

Case II

Item	Quantit y	Unit Cost SEK	Net Cost (MSEK)
Membrane	1141 m^2	290 SEK/m^2	0,33
MD modules	496	5000	2,48
Heat Exchangers PH1	5	40000	0,20
Heat Exchangers PH2	5	30000	0,15
Pump	10	30000	0,30
Water tank	1	20000	0,02
Piping	700 m	600 SEK/m	0,42
Valves	100	2000	0,20
Temperature Indicators	25	2300	0,06
Pressure indicators	20	2800	0,06
Security system	4	5000	0,02
Control system			0,02
Construction	1120 h	350 SEK/h	0,39
Others			0,30
Subtotal			4,95
50% margin of error/profit			2,50
Total investment			**7,45**

Table C-3: Investment cost of Case II

Item	Quantity	Unit Cost SEK	Net Cost (MSEK)
Pump electricity	56 MWh	500	0,03
Heat energy	960 MWh	400	0,38
Maintenance cost			0,05
Variable cost			0,07
Total annual cost			**0,53**

Table C-4: Annual cost of Case II

The specific water cost for Case I, Case II and Alternative 1 (Prestudy) are summarized below in table C-5:

	Specific water costs, low end estimate (SEK/m^3)	**Specific water costs, high end estimate including 50% uncertainty/profit (SEK/m^3)**
Case I	19	22,5
Case II	14	17
Alternative 1	11*	14,4 *

Table C-5: Comparison of specific production cost

* Cost adjusted to account for heat losses

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