DISTRICT HEAT-DRIVEN WATER PURIFICATION VIA MEMBRANE DISTILLATION

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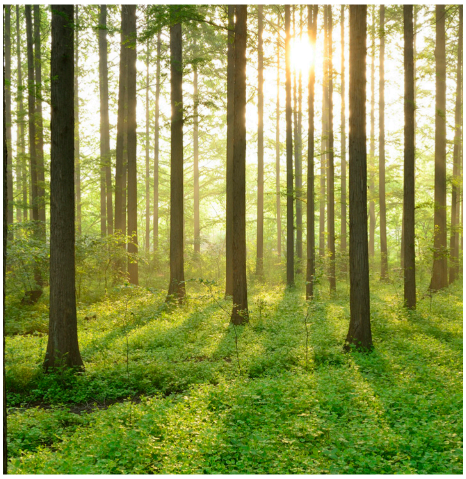

FJÄRRSYN

Energiforsk

DISTRICT HEAT-DRIVEN WATER PURIFICATION VIA MEMBRANE DISTILLATION

New possibilities for applications in various industrial processes

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FÖRORD

Den här rapporten har som syfte att studera om membrandestillering, så kallad MD-teknik, kan appliceras i relevanta industriprocesser med fjärrvärme som drivkälla.

Projektet har genomförts av Andrew Martin, Daniel Woldemarim, Ershad Khan och Alaa Kullab på KTH, Centre for Sustainable Communications. Projektet har också haft ett bra stöd av referensgruppen som har bestått av Rickard Pellny från Övik Energi AB, Jan Baldefors från Svensk Fjärrvärme, Eva-Katrin Lindman från Fortum AB, Erik Dahlquist från Mälardalens Högskola, Marie Cesar från Kraftringen AB, Gunilla Jacobsson från Vattenfall AB och Aapo Sääsk från Scarab Development AB.

Projektet ingår i forskningsprogrammet Fjärrsyn, som finansieras av Svensk Fjärrvärme och Energimyndigheten. Forskningen inom Fjärrsyn ska stärka fjärrvärme och fjärrkyla, uppmuntra konkurrenskraftig affärs- och teknikutveckling och skapa resurseffektiva lösningar för framtidens hållbara energisystem till nytta för fjärrvärmebranschen, kunderna, miljön och samhället i stort.

Membrandestillering är en värmedriven process som utnyttjar partialtryckdifferenser över ett mikroporöst, hydrofobiskt membran för vattenrening. Då tekniken kan drivas vid ett brett temperaturintervall, 50-90 °C, är den intressant för fjärrvärmetillämpningar. MD-tekniken kan innebära lägre el- och värmeanvändning jämfört med befintliga vattenreningsprocesser baserade på membranteknik. Då MD innebär mindre mekanisk påfrestning på membranen kan tekniken leda till en ökning i värmeunderlag med ökad elproduktion i fjärrvärmeverk som följd.

Industrins egna miljö- och hållbarhetsmål är en bidragande orsak till att man vill öka användningen av fjärrvärme. Fossil produktion ska minimeras och elanvändning ska reduceras i samband med att man gör översyner av egna processer. MD- tekniken är en sådan process där man minskar sin elanvändning och ökar sin fjärrvärmeanvändning. Detta är något som industrin strävar efter för att få mer hållbara processer i industrin. Projektet förväntas bidra till att utveckla nya tillämpningar för fjärrvärme inom industrin.

Jan Berglund
Ordförande i Svensk Fjärrvärmes teknikråd

Rapporten redovisar projektets resultat och slutsatser. Publicering innebär inte att Fjärrsyns programråd, Svensk Fjärrvärme eller Energiforsk har tagit ställning till innehållet.

SAMMANFATTNING

Vattenrening är en viktig separationsprocess som förekommer i en mängd olika industritillämpningar. Kraven på processvattenkvaliteten varierar beroende på tillämpningarna, men trenden med ökad återföring av procesströmmar i kombination med strängare regler mot utsläpp kommer att öka behovet av vattenrening i framtiden. Membranteknik som omvänd osmos, ultrafiltrering och nanofiltrering används ofta. Men även om dessa processer är väl utvecklade finns det ett behov av att undersöka annan teknik som kan leda till mer robust prestanda, lägre energiförbrukning och mindre miljöbelastningar samt lägre kostnader. Membrandestillering, så kallad MD-teknik, är ett sådant alternativ. Det är en värmedriven process som lämpar sig väl till fjärrvärme där en hög renhetsgrad önskas.

Syftet med projektet är att utveckla MD-teknikens roll i fjärrvärmeperspektivet. Följande tre delmål har behandlats:

Delmål 1 handlande om kartläggning av vattenrening i relevanta industriprocesser. Olika möjliga tillämpningsområden identifierades, inklusive rening av rökgaskondensat, ultrarent vatten, processer i livsmedelsindustrin, uppkoncentrering, avsaltning med flera. Tre fallstudier av intresse för fjärrvärmedriven MD-teknik identifierades för vidare analys.

Delmål 2 omfattade systemanalyser och optimeringsstudier för fallstudierna:

- Rening av avloppsvatten från Astra Zeneca i Södertälje. Det vore teoretiskt möjligt att sätta MD-tekniken mellan fjärrvärmenätet och slutanvändarna för att åstadkomma avancerad rening hos en mindre avloppsström. Fjärrvärmebehovet skulle öka med omkring 7-13% och kostnaderna (exklusive omkopplingar) uppskattas öka med 60%, jämfört med dagens reningsteknik (aktiv kol).
- Uppkoncentrering av etanol i CO_2-skrubbervatten hos Agroetanol i Norrköping. Konceptet handlar om avlastning av destillationskolumnen genom införande av MD-tekniken. Utbytet mellan ånga (destillationskolumnen) och fjärrvärme (MD-systemet) är ungefär lika, vilket innebär stora möjligheter för kostnadsbesparingar med MD-tekniken.
- Vattenrening och uppkoncentrering hos Arla Foods, Kalmar-mejeriet. En svag värmeintegration innebär stora fjärrvärmebehovför MD-tekniken i just denna tillämpning

I Delmål 3 undersöktes två olika MD-prototyper experimentellt. En nyare generation av MD-prototyp visade en väsentlig minskning i värmeförluster med ökat utbyte, vilket leder till lägre energibehov.

SUMMARY

Water purification is an important separation processes that can be found in various industrial applications. Process water quality varies between applications, but the trend of increased reuse or recycling of process streams in combination with stricter emission regulations lead to additional needs for water purification in the future. Membrane technology such as reverse osmosis, ultrafiltration and nanofiltration are often employed. However even if these technologies are mature there is still a need to explore alternatives that can lead to more robust performance, lower energy consumption, reduced environmental effects, and lower costs. Membrane Distillation (MD) is such an alternative. The process is heat driven and can be paired up with district heating when a high degree of purity is desired.

The purpose of this project is to develop MD's roll within a district heating perspective. The following three sub-goals have been investigated:

Sub-goal 1 considered a mapping of water purification in relevant industrial processes. Various possible application areas were identified, including treatment of flue gas condensate, ultrapure water, processes in the food industry, reconcentrating, desalination, and others. Three case studies with relevance to MD driven by district heating were identified for further analysis.

Sub-goal 2 encompassed system analyses and optimization studies for the particular case studies:

- Wastewater treatment at Astra Zeneca in Södertälje. It is theoretically possible to place MD between the district heating network and end users in order to achieve advanced treatment for a smaller wastewater line. The district heating demand would increase by about 7-13%, and costs (excluding retrofitting) are estimated to increase by 60% compared to the available treatment method (activated carbon).
- Reconcentrating of ethanol from CO_2 scubber water at Agroetanol in Norrköping. The concept involves off-loading of the distillation column via introduction of MD technology. Exchange between steam (distillation column) and district heating (MD system) is roughly balanced which implies significant cost savings potential with MD.
- Water purification and reconcentrating at Arla Foods, Kalmar facility. A poor thermal integration yields a high district heating demand for MD in this particular application.

For sub-goal 3 experimental investigations were conducted for two MD prototypes. A newer generation of MD prototype showed a marked reduction in heat losses with increase in yield, which in turn leads to lower energy demand.

UTÖKAD SAMMANFATTNING

Introduktion och mål

Vattenrening är en viktig separationsprocess som förekommer i en mängd olika industritillämpningar. Pannor, fjärrvärmenät och annan utrustning inom kraftvärme eller lokalvärme har stora behov av ultrarent (dvs mycket högkvalité) vatten. Halvledarindustrin och läkemedelsindustrin ställer också höga krav på vattenkvalitet i sina respektive processer. Stålindustrin, pappers- och massaindustrin samt andra tillverkningsindustrier använder stora mängder av vatten. Processvattenkvaliteten variarar mellan dessa industrier, men trenden med ökad återföring av procesströmmar i kombination med strängare regler mot utsläpp kommer att öka behovet av vattenrening i framtiden. Industrins uttag av vatten uppgick till ca 1,7 miljarder m^3 under 2010, ungefär två-tredjedelar av den totala svenska uttaget för detta år (källa: SCB). Även om bara en mindre del av denna ström kräver extra uppmärksamhet när det gäller reningsgrad är potentialen för ny, förbättrad vattenreningsteknik mycket stor.

Ett antal tekniker för vattenrening har utvecklats genom åren. Många av dagens kommersiella metoder är baserade på membranteknik, till exempel omvänd osmos (RO), ultrafiltrering och nanofiltrering. Även om membranmetoder är beprövade finns det vissa nackdelar. De arbetar ofta med höga tryckskillnader (framförallt för RO) vilket skapar stora mekaniska belastningar som kan minska membranets prestanda och livslängd, särskilt för orena råvaror. Vissa högkoncentrerade strömmar kan inte hanteras av sådana tekniker på grund av praktiska begränsningar (till exempel övre tryckgränsen för RO). Dessa metoder kräver relativt stora mängder el (pumparbete) samt komplexa förbehandlingssteg och styrsystem. Alternativa membranprocesser som behåller fördelarna av traditionell membranteknik men har utökad robusthet, lägre elförbrukning och enklare processintegrering är därför av stort intresse.

Membrandestillering (MD) är en lovande teknik i sammanhanget. MD är en värmedriven process som utnyttjar ett mikroporöst, hydrofobiskt membran. Strömmen från vilken vattnet ska avskiljas värms upp till ca 50-90°C och leds till den ena sidan av membranet. Vattnet (och eventuella flyktiga ämnen) förångas och tränger genom membranets porer, medan vätskan hålls bort. Därefter passeras ångan genom en tunn luftspalt innan kondensering sker på en kylplatta. Metoden benäms Air Gap MD till skillnad från Direct Contact MD, där ångan kondenseras direkt i en kall renvattenström. Egenskaperna hos MD-tekniken är: (1) 100% (teoretiskt) separering av joner, makromolekyler, kolloider, celler och andra icke-flyktiga ämnen; (2) lägre driftstemperaturer än konventionella destilleringsmetoder; (3) lägre arbetstryck än konventionella tryckdrivna membranprocesser och (4) låga mekaniska påfrestningar tack vare en mycket låg tryckskillnad över membranet.
Eftersom MD-tekniken drivs av relativt låga temperaturer är fjärrvärme en ideal

värmekälla. Efterfrågan på fjärrvärme i traditionell bemärkelse kommer sannolikt att minska i framtiden. T ex visar en publicerad prognos på en 10%-ig minskning i efterfrågan 2025 jämfört med nivån från 2007[1]. Genom innovativ integrering av MD-teknik i fjärrvärmesystem kan vattenrening drivas av fjärrvärme, och således ge fjärrvärmebranschen en möjlighet att vidga marknaden samtidigt som utsikterna till kostnads- och energieffektiv vattenrening är stor.

MD-teknikens prestanda gynnas av höga källtemperaturer (upp till 90 °C) och låga kyltemperaturer, men processen kan även drivas med andra temperaturförhållanden. Till exempel är det fullt möjligt att värmeväxla MD-processen med en lågtemperaturström från fjärrvärmereturlinjen, även vid temperaturnivåer där nuvarande efterfrågan är lågt. MD-processen kan också integreras mellan fjärrvärmenätet och den tilltänkta värmesänkan. Det vill säga att MD-processen förses med temperaturer omkring 80-90 °C och kylar ned den till ca 50-60 °C. På detta vis får kunden rent vatten samtidigt med värmeleveransen.

MD-tekniken har inte tidigare studerats i fjärrvärmesammanhang varför det är motiverat att undersöka dennes roll som en energieffektiv, miljövänlig och ekonomisk vattenreningsprocess i industrin. Resultatnyttan finns inom utökning av fjärrvärmeunderlaget genom nya tillämpningar, förbättrad industriprocesser med lägre kostnader och förbättrad miljöprestanda, och utveckling av MD-tekniken genom anpassning till fjärrvärmebranschen.

Huvudmålet med detta projekt har varit att utveckla MD-tekniken som en ny vattenreningsprocess i industrin med fjärrvärme som drivkraft. Projektet har utförts under perioden 2013-2015 och då berört tre delmål (metoder i parentes):

- Delmål 1. Kartläggning av vattenrening i relevanta industriprocesser (litteratursökning, kontakt med industrier och företag)
- Delmål 2. Systemanalyser och optimeringsstudier (material- och energibalanser med input från respektive industrier kopplad till experimentella undersökningar av MD-tekniken)
- Delmål 3. Komponentstudier med fokus på fjärrvärmeanpassning av MD-tekniken (experimentella undersökningar av två olika MD-prototyper)

Nedan sammanfattas resultaten av arbetet.

<u>Kartläggning av vattenrening i relevanta industriprocesser</u>
En rad tillämpningar har identifierats som relevant för MD-tekniken, men har sedan <u>inte</u> undersökts i mer detalj:

- Rening av rökgaskondensat: har studerats tidigare inom ramen för Värmeforsk och ett nyligen avslutat projekt mellan IVL och KTH. Anses inte centralt till fjärrvärmedistributionen.

[1] Se huvudrapporten för källor.

- Avskiljning av läkemedelsrester från renat avloppsvatten. Försök på Hammarby Sjöstadsverk har visat överlägsen prestanda gällande separationsverkningsgrad för ett dussintal substanser. Tillämpningar är dock inte aktuellt med hänsyn till de stora vattenvolymer som måste hanterats vilket gör MD-tekniken för kostsam (stor antal moduler, stor membranyta).
- Ultraren vattenproduktion omfattande pannvatten, spädvatten, spolvatten i halvledarindustrin och processvatten i läkemedelsindustrin. Kraftvärmetillämpningar ligger utanför projektets ramar och halvledartillverkning finns inte längre i Sverige. Enbart RO får användas som reningssteg för processvatten i läkemedelsindustrin på grund av rådande standard (farmakopén), vilket därmed utesluter MD-tekniken.
- Livsmedelindustrin exklusive mejeriindustrin.Inga tillämpningar hittades i Sverige.
- Uppkoncentrering av icke-flyktiga syror Dessa finns i gruv- och stålindustrin, fast inga tillämpningar hittade i Sverige.
- Avsaltning: stor intresse utomlands i Mellanöstern till exempel, men inte aktuellt för Sverige. (Det har skrivas om avsaltningsanläggningar på Gotland, men kontakt med kommunen har inte besvarats.)
- Andra tillämpningar. Skärvätskor (experimentella försök misslyckades), avloppsvatten från sjukhus (bedöms vara svåra att hantera ur hygieniska och toxiska perspektiv) och vattenrening i simhallar (intressant, men idén kom in för sent i projektet).

De tillämpningar som bedöms mest lämpliga för vidare studier består av följande:

- Avloppsrening hos AstraZeneca i Södertälje
- Uppkoncentrering av etanolhaltigt skrubbervatten hos Agroetanol i Norrköping
- Ersättning av olika reningssteg hos Arla Foods mejeri i Kalmar

Dessa industrier har även visat ett stort intresse för tekniken, och kopplingen till fjärrvärme.

<u>Fallstudie: AstraZeneca</u>

AstraZeneca tillverkar en rad olika läkemedel (piller, inhalatorer mm) vid Gärtuna. Spol- och tvättvatten leds till två avloppsströmmar. Huvudavloppströmmen på 33 m^3/tim leds bland annat genom ett avancerat (svampbaserat) biologisk reningssteg, medan en mindre avloppsström på 3 m^3/tim behandlas med aktiv kol för att ta hand om rester från olika magmediciner (som annars ställer till problem för biologisk rening vid högre koncentration). Båda strömmarna blandas för vidare behandling.

Den mindre avloppströmmen är av intresse för MD-tekniken för att kunna ersätta aktiv kol; på så vis skulle man nå så kallad ”zero liquid discharge” med MD-tekniken om koncentratet tas om hand genom destruering. Huvudavloppströmmen behöver uppvärmning från 15 till 25 °C, vilket för nuvarande sker genom fjärrvärmen. Med MD-teknik är det möjligt att i stället tillgodose detta värmebehov genom att sätta MD-systemet mellan fjärrvärmenätet och denna ström så att kylvärmen från MD-enheten används ytterligare. Vidare kunde byggnader och processer värmas med MD-returen, särskilt om värmebehovet för vattenreningen är stort.

Under 2014 köpte AstraZeneca Gärtuna 20 205 MWh fjärrvärme inklusive uppvärmning av huvudavloppsströmmen (3 240 MWh). I projektet har en potentialbedömning utförts vilken visar på ett årligt värmebehov (fjärrvärme) för MD-systemet mellan 17 600 – 21 600 MWh beroende på modultyp (Xzero respektive Elixir). Därmed bedöms energianpassning som god åtminstone från ett teoretiskt perspektiv. Kostnaden för vattenreningen exklusive nödvändiga omkopplingar uppskattas mellan 37 och 77 SEK/m^3 (Elixir respektive Xzero), som kan jämföras med dagens kostnad för aktiv kol (inklusive avfallshantering), 23 SEK/m^3. Resultaten är lovande för Elixir-modulen.

Fallstudie: Agroetanol
Agroetanol producerar 250000 m^3/år teknisk sprit från spannmål och olika jordbruksrester. Processen som är aktuellt för MD-tekniken handlar om uppkoncentrering av skrubbervattnet från koldioxavskiljaren (6-12 m^3/tim, 5% etanol/95% vatten). En ökad etanolhalt i denna ström innebär en mindre last på destillationskolumnen, vilket i sin tur leder till ett minskat ångbehov (processången levereras av E.ON Norrköping). MD-tekniken har här studerats för uppkoncentrering av etanol, och labboratorieförsök (se nedan) visade positiva resultat.

För energianalysen antogs ett flöde på 9 m^3/tim med uppkoncentrering av etanol från 5% till 77%, med temperaturerna begränsade till 40 °C för att säkerställa en relativ hög separationsverkningsgrad (att nå närmast 100% etanol går inte med ett en-stegs MD-system). Det årliga fjärrvärmebehovet för denna processström beräknas då till 6300 MWh, vilket kan jämföras med uppskattat ångbehov av 5 600 MWh hos destillationskolumnen. Man kan därmed konstatera att MD-tekniken är utbytbar gällande värmebehovet, och finns det potential för årliga värmekostnadsbesparingar på över 1 MSEK (priser för ånga och fjärrvärme antas som 800 SEK/MWh respektive 500 SEK/MWh). Återbetalningstid för MD-utrustningen uppskattas till cirka ett år. Det även finns positiva aspekter för energileveratören E.ON-on: ökad elproduktion genom minskade ångleveranser och försäljning av (lågtemperatur) fjärrvärme.

Fallstudie: Arla Foods
Arla Foods liksom andra processindustrier vill förbättra sin miljöprofil och användning av fjärrvärme anses positiv eftersom andel förnybar energi ökas. Anläggningen i Kalmar, som är kopplad till fjärrvärmenätet tar emot 160 000 ton/år mjölk för att producera olika mjölkkvalitéer, ost och vassle. Två möjligheter har identifierats för MD-tekniken Dels uppkoncentrering av vasslen, dels

utbyte av ett RO-poleringssteg i processen. Brineströmmen som finns kopplad till ost-tillverkningen kunde också utnyttjar MD-tekniken.

Energianalysen visar dock en alldeles för hög fjärrvärmeförbrukning när MD-tekniken sätts in i systemet. Detta beror på svårigheter att integrera MD mellan fjärrvärmenätet och de olika värmekrävande processerna. Även med värmeintegrering ligger MD-systemets årliga fjärrvärmebehov på nästan 10 000 MWh för båda ovanstående tillämpningar. Då blir MD-tekniken alldeles för kostsamt i förhållande till andra metoder.

Komponentstudier och experimentella undersökningar

Experimentella undersökningar har också genomförts inom projektet. Här har två olika MD prototyper testats i KTH:s energiteknik lab. Båda använder samma upplägg med kassett/membran. Den ena, HVR, är en äldre prototyp med en tjock aluminium kylplatta.Den andra, Elixir, är en ny prototyp med en kylplatta av tunnmetall, liknande material som finns i plattvärmeväxlare.

Modulernas prestanda karakteriserades genom försök med olika inloppstemperaturer på varm respektive kall sidan, kyltemperaturer och flöden. Utbytet ökar med ökade inloppstemperatur på den värma sidan och högre temperaturskiljnader mellan varma och kalla sidan. Elixir-modulens prestanda ($m^3/hr\text{-}m^2$) ligger alltid över prestandan för HVR-modulen, med upp till 22% högre utbyte. Även energiprestandan är fördelaktigt med Elixir-modulen. Värmebehovet är likvärdigt för båda modulerna (cirka 1000 kWh/m^3). Elixir-modulens nettovärmebehov är omkring 36 kWh/m^3, vilket är ungefär hälften av HVR- modulens värden. Skillnaden i prestandan kan härledas till den förbättrade kylplattedesignen.

En jämförelse med Xzero-modulen som är en fullskalig kommersiell prototyp, visar att Elixir-modulens design har klara fördelar i både utbyte och energiprestanda. Dessa fördelar har visats sig i uppskalningen som presenterats tidigare.

Slutsatser

Inom ramen för projektet har en rad industriella tillämpningar för fjärrvärmedriven MD-teknik undersökts. Av många alternativ har tre fördjupade fallstudier utförts, där resultaten belyser fördelar och kostnadsbesparingar med MD-tekniken, men även situationer där tekniken inte är lämplig. Vidare har projektets experimentella utvecklingsdel visat på en positiv utveckling av MD-modulens prestanda. Därmed är projektens mål uppnådda.

Av de tre fallstudier är Agroetanol mest lovande eftersom MD-tekniken kan leda till besparingar för industrin och ökad användning av fjärrvärme. Dessutom bör praktiska aspekter som värmeväxling och rörkopplingar vara relativt lätta att åtgärda. Fjärrvärme levereras inte till Agroetanol i nuläget, men anläggningen ligger i närheten av E.ON:s avfallsförbränningsanläggning.

Fallstudien med AstraZeneca visar framförallt på möjligheten att förbättra rening av avloppsvatten kopplad till värmeåtervinning gentemot olika värmebehov. Inköp av fjärrvärme skulle öka med 7% med utnyttjande av MD-tekniken. I detta är det inräknat att MD-tekniken integreras med de andra värmebehoven inom området (byggnader och processer). Då används, där det är möjligt, fjärrvärmereturen från MD-systemet till ytterligare värmning i andra delar av process och byggnader, vilket medför att fjärrvärmereturen från AstraZeneca kan minimeras.

Slutligen bedöms utsikter för fjärrvärmedriven MD-tekniken som små för mejeriindustrin eftersom värmeåtgången är alldeles för stor på grund av svag värmeintegrering.

Framöver finns det planer att följa upp projektet genom diskussioner med Agroetanol och AstraZeneca för eventuella försöksprojekt. Vissa tillämpningar som identifierades i kartläggningen (sjukhus, simbassänger) blir mål för nya studier genom till exempel examensarbete. Andra möjligheter såsom koppling till t ex solvärme eller industriell spillvärme är också av intresse för framtida studier.

CONTENTS

ABBREVIATIONS

ABE: Acetone, Butanol and Ethanol

AC: Activated Carbon

AGMD: Air Gap Membrane Distillation

CHP: Combined Heat and Power

DCMD: Direct Contact Membrane Distillation

DDGS: Dried Distillers Grains with Solubles

DH: District Heating

FAO-UN: Food and Agriculture Organization of the United Nations

IVL: Swedish Environmental Research Institute

KTH: Kungliga Tekniska högskola

MD: Membrane Distillation

MEDSOL: Solar-powered Membrane Distillation

MF: Micro Filtration

MSW: Municipal Solid Waste

O&M: Operation and Maintenance

RES: Renewable Energy Sources

RO: Reverse Osmosis

SGMD: Sweep Gas Membrane Distillation

UF: Ultra Filtration

VMD: Vacuum Membrane Distillation

V-MEMD: Vacuum - Multi Effect Membrane Distillation

WBCSD: The World Business Council for Sustainable Development

WDG: Wet Distillers Grains

WTP: Wastewater Treatment Plant

1 INTRODUCTION AND OBJECTIVES

Water and energy have always been intertwined as water is used in virtually all industrial processes, and energy is required to extract and deliver water of appropriate quality for use and to treat wastewater for reuse or safe release to the environment (U.S. Department of Energy, 2014). Second only to agriculture, the industrial sector is one of the largest users of water worldwide (FAO-UN, 2015). Among industries thermal power plants exhibit one of the highest water demands, however many other industrial processes follow closely behind. The amount of water used in industries varies widely from one to another depending on the type of water use and scale of processes. Water can be used as process water in industries such as pulp and paper, textile, and semiconductors, or to enable steam-driven power and for use in various production processes including cooling and cleaning (WBCSD, 2005). In other industries, notably the food, beverage and pharmaceutical sectors, water is an ingredient in finished products for human consumption (Förster, 2014). Examples of such products include dairy products, soups, beverages and medicines that are delivered in liquid form.

Environmental regulations such as the EU Water Directive and other national and local statutes have led industry to recycle more water and to raise standards of effluents, thus adding more complexity and cost to the water treatment methods employed. Various types of large-scale water treatment technologies are in use to supply the quality of water required for the specific purpose. Most of these water treatment processes involve a number of sequential processes like filtration, biological or chemical reaction, adsorption and related procedures. Advanced treatment technologies such as reverse osmosis (RO) are necessary for achieving high separation efficiency and are often used in ultrapure water applications. A drawback of RO is its relatively high specific electricity demand. Hence alternatives featuring similar or better separation efficiency with lower energy impact are of high interest. Membrane distillation (MD) is one such technology. MD combines the benefits of membrane separation with reduced electricity needs, and can be paired to heat sources and sinks in the range of 40-90ºC and 5-50ºC, respectively. This makes MD highly suitable for integration with district heating (DH) networks, which until now has not been investigated despite the fact that there is more than sufficient DH capacity to drive such a process.

The main objective of this investigation is to develop new industrial applications for MD with district heating as the heat source and/or sink. The project encompasses three tasks with respective methodologies:

Task 1: Survey of water purification in relevant industrial processes – literature survey and contact and interviews with Swedish-based industries

Task 2: System analyses and optimization studies – mass and energy balance calculations with input obtained from industry coupled to experimental MD investigations

Task 3: Component studies with focus on tailoring MD to district heating networks – experimental MD investigations for two different prototype modules

This report is divided into the following sections: background with DH overview and additional details on MD technology; survey including a range of possible MD applications and presentation on the specific industries identified; results from three case studies (system analyses) and laboratory experimentation of new MD prototype; and conclusions and future work.

2 BACKGROUND

2.1. District heating: Opportunities and Challenges

District heating is a centralised facility used for heating and distributing water to its consumers by using a network of pipelines. When the heated water reaches the consumer (for example a residential building), the heat is transferred through heat exchangers to supply domestic hot water and radiators for space heating. The centralised heat production within district heating systems can be placed in one or several heat production units such as hot water boilers, heat pumps or combined heat and power (CHP) plants where heat is co-produced together with electricity. Different kinds of heat production processes use a variety of fuels such as; biomass, waste, peat, oil, coal, natural gas and electricity (Eriksson et al, 2007).

DH systems can play an important role in increasing the use of renewable energy sources (RES) for heating purposes, but also in reducing total primary energy demand. Due to economy of scale, the DH systems provide an opportunity of using deep geothermal heat as well as unrefined biomass (e.g. waste wood, straw, forestry residues) and municipal solid waste (MSW). The DH systems also enable the utilisation of surplus heat in industries (industrial waste heat) and thermal power plants via combined heat and power production (CHP), thus reducing primary energy demand. In Europe, DH systems are particularly common in Sweden, Finland, Denmark, the Baltic countries and Eastern Europe.

For 2013-2014 the Swedish heating market amounted to 100 TWh per year and 100 billion SEK turnover (Sköldberg & Rydén, 2014). District heating stood for more than half of the total heating demand and a third of the heating was based on electricity (see Figure 1). In terms of total annual turnover, district heating represents 40% of the total and electric heating and heat pumps

45%. District heating was found to be the largest heating category in multi-family houses and premises, whereas electricity based heating was largest in single-family houses (Sköldberg & Rydén, 2014).

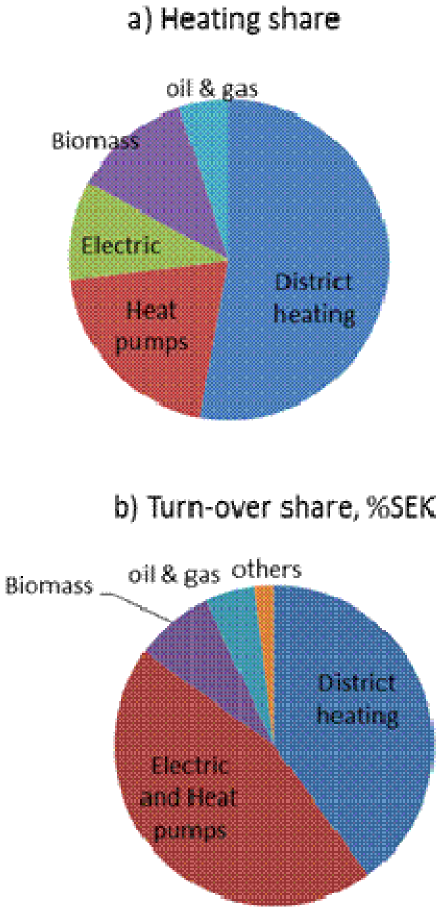


Figure 1. Different heating sources share in terms of Annual heating (a) and total turnover (b) share in the Swedish heating market for the year 2012-2014 (Sköldberg & Rydén, 2014).

As shown in Figure 2, even though district heating has a large share of heating among the energy sources, the annual heating consumption is forecasted to reduce by nearly 10% in 2025 when compared with the estimated consumption for 2007 (Skoldberg, et al., 2011). The main contributor for this decrease is the improvements in energy efficiency of multi-family houses and premises. This is most obvious for building heating, where the specific heat consumption per square meter has been reduced by the magnitude of one percent per year for the period of 1995 – 2012. (Sköldberg & Rydén, 2014). Of course climate change specifically global warming could have significant contribution as well. Yet, there are opportunities for district heating market in Sweden. As it is mostly derived from biomass, district heating is considered environmentally friendly and contributes to the renewable share of Swedish energy composition. The increase in CO_2 taxes on fossil fuels, the green certificate system and subsidies to biomass cogeneration plants will make the district heating market still attractive in the future. As most of the district heat is produced in combined heat and power plants, the development and increase in the supply of heat through district heating technology will also lead to an increase in electric production as a result.

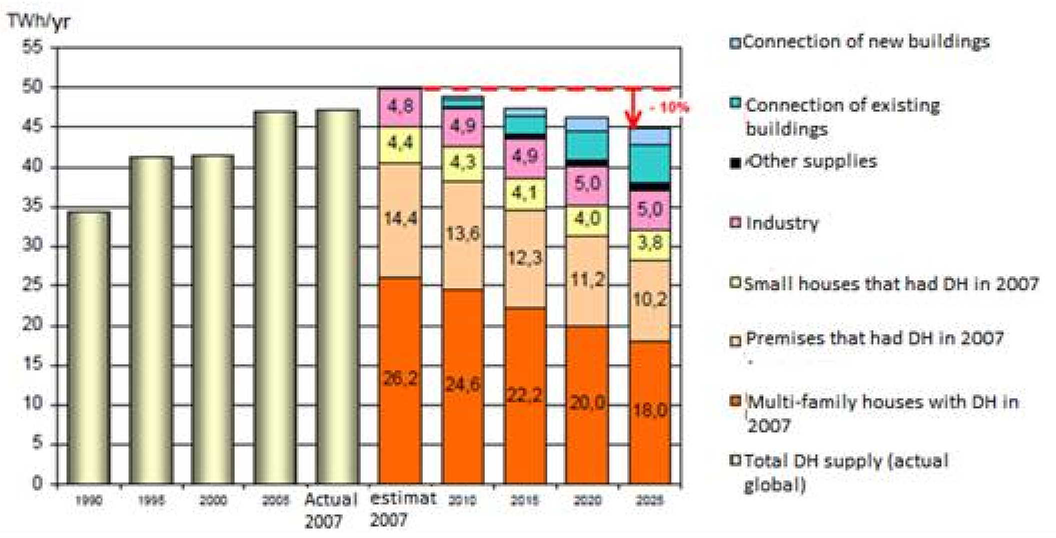


Figure 2.The Swedish district heating supply development according to calculations carried out in project "District heating in the future:the need'' (Skoldberg, et al., 2011).

2.1 Membrane Distillation

Membrane distillation is an emerging thermally-driven, membrane-based separation process in which the hydrophobic microporous membrane separates two solutions with different temperatures. The temperature difference across the membrane causes water vapour pressure gradient, resulting in water vapour transport through the pores from the feed to the cooling side of the membrane. Some of the key advantages of membrane distillation processes over conventional separation technologies are: relatively lower energy costs and reduced vapour space compared to conventional distillation; a complete rejection of dissolved, non-volatile species to produce ultrapure water (electrical conductivity of 5 μS/m); much lower membrane fouling as compared with microfiltration, ultrafiltration, and reverse osmosis; lower operating pressure than pressure-driven membrane processes and lower operatingtemperature (60ºC-90ºC) as compared with conventional evaporation integrated with low grade heat sources such as waste heat in power plants and related industries, solar thermal collectors and geothermal heat sources. The possibility of integration with low grade heat sources enables MD to be installed from small family house for drinking and hot water supply (Kumar & Martin, 2015) to large scale water purification in semiconductor industries (Liu & Martin, 2005) which makes the running cost feasible for such applications.

Four common MD system configurations, as shown in Figure 3, have been in use where the hot feed side must be maintained in direct contact with one side of the membrane in all types. Changes are made only in the permeate side. Direct Contact Membrane Distillation (DCMD), in which the membrane is in direct contact with permeate water. This is the simplest configuration capable of producing reasonably high flux. It is best suited for applications such as desalination and concentration of aqueous solutions. Air Gap Membrane Distillation (AGMD) is the other type module configuration in which an air gap is interposed between the membrane and a condensation surface. The configuration has the highest energy efficiency, but the flux obtained is generally lower. The air gap configuration can be widely employed for most membrane distillation applications, particularly where energy availability is low. In this study, the membrane distillation modules considered in all experimental and theoretical investigation were all based on air gap membrane distillation modules developed by Xzero AB and its sister company HVR AB (Figure 4). Vacuum Membrane Distillation (VMD), in which the permeate side is air under reduced pressure and permeate is condensed in a separate device. This configuration is useful when volatiles are being removed from an aqueous solution. The fourth configuration, Sweep Gas Membrane Distillation (SGMD), involves stripping gas is used as a carrier for the produced vapor. It is used when volatiles are removed from an aqueous solution.

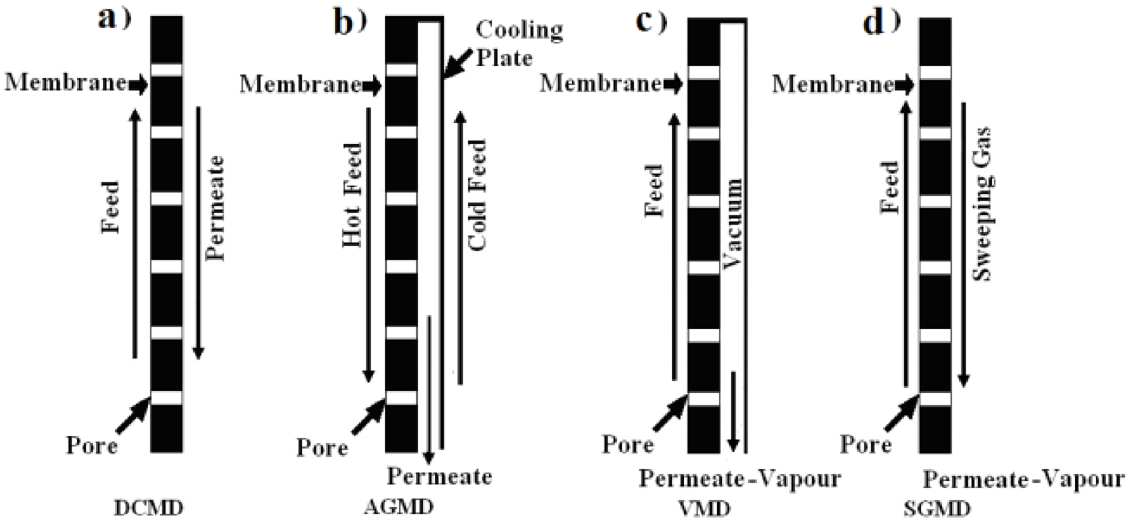
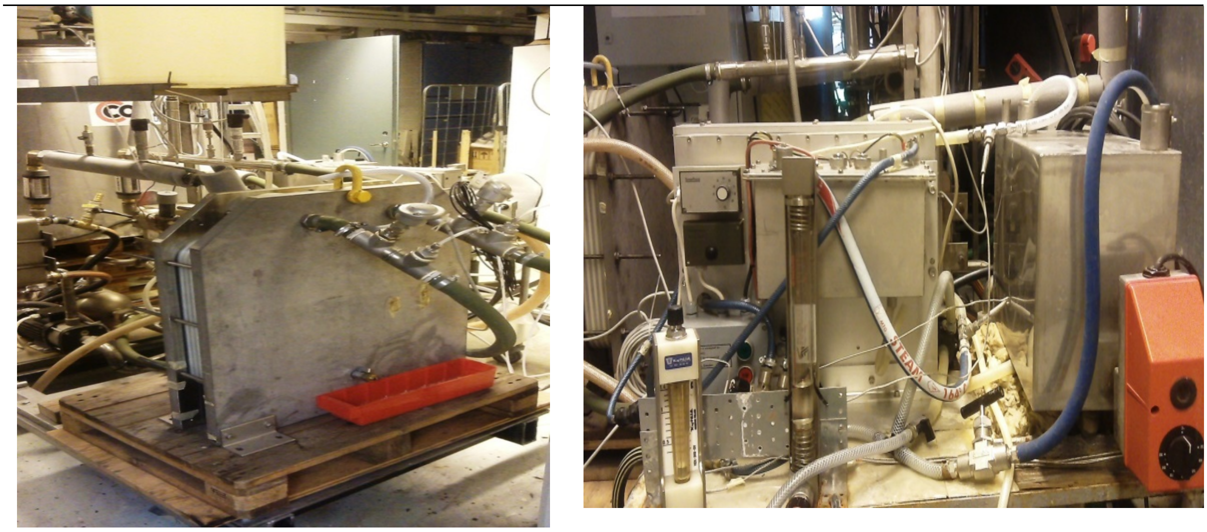


Figure 3. The four most common membrane distillation module configurations (Camacho, et al., 2013).



a) Xzero's 10-cassettes (membrane area of 2.8 m^2) b) HVR lab scale unit (membrane area of 0.23 m^2)

Figure 4. Laboratory scale membrane distillation test systems at KTH Energy Department.

Even though membrane distillation has gained significant interest from an industrial and academic perspective in recent years, there are still challenges in commercialization of the technology mainly due to module design and membrane development issues. There are only a few MD pilot plants all over the world. One such facility is MD pilot plant for research and demonstration at Hammarby Sjöstadsverk in Stockholm which is based on AGMD (Figure 5). The membrane distillation modules are supplied by Xzero AB and are driven by heat from district heating network. The system has a production capacity of 5-10 m^3 pure water per day (Woldemariam, 2014). In February 2014, Aquaver, a Dutch cleantech company also commissioned and installed the first full-scale commercial MD facility for desalination in a small Maldivian Island of Gulhi (Aquaver, 2014). The membrane distillation system supplied by memsys is based on Vacuum-Multi Effect Membrane Distillation (V-MEMD) configuration (Figure 6). It utilizes waste heat of a local power generator and has a production capacity of 10 m^3 drinking water per day.

Figure 5. Xzero membrane distillation system at Hammarby Sjöstadsverk testing and demonstration facility.

Figure 6. Memsys membrane distillation system integrated by Aquaver (Aquaver, 2015)

3. SURVEY OF POTENTIAL MEMBRANE DISTILLATION APPLICATIONS WITH RELEVANCE TO DISTRICT HEATING

3.1. Flue gas condensate treatment

A pilot plant trial of membrane distillation has been investigated previously (Kullab & Martin, 2011) for the treatment of flue gas condensate. The results from this study showed the performance of MD in removing many of the heavy metals and ions present in the condensate with high efficiency up to 99.9% removal except for the ammonia and carbon dioxide which passed through the membrane water vapors as volatiles. Later on, there has been an ongoing research in collaboration between KTH and IVL (Swedish Environmental Research Institute) at Hammarby Sjöstadsverk using Xzero's AGMD modules. The test aims at reducing the pH of the feed so that the ammonia gas in the condensate would be dissolved and leaking through the membrane would be prevented. (A report is expected to be available in early 2016.) This application has not been considered in the present study.

2.1. Removal of pharmaceutical residues from treated wastewater

Treated wastewater contains minute concentrations of a variety of pharmaceuticals, and the elimination of such substances is very difficult even with modern water treatment methods. In 2013 the EU added three pharmaceuticals (two hormones and a pain killer) to a watch list of emerging pollutants; moreover pharmaceuticals are expected to be part of upcoming EU Water Directives (Chatain, 2013; 2008/105/EC, 2015). Separation of pharmaceutical residues with MD was the topic of an investigation conducted by IVL and KTH. MD technology exhibited outstanding separation efficiencies for more than twenty substances (Martin, 2013). Even though results are promising in terms of completely eliminating pharmaceuticals from product water, this application has not been considered owing to the extremely high water volumes involved.

2.2. Ultrapure water production

Ultrapure water (UPW) is required for processes in a variety of fields: thermal power plants and district heating networks (boiler make-up water, district heating make-up water); semiconductor manufacturing (Liu & Martin, 2005) (UPW, 2015); and pharmaceutical manufacturing (Judd & Jefferson, 2003). UPW needs for thermal power plants are outside the scope of the present work. While there is a significant amout of interest in employing MD technology in semiconductor manufacturing (Sääsk, 2013), the industry is no longer present in the Nordic countries. UPW standards for pharmaceuticals require use of RO as a water treatment step, so the viability of MD as a replacement technology is judged to be low.

2.3. Food industry applications excluding dairy

Membrane distillation (DCMD) has also been studied for concentrating of juice products and for the treatment of wastes from the production plant. For treatment of wastewater from Olive mill with a combination of pretreatment processes such as coagulation and flocculation microfiltration (A. El-Abbassi, 2013), for orange juice concentration (Alves & Coelhoso, 2006) by comparing MD with Osmotic Evaporator in terms of product flux. Apple juice concentration was also studied using DCMD (Gunko et al, 2006) and it was shown a concentration of up to 60-65% though with a huge decrease in the product flux. Sweeping gas MD (SGMD) and vacuum MD (VMD) were also shown to be promising techniques for gentle stripping of berry juice aroma compounds with lower energy consumption than the conventional high-temperature evaporation (Bagger-Jørgensen, 2011). None of these applications are judged to be of interest for the Swedish or Nordic industrial sector.

2.4. Concentration and recovery of non-volatile acids

Concentrating used acids and such sulfuric acid, hydrochloric acid and valuable metals from acidic mining wastes are among the potential applications of membrane distillation in not only recovery of important and limited metals but also as part of environmental protection (Tomaszewska, 2000; Kesieme, 2015; Tang & Zhou, 2006). In these studies different DCMD and VMD modules were tested to separate out the acids and other important mineral ions from wastewaters used in mining and recover the water up to 80% recovery. Swedish or Nordic applications of these processes could not be identified.

2.5. Desalination

Membrane distillation has been more extensively investigated for desalination than other applications using different energy sources (e.g. Cipollina, 2012; Qtaishat & Banat, 2013; Schwantes, 2013). Desalination is not a priority water supply technology in the Nordic countries, so this application has not been considered.

2.6. Other applications

Through contacts at IVL cutting fluid wastewater treatment was identified as a potential application. Volvo Trucks in Skövde employs ultrafiltration (UF) and RO to treat cutting fluid effluent. The present wastewater treatment line, while fairly advanced, is not fully capable of removing all organics. A sample of UF-treated cutting fluid wastewater was sent to KTH for testing in a bench-scale MD unit. Results showed a low separation efficiency, which was linked to undesired wetting (i.e. penetration of liquid phase across the membrane). Therefore this application was judged to be not viable.

Skåne University Hospital in Lund has been in contact with Kraftringen Energi AB to explore the possibility of treating small amounts of wastewater associated with cancer treatment. Although promising this application has some complexities in terms of hygiene and toxicity, hence it is suggested that it be considered for future work.

Tekniska verken in Linköping suggested MD as a water treatment technique for use in municipal swimming pools. The suggestion at the latter part of the project period, hence it could not be considered.

3. GENERAL DH INTEGRATION FOR SELECTED CASE STUDIES

As this study is aimed at investigating potential applications of membrane distillation powered by heat from district heating in industries, the systems in focus will consider the integration of the three different processes i.e. the industrial process, membrane distillation and district heating as shown in Figure 7.

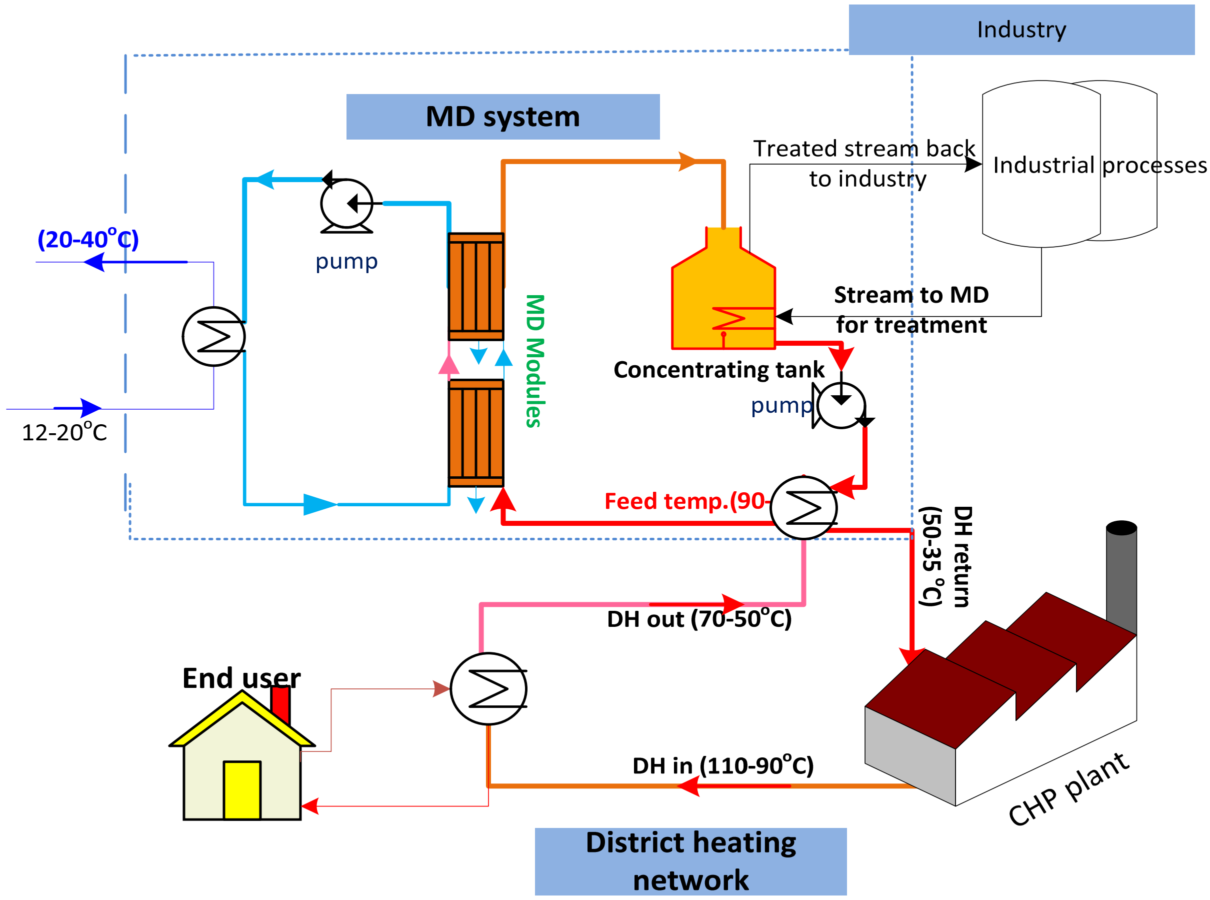


Figure 7. Typical District Heating-Membrane Distillation-Industrial process integration system.

The MD system will be supplied by a stream of solution from the industrial process for separation. The heat will be supplied to the MD system by DH through plate heat exchangers either from the high-temperature line of the district heating or from the lower side of the DH network, depending on the production requirement of the MD. The MD system can provide low-grade heat for subsequent processes by adjusting the cooling loop temperature level. (Use of the DH supply line as a heat source and DH return line as a heat sink is theoretically possible but has not been considered owing to practical limitations.) This arrangement allows for solution reconcentration by making use of the circulation tank and there is a continuous supply of the permeate product which can be reconnected to the plant for further process or reuse. The three case studies considered herein are batch processes that can be integrated with batch MD process where the feed to the MD can be concentrated.

3.1. AstraZeneca Case Study

Wastewater from pharmaceutical plants usually contains very complex mixtures of contaminants including antibiotics, antacids, steroids, antidepressants, analgesics, anti-inflammatories, antipyretics, beta-blockers, lipid-lowering drugs, tranquilizers, and stimulants (Burke, 2008). These are classified as hazardous wastes which are too strong to dispose and are mostly non-degradable; they pose a potential long-term risk for aquatic and terrestrial organisms. In this study membrane distillation is considered for the treatment of wastewater from AstraZeneca Gärtuna pharmaceutical production plant. The analysis is based on both the performance of MD in removing the common pharmaceuticals and system thermo-economy for the capacity required by the waste water treatment plant (WWTP). Contact details for AstraZeneca:

- Peter Gruvstedt, Process Engineer - Wastewater treatment plant, AstraZeneca Sweden Operations, peter.gruvstedt@astrazeneca.com
- Alexis Midestad, Energy Manager, Site Services

Södertälje, AstraZeneca Sweden Operations Hard Service, alexis.midestad@astrazeneca.com

3.2. Agroetanol Case Study

Agroetanol in Norrköping produces fuel ethanol from agricultural wastes and similar feedstocks. In this case study MD will be considered as a technology for recovering water for reuse and ethanol for further dehydration back in the plant. Even though membrane distillation has been rarely studied compared to the well-established and commercialised pervaporation method, it had been claimed to separates ethanol from solution 100 times faster than pervaporation (Benavides-Prada et al., 2013), due to the nature of driving force and enhanced separation by feed and cooling temperatures manipulation. Separation of ethanol from the fermentation broth and ethanol recovery from aqueous solutions has also been studied by a number of researchers at a laboratory scale (Lewandowicz, 2011). Results from the studies showed that membrane distillation (VMD) increases ethanol production by removing the ethanol produced from the fermentation broths continuously, even though membrane wetting at higher ethanol percentage. A theoretical analysis of removing the solvents acetone, butanol and ethanol (ABE) from the fermentation broth by using AGMD was done as a mechanism of increasing fermentation and production (Banat & Al-Shannag, 2000). Membrane distillation system can be applied to remove bioethanol from a reactor and so a possible solution to overcome the disadvantages of conventional fermentation systems by reducing the yeast stress commonly occurring in industrial fermentation. Contact details at Agroetanaol:

- Anders Holmbom, Process Engineer, Lantmännen Agroetanol, Norrköping, Sweden anders.holmbom@lantmannen.com

3.3. Arla Foods Case Study

MD has been considered previously for applications in the dairy process.

Separation of skim milk and also whey filtrations were among the studied processes incorporating MD (Hausmann, et al., 2011, 2014). In these studies, a hybride of MD and FO and RO were considered and proved to be efficient in separation performance and energy saving especialy in the presence of waste heat. Membrane fouling and reduced flux were stated as the drawbacks. Arla Foods utilizes RO in various polishing steps, and there is potential to use MD as a replacement. Contact details at Arla Foods:

- Inger Larsson, Environmental Manager SE Global EH&S, Arla Foods AB inger.larsson@arlafoods.com
- Oskar Räftegård, SP Sveriges Tekniska Forskningsinstitut, oskar.raftegard@sp.se, was approached for discussions related to the different processes and their energy demands at the Kalmar facility.

4. SYSTEM ANALYSIS OF SELECTED MEMBRANE DISTILLATION CASE STUDIES FOR DISTRICT HEATING INTEGRATION

This chapter contains results from system studies conducted on case studies at AstraZeneca, Agroetanol, and Arla Foods. Input data for MD module performance was obtained from experiments conducted at Hammarby Sjöstadsverk (Xzero modules) and KTH (Elixir modules).

4.1. Wastewater Treatment at AstraZeneca

4.1.1. Background on Current Process

Production facilities in Sodertalje (Snäckviken and Gärtuna) are home to AstraZeneca's biggest global tablet and capsule manufacturing plants. The wastewater treatment plant considered in this study is found in Gärtuna.

The AstraZeneca Gärtuna facility as a whole uses district heat, steam and also cooling to maintain climate control such as humidity and temperature. The ventilation is also crucial in order to keep a clean environment in favor of medicine production quality and a healthy working environment. To provide heat to the climate control, AstraZeneca not only imports heat from the power plant Igelstaverket but also produces its own heat with heat pumps and condensate heat exchangers. The steam is directly imported from Igelstaverket and the cooling is produced in different buildings operated by both AstraZeneca and Caverion, within the facility (Björk, 2015). All electricity is produced outside of the facility and is bought from the company Telge Energi.

The treatment process for wastewater from pharmaceutical plants is mostly done by decomposing the organic compounds in bioreactors and using activated sludge treatment. However, these processes are not efficient enough to fully remove the pollutants and hence additional series of treatment steps are required such as activated carbon treatment, advanced oxidation, photolysis and nanofiltration.

In the pharmaceutical WWTP considered in this study, there are two separate wastewater streams. The one under consideration, which has an average flowrate of 3 m^3/hr, originates from tablet production consisting of substances that otherwise, would be decomposed at low pH and destroy the fungi in the biological treatment step. Therefore, this line has to be pretreated separately before joining the second wastewater stream. This main wastewater line has an average flowrate of 33 m^3/hr and is passed through several stages of treatment. The schematic diagram of the WWTP showing the two supply lines and the major treatment components is illustrated in Figure 8.

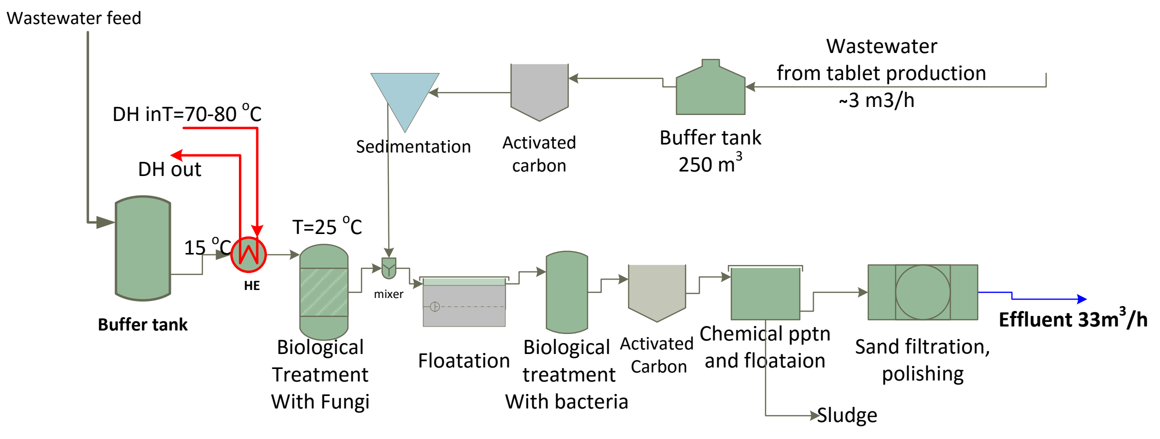


Figure 8. The major processes and flows in the wastewater treatment process at Gärtuna WWTP.

After being mixed in the buffering tank, the main wastewater stream is heated from about 10-20 ºC to up to 25-30 ºC before entering the bioreactor.

The treatment procedure for the tablet wastewater (3 m^3/hr) involves activated carbon followed by sedimentation, where the pollutants that would have been destroyed by the bio-agents are removed. Effluent is discharged into a mixer, where it combines with the main wastewater stream.

4.1.2. Options for Membrane Distillation at AstraZeneca

Membrane distillation is considered for removal of pollutants from the tablet production wastewater stream (3 m^3/hr). The removal efficiency of membrane distillation has been previously investigated at Hammarby Sjöstadsverk in research collaboration between KTH, IVL, and Xzero (Larsson, 2013), so the analysis considers energy aspects only

The wastewater stream from tablet production has a less complex mixture as it is collected from selected and few processes, unlike the main wastewater where it is collected from every pharmaceutical production and cleaning units. This stream will directly pass through the MD unit for treatment, replacing the activated carbon and sedimentation procedures. The schematic diagram for the integration of MD in the WWTP is shown in Figure 9. Such an approach eliminates the need for disposal of spent carbon or sedimentation residues; instead a highly concentrated liquid wastewater is produced, and the product water is contaminant free and could conceivably be reused. Provided that the concentrate is disposed of properly (i.e. incinerated), MD can thus lead to a zero liquid discharge treatment system.

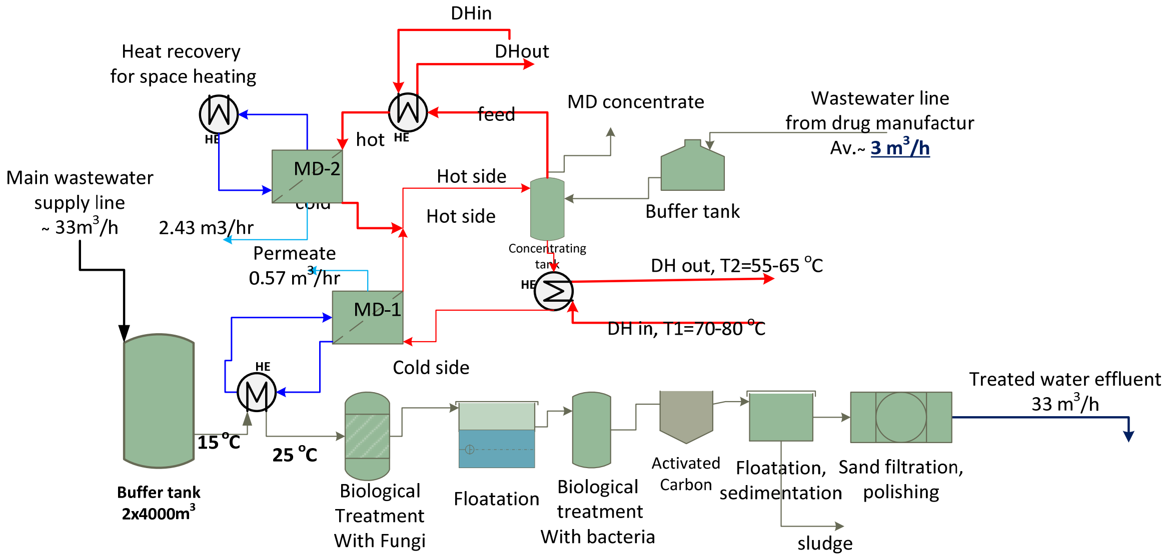


Figure 9. The WWTP with MD integrated for removal of pollutants from the tablet production wastewater stream.

The heat required for powering the MD unit is supplied by the DH network, and the MD system is placed between the tablet production pretreatment stream and the main wastewater stream. The main wastewater line will be heated (from 15°C to 25°C) by the recovered heat from the MD process, with excess heat available for process or building heat.

5.1.3. Heat demand and loss calculations

MD heat demand was calculated, using data obtained from experiments carried out on a pair of series-connected Xzero modules. A simple up-scaling was then applied in order to obtain the necessary number of MD modules and the annual heat demand required for 3 m^3/hr production capacity. The different heat flows in and out of the module pair and entire array are summarised in Figure 10; this example also serves valid for the other case studies. The permeate production rate of the pair of MD modules was taken from the previous experimental study on pharmaceutical removal test at Hammarby Sjöstadsverk (Woldemariam, 2014).

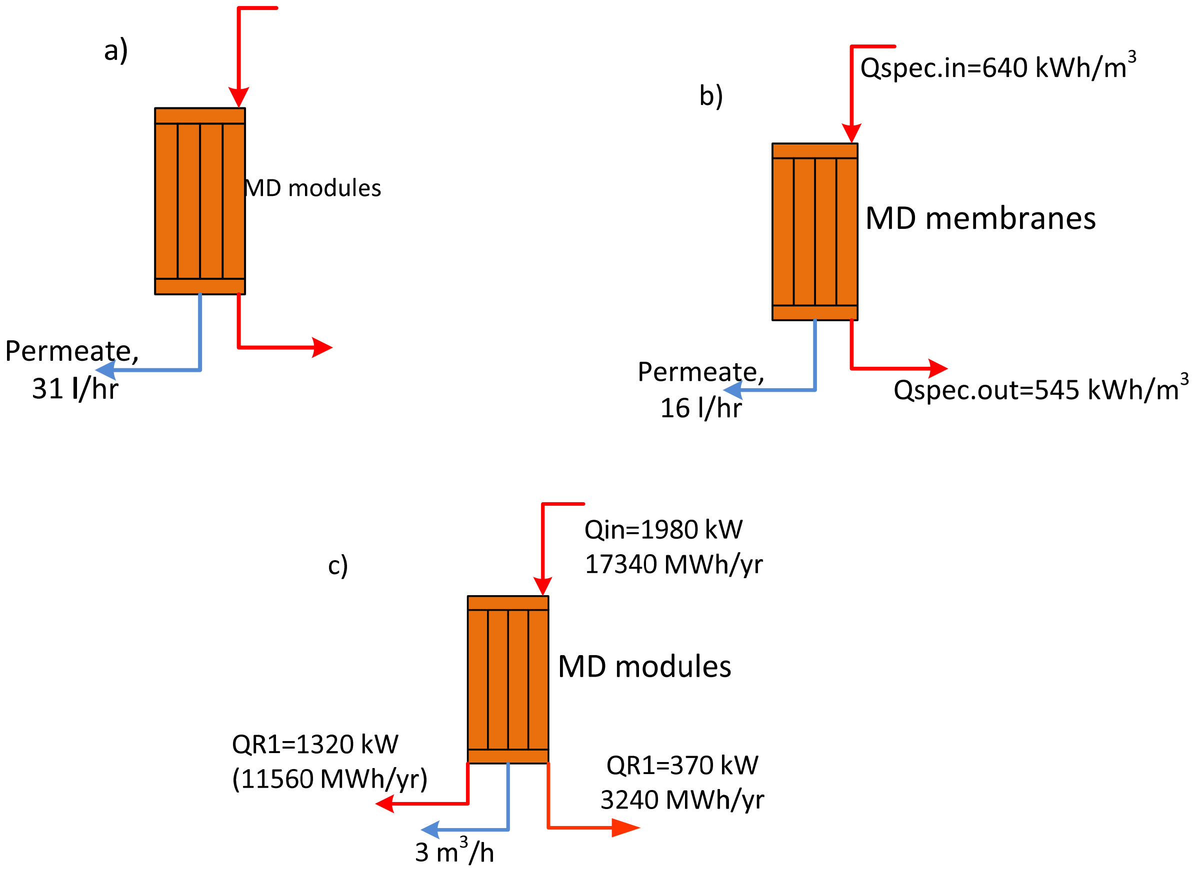


Figure 10. Heat flows into and out of Xzero AGMD modules for a&b are for series-connected pair for cooling 15 and 50 °C respectively, and c) full array for both 15 and 50 °C cooling a&b are for Th=80°C, Tc=15°C, where as c at Th=80°C, Tc=50°C

At a feed temperature of 80°C, the paired Xzero MD modules require a specific heat input ($\dot{q}_{spec.in}$) of 750 and 640 kWh/m^3 permeate at cooling temperatures of 15 °C & 50 °C respectively. Out of this heat input, about 650 kWh/m^3 is transferred to and recovered by the cooling water ($\dot{q}_{spec.out}$) for heating up the main wastewater steam and 545 kWh/m^3 is recovered at higher cooling temperature for space heating. The net specific heat demand ($\dot{q}_{spec,net}$) for a) is the difference between the heat input and heat recovered by cooling, which is 100 kWh/m^3 for the series-connected module pair. For the AstraZeneca array case study, the annual heat demand for the MD system is 17367 MWh of which 3240 MWh would be recovered in the wastewater treatment plant for raising the temperature of waste feed. The remaining 11604 MWh of heat is in theory available for space heating.

Table 1 presents the key findings for this case study. In 2014, the total DH demand of AstraZeneca (Gärtuna) was 20205 MWh (Midestad, 2015); this figure includes wastewater heating and space heating. Annual demand for space heating is thus 16965 MWh. The MD system with either Xzero or Elixir modules covers 100% of the wastewater heating demand and could provide up to 70%-100% of the building heating needs. The total DH demand with an integrated MD system implies an increase of 7-13% as compared to the base case. Hence, the integration seems promising from an energy perspective. Table 1 also includes MD membrane area requirements for the full-scale production at the WWTP.

Table 1.Summary of the MD system's annual heat demand and membrane area requirement when integrated with WWTP at AstraZeneca Gärtuna

Module	MD system heat demand (MWh)	Heat recovered from MD system at ca 50 C (MWh)	Heat supplied to 33 m^3/hr wastewater stream (MWh)	Net MD thermal energy demand (MWh)	Membrane area required (m^2)
Xzero	17367	11560	3240	2523	445
Elixir	21637	18252	3240	1432	293

5.1.3. Economic evaluation

The cost related to the MD system is derived from the DH demand and electricity requirement for the pumping, in addition to investment and maintenance costs (membrane cleaning, replacement of membranes every five years, etc.). The capital cost is distributed over a 15 year period (Liu & Martin, 2005) in order to calculate unit cost of purified water. The comparison of estimated cost of employing an MD system for tablet wastewater treatment is summarised in Table 2. The current annual activated carbon (AC) purchase and disposal related costs are about 0.55 MSEK (Gruvstedt, 2015) equivalent to a pretreatment cost of 23 SEK/m^3. Replacing the AC pretreatment process with an MD system yields specific costs of about 77 SEK/m^3 for Xzero modules and 37 SEK/m^3 for Elixir modules. (It should be noted that this cost does not include retrofitting, i.e. placement of MD system between district heating network and end users, with accompanying needs for piping, heat exchange, and control.) At first glance this makes MD technology uncompetitive, however one should keep in mind that AC pretreatment in its current form is unable to yield a zero liquid discharge scenario (see previous section).

Table 2. Summary of the wastewater treatment costs using Xzero and Elixir MD modules in AstraZeneca case

Expense type	**Amount**	
	Xzero	**Elixir500**
Capital expenditure	3.39MSEK	1.5 MSEK
Annual district heating	1.2 MSEK/yr	0.7 MSEK/yr
Other annual O&M costs	48.4 KSEK/yr	31.7KSEK/yr
Total annual expenditure	4.6 MSEK	2.2MSEK
Unit cost of purified water	77 SEK/m^3	37 SEK/m^3

5.2. Membrane Distillation Applications at Agroetanol

5.2.1. Background

Läntmännen Agroetanol is the only large-scale manufacturer and supplier of grain-based fuel ethanol in Sweden. The plant which started its first production in 2001 uses wheat, rye and barley as raw material cereals. Around 660,000 tonnes of grain are required to produce 250,000 m^3 of ethanol per year. The major processes in the ethanol production plant consist of four steps: a) mash preparation (which includes milling and liquefaction); b) fermentation, where yeast is added to the liquefied mash solution, converting sugars to ethanol and carbon dioxide; c) distillation and dehydration, where ethanol is distilled from the mash with remaining water separated by molecular sieve to achieve a product of 99.7% ethanol; and d) evaporation and drying, where the alcohol-free mash is dried and pelleted for animal feed.

The major processes of the considered plant are shown in Figure 11. The fermentation process produces ethanol and carbon dioxide along with glycerol, lactic acid, succinic acid, xylose, butanol, and acetone. During separation of carbon dioxide, a small amount of ethanol is evaporated with the gas mixture. After CO_2 is removed from this mixture, ethanol is recovered by first dissolving the vapors in water (scrubber); this aqueous mixture is fed into the distillation column.

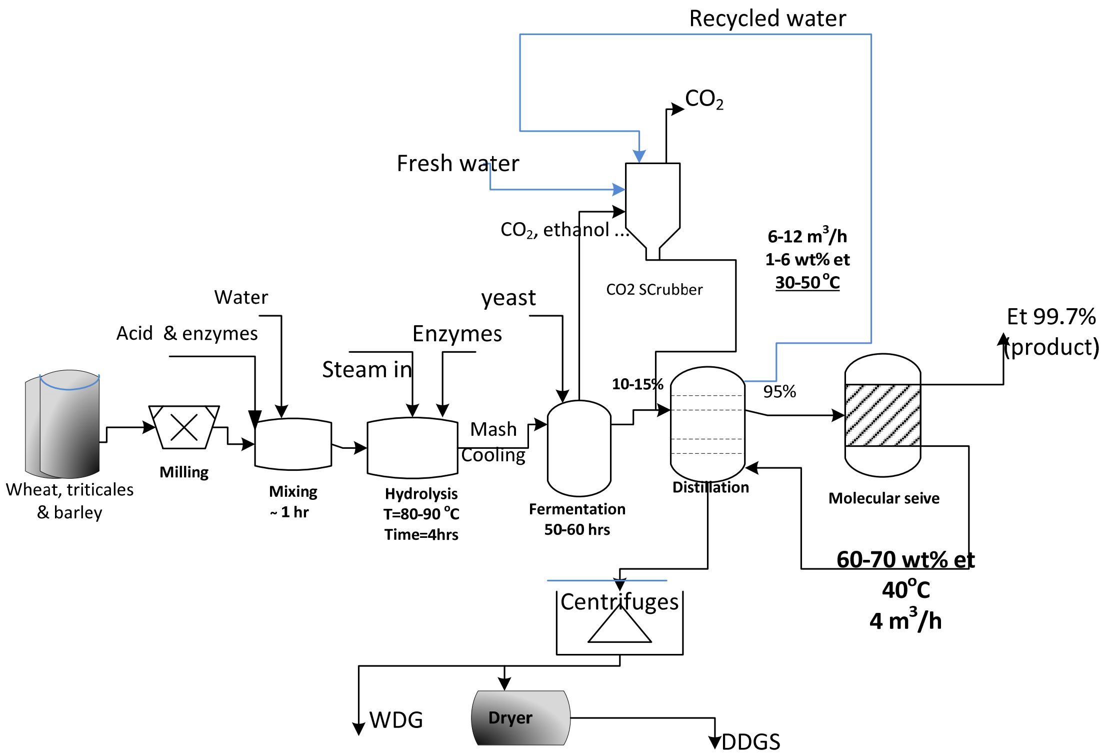


Figure 11. Schematic layout representing the major processes and components in the bioethanol production plant at Läntmännen Agroetanol.

The distillation column uses steam as a heat source, which is costly for the distillation of low percentage of ethanol from scrubber (1-6% eth. wt/wt). Taking specific heat demand for distillation 1825 kWh per cubic meter of ethanol produced (Rajoli, 2015), the annual heat demand for the distillation column can be calculated to be 5600 MWh/yr.

5.2.2. Membrane distillation integration and energy analysis

The MD system considered in the bioethanol production plant involves the recovery of the ethanol from the scrubber water. In the current plant, this water-ethanol mixture is fed to the distillation column where high-grade heat (steam) is used for distillation, as shown in Figure 11. As the percentage of ethanol in this feed is very low (1-6% wt/wt), the heat demand for the distillation process will be higher than the regular heat demand for feed from the fermentation broth with 12- 16 % ethanol. Reconcentrating the reject stream from the molecular sieve is another option for MD integration; however, the relatively high ethanol concentration is expected to be too demanding for this technology, and hence this option is not considered here. Figure 12 illustrates the chosen MD integration; care has been taken to ensure proper heat and water balances.

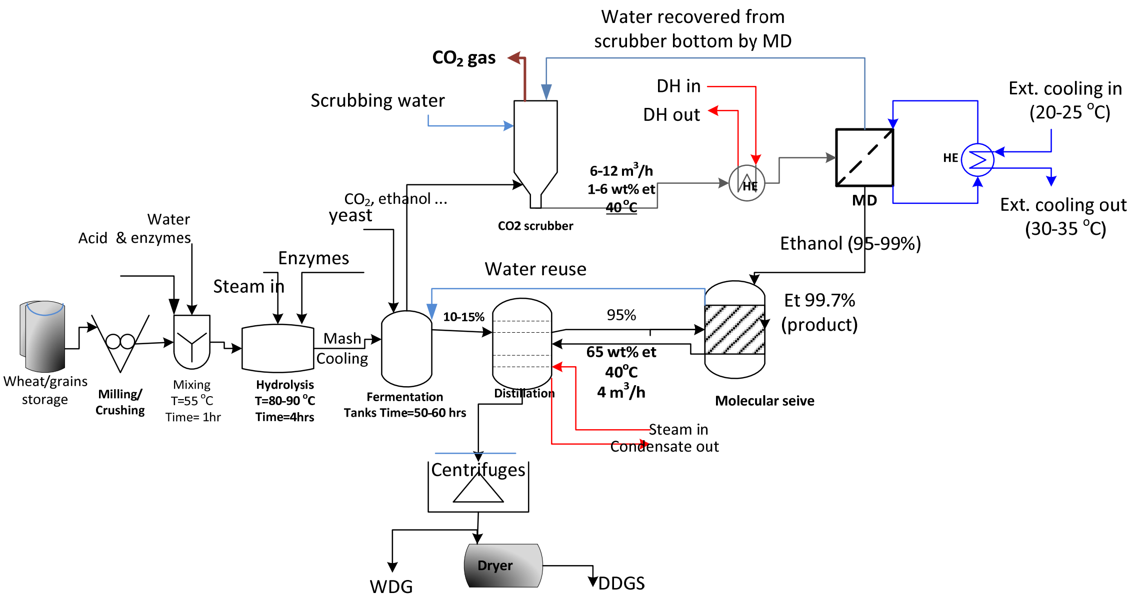


Figure 12 The bioethanol production plant layout with the integration of MD for separating water-ethanol mixture from scrubber water.

The MD feed is at a fairly high temperature (35ºC), and the additional few degrees temperature rise can be readily obtained from district heating. (Currently Agroetanol is not connected to the DH system, however E-on's CHP facility is located nearby.) Heat is rejected from the MD system at a low temperature, and it is assumed that this heat cannot be recovered for other purposes. Test for ethanol separation was done at KTH using Elixir MD modules whose summary is given in Table 3.

Table 3. Summary of performances of the Elixir MD module types for ethanol-water separation

Parameters	Value
Feed Ethanol % (wt/wt)	5
Feed temp. (°C)	33
Cooling temp. (°C)	15
Permeate flux, L/m²h	1.89
Spec. heat input (kWh/m³)	1633
Permeate ethanol %	77

The annual heat demands summarised in Table 4. The data shows that steam can be more or less evenly traded for district heating, putting MD technology at an advantage.

Table 4.Annual heat demand for the MD module and the distillation column.

Module /unit	Heat demand(Mwh/year)
Elixir	6258
Distillation C.	5600

5.2.3. Economic evaluation of scrubber water recovery by MD

The economic evaluation considers the capital investment along with operation and maintenance costs for the MD system. The capital investment is based on the required number of modules (or rather necessary membrane area) along with auxiliary equipment such as pumps, piping, valves, storage tanks, and control system. This calculation is based on data presented in the previous section. Assuming a price of 500 SEK/MWh for district heating, and taking other operation and maintenance costs into perspective, the net specific cost on a 95% ethanol basis is 1030 SEK/m^3 (capital costs included; 15 years span assumed). In comparison, the specific cost of providing steam to the distillation column for treating this stream is estimated at 1747 SEK/m^3, assuming a steam price of 800 SEK/MWh. A cost saving is therefore achieved with MD technology, and the payback period is close to one year. By offloading the distillation column, further gains could be made in productively, although these aspects have not been considered here. Further details on the economic evaluation are included in Table 5.

Table 5. Comparison between MD and conventional distillation for recovering ethanol from same low percentage ethanol-water feed (cost of steam was assumed to be 800 SEK/MWh and 500 SEK/MWh for DH heat). Capital cost for the distillation column is taken from (Crespo & Böddeker, 2013).

Module	Membrane area, m^2	Capital (MSEK)	Annual heat cost MSEK/yr	Other O&M, KSEK	Total cost SEK/m^3
Elixir500	190	2.25	3	7.9	890

5.3. Membrane distillation applications at Arla Foods

5.3.1. Background

The Kalmar Dairy Facility is one of the twelve dairies that Arla Foods operates in Sweden. Kalmar Dairy receives 160000 tonnes of milk per year from dairy farms in the region to produce various mild products, cheese, and whey. A large amount of energy is demanded in dairy plants in the form of steam, electricity, cooling and ventilation. Arla Foods had planned in 2011 to reduce the energy consumption and water use by 26% by the year 2020 (Figure 13). The introduction of such systems as membrane distillation could play a role to facilitate the not only in the reduction of energy and water consumption, but also the shift to renewables.

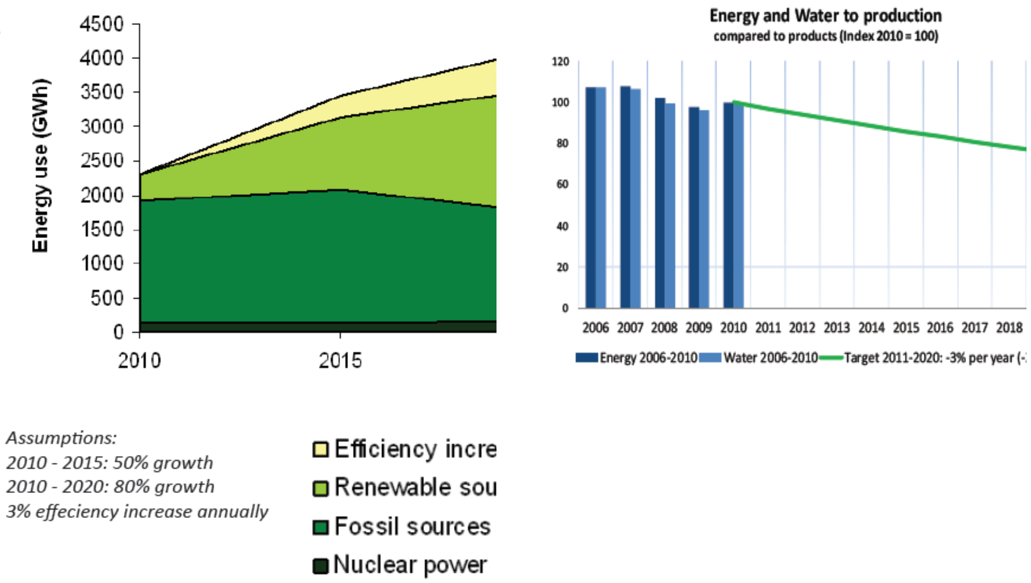


Figure 13. ArlaFoods' energy and water consumption scenario for 2020 a) Energy efficiency and composition b) Water and energy consumption % (ArlaFoods, 2011).

The major streams and components of the dairy plant are represented in Figure 14. Major processes include; milk skimming to separate some fat, cheese making and why concentrating.

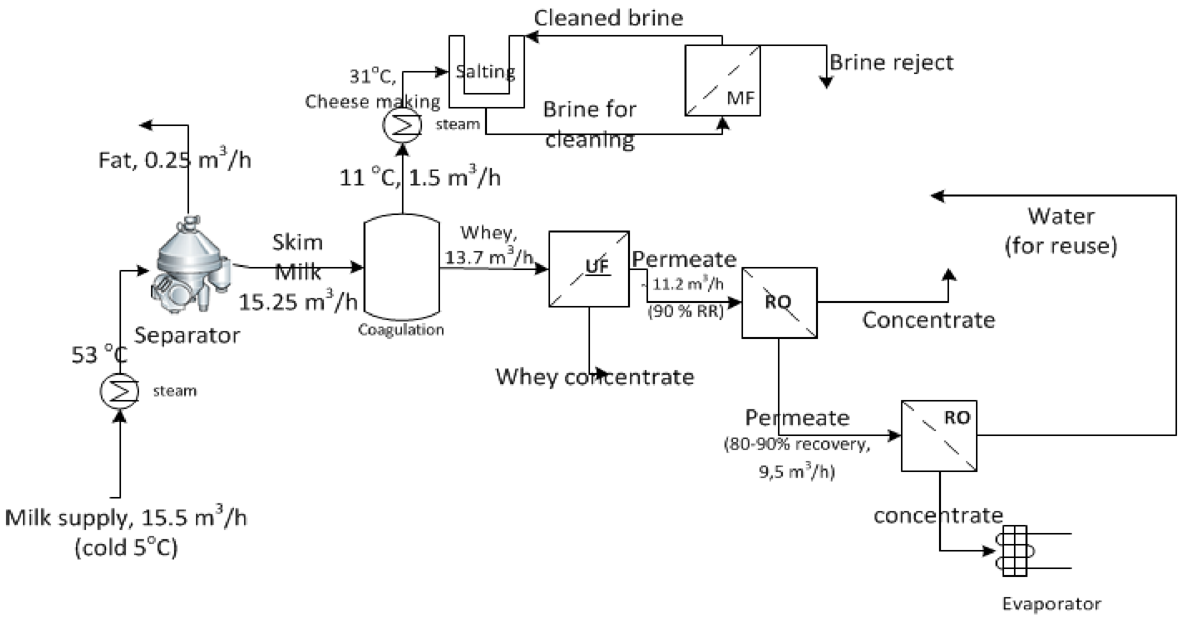


Figure 14. Process diagram for the major procedures in Kalmar dairy, ArlaFoods.

The three relevant processes that were considered for possible applications of membrane distillation were as follows: a) whey permeate concentration, b) replacement of second-stage RO polishing loop; and c) desalination of brine used for cheesemaking. The dairy process is supplied with steam and a number of cooling processes are also involved, which means it could be possible to integrate MD in the system so that the heat demand can be supplied within the system without affecting the total annual heat demand of the plant. District heat can be employed to supply the remaining thermal demand.

Membrane distillation has been investigated for two of the above potential applications, namely, brine concentration and RO polishing; see Figure 15. (Whey permeate concentration was not of interest to Arla Foods.) MD unit 1 is introduced in order to concentrate the brine and purify microfiltration water, and both MD permeate streams can be reused in the plant. The system requirement calculation for MD unit 1 is based on scaling up for a production of 1.75 m^3/hr pure water. Membrane distillation unit 2 (MD-2) is introduced replacing the second reverse osmosis polishing step. Replacing RO with MD unit will have the advantage of removing high-pressure pumps and so reducing the electrical energy demand of the plant. The product water from the MD can be used as a fresh water source which contribute to the reduction of fresh water consumption by the plant.

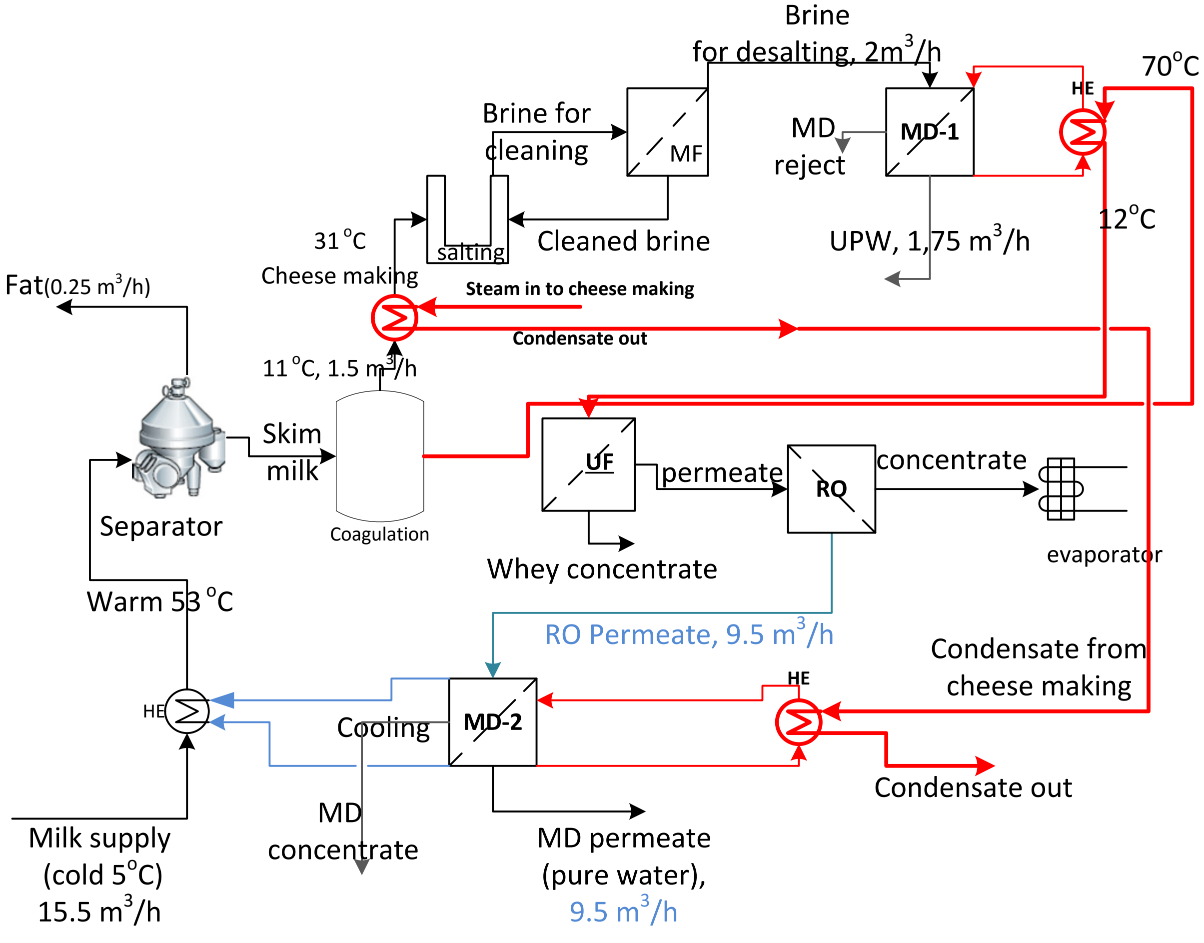


Figure 1. The dairy process scheme with MD systems introduced for desalinating the brine (MD-1) and polishing RO permeate applications (MD-2).

5.3.2. Energy demand and cost comparison

The main concept of integrating MD in to this type of dairy processing plant is to make use of any waste heat for separation process and so reduce the steam demand of the plant. In this regard, MD-1 can be integrated in such a way that the waste heat from whey cooling can be recovered by the MD for the purpose of concentrating the salt and producing fresh process water at the same time. The whey cooling releases about 8100 MWh per year from which about half of this can be usable to run the MD. The second MD system (MD-2) can be connected between the condensate return from cheese making and the cheese making process. The steam demand for the cheese making process, MD heat demand and heat flows around the MD systems are summarized in Figure 16**Fel! Hittar inte referenskälla.**. (Performance characterization of Xzero and Elixir modules has been presented in the previous section. Allowance has been made for high salt concentration according to theoretical considerations presented in Alkhudhiri et al. (2012).

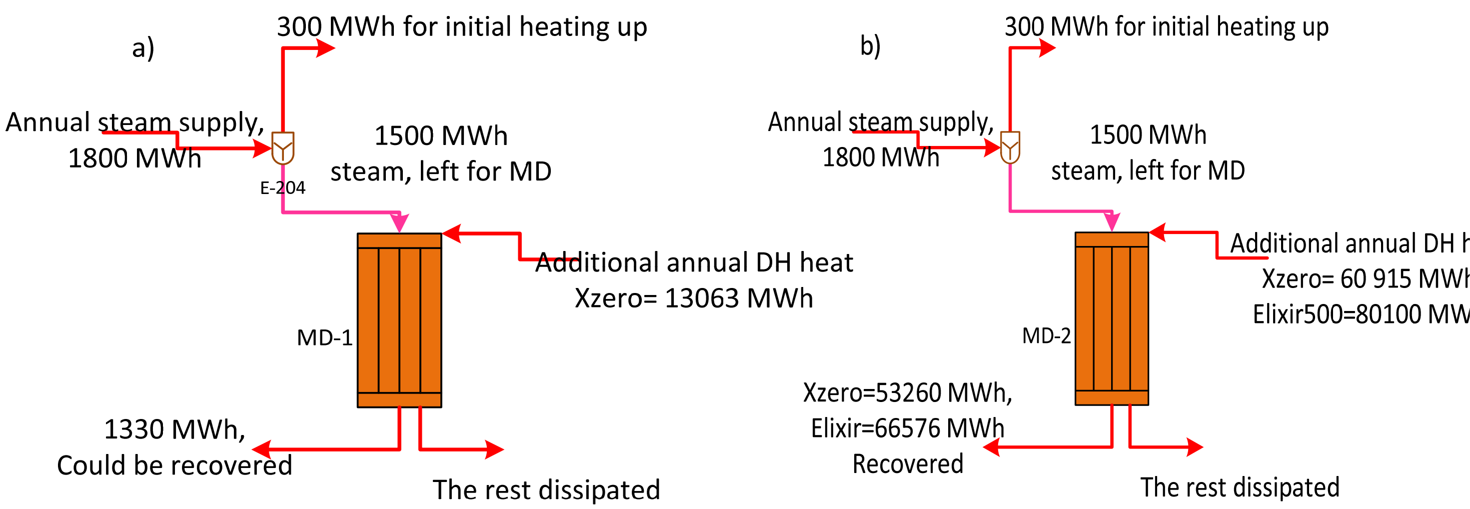


Figure 2.Available heat and additional heat demand of the two MD types for each application considered.lishing.

Based on the heat demand from the basic modules and linear scaling up to the large scale of 1.75 and 9.5 m^3/hr MD production capacity required to line up with UF and RO permeates respectively. As summarized in Table 6, Xzero modules require less additional heat due to the lower specific heat demand than for Elixir modules. Some amount (7420 MWh) of the heat required for either of the MD units can be recovered for raising temperature feed milk from silons before enjection to separator section. There is not much usable heat that can be recovered in the plant and also no district heating supply for the dairy considered in this study. Further study of the process could lead to replacing steam with district heating in other processes, which in turn might make a more favorable case for MD integration. Such an investigation is beyond the scope of the present study.

The total unit cost of treatment is also summarised in Table 6. For Elixir modules, test data was available only for RO permeate polishing. So when one compares the capital cost which is proportional to the membrane area, Xzero requires larger membrane areas ans so higher capital expenditure. But the annual heat demand is about two thirds of the heat demand by Elixir. The final lower higher unit price is therefore maily due to the higher heat demand by Elixir. In general the chice of modules can depend on the availability and price of heat to the plant in addition to the targeted production capacity.

Table 6. Heat demand and related costs for the two membrane distillation systemssimulatedforthe dairy plant case

		Capital MSEK	**Annual heat demand**	**Total unit cost**	**Membrane area**
Xzero	MD-1	1.48	14563 MWh	410 SEK/m^3	287 m^2
	MD-2	7.2	62415MWh	447 SEK/m^3	1409 m^2
Elixir	MD-2	4.7	82387 MWh	860 SEK/m^3	925 m2

6. EXPERIMENTAL INVESTIGATION OF TWO MEMBRANE DISTILLATION PROTOTYPES

6.1. Experimental setup

Two MD module prototypes, HVR and Elixir, were tested at KTH. Both have identical cassette-membrane inserts and differ only in condensation surfaces: the HVR module utilizes a heavy aluminum plate, characteristic for a laboratory rig only; while the Elixir module employs a thin-plate stainless steel condensation surface, with design features taken from commonly used plate-and-frame heat exchangers.

6.1.1. Experimental setup

Figure 3 shows a schematic of the experimental setup.

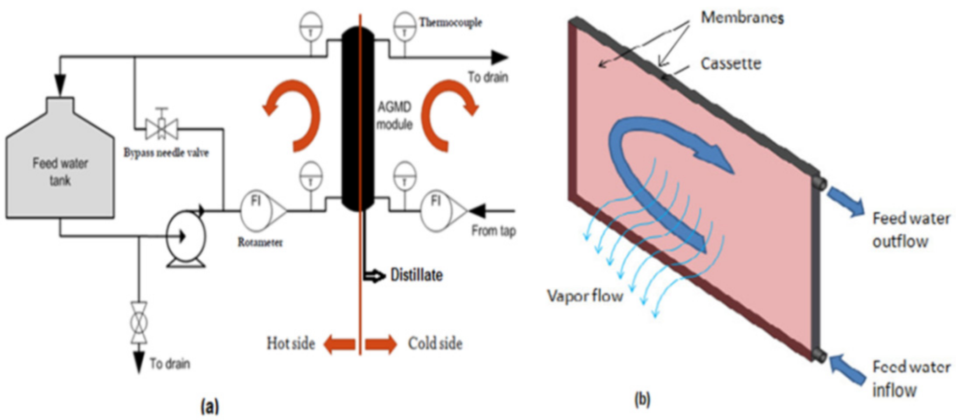


Figure 4. (a) MD bench scale unit setup at KTH lab (b) Membrane Cassette (MD) (Khan & Martin, 2014).

Two immersion heaters (combined rate of 4.5 kW) provide temperature control to feed water contained in a 24-liter tank. A small circulation pump and bypass valve allow the hot-side flow rate to be controlled, and a rotameter is employed to measure flowrate. Once-through tap water is used as a heat sink, which can be heat exchanged with an external source to raise the inlet coolant temperature to the desired level. Here a second rotameter with built-in control valve measures cold water flowrate in the cooling channel. Product water is measured with a graduated cylinder and stopwatch, typically during a 30-minute period of steady operation. The obtained permeate was returned to the feed tank. In this case, the feed concentration was constant over the entire course of the AGMD investigations. To measure the feed and cold temperatures, thermocouples were installed at the inlets and outlets of the module and were connected to a data logger (Keithley 2701 DMM with a 7706 card). Experimental errors are as follows: temperature, ±1.50^{o}C; flow rate,± 0.1 L/min; yield, ± 0.02 L/hr; and conductivity ±0.5µS/cm. The AGMD-HVR module consists of a 2.4 cm separation between two vertical condensations plates, behind which are located serpentine cooling channels. The experimental setup is almost similar for the case of AGMD-Elixir module but the condensate plate design is different (plate surface shape and thickness) and it consists of 2.1 cm separation between two stainless steel thin plates. A polypropylene cassette with membranes attached to either side is placed between the condensation surfaces (cassette dimensions 42 cm wide by 24 cm high, total membrane area 0.19 m^2) for both MD modules. This arrangement provides for an initial gap of 9 mm on each side, although the actual gap size is reduced by bulging of the membranes during operation. The feedstock is introduced at the bottom of the cassette and flows out from the top, as seen in Figure 17 (b). The membrane material is PTFE (polytetrafluoroethylene, supplied by Gore) with a porosity of 80% and thickness of 0.2 mm.

6.1.2. Experimental procedure

The AGMD experiments consist of analyzing the performance of the system under different operating conditions, namely various cold-side temperature levels. The operating conditions on the cold side included a flow rate of 1.9 L/min and a range of coolant inlet temperatures: 15ºC, 30ºC, 45ºC, 55ºC, and 70ºC. On the hot side, the temperature was kept constant at about 80ºC, with a constant feed flow of 3.8 L/min. Flowrates are selected to lie in the upper range in terms of ensuring low ΔT across the particular side while keeping pressure drop (linked to pumping power requirements) and absolute pressure (linked to membrane liquid entry pressure limitations) at reasonable levels. The initial conductivity of plain tap water is about 250 µS/cm at 25ºC.

6.2. Test results from experimental analysis – water as feedstock

The performance of the AGMD-HVR and AGMD-Elixir prototype are evaluated by analyzing product water flow rates (flux, L/m^2h) and specific thermal energy requirements (kWh/m^3) as a function of feed and coolant temperature difference (ΔºC). As mentioned previously, experiments were performed for high and low-temperature differences across the membrane for tap water.

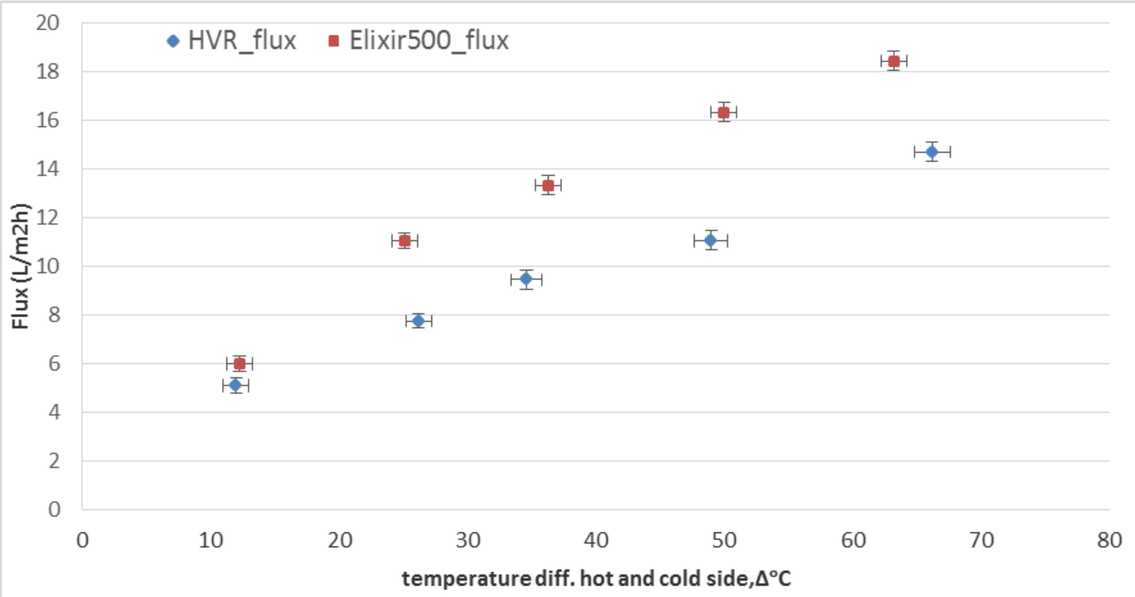


Figure 18. Product water flux as a function of temperature difference across the membrane (feedwater flow 3.8 L/min, coolant flow 1.9 L/min, constant feedwater inlet temperature ca 80ºC, coolant inlet temperature varying).

A feedstock-to-coolant temperature difference ΔT is defined for reference purposes:

$$\Delta T = T_{fi} - T_{ci} \quad . \qquad (1)$$

where T_{fi} and T_{ci} are the inlet temperatures of the feed and coolant, respectively.

Figure 18 shows the effect of this temperature difference on permeate flux at constant feed flow and coolant flow rate (the temperature difference is based on inlet conditions). The results show the increase of permeate flux with increase in temperature difference, an expected observation owing to the higher driving forces in this scenario. A small but significant flux is measured at a very low temperature difference, corresponding to a coolant temperature of around 70ºC, which has implications for heat recovery on the cold side (see next section). Figure 18 also shows that permeate flux was considerably improved by the AGMD-Elixir module compare to AGMD-HVR module and the flux improvement is gradually higher at level of high temperature difference. The major reasons for enhanced performance could be favorable condensate surface geometry, smaller air-gap resistance, and improved heat transfer with a thinner cooling plate thickness. The specific thermal energy consumption has been estimated in two ways:

Enthalpy drop across the hot side,

$$Q_2 = [\dot{m}_h c_{ph} (T_{hi} - T_{ho})] / \dot{m}_p \quad (2)$$

Net enthalpy change,

$$Q_2 = [\dot{m}_h c_{ph} (T_{hi} - T_{ho}) - \dot{m}_c c_{pc} (T_{co} - T_{ci})] / \dot{m}_p \quad (3)$$

where $\dot{m}$ is the mass flow rate and c_p is the specific heat for cold (subscript c), hot (subscript h), and product water streams (subscript p); subscripts i and o denote inlet and outlet, respectively. Figure 19 hows this data as a function of feedstock-to-coolant temperature difference. Specific thermal energy consumption shows a weak correlation to temperature difference.

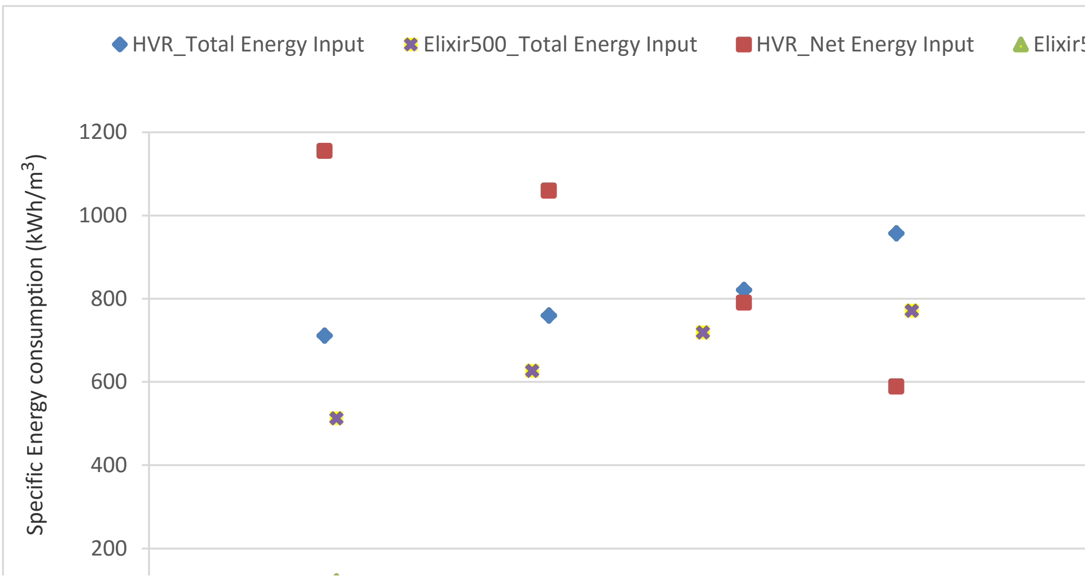


Figure 19. Specific thermal energy consumption for AGMD modules (feedwater flow 3.8 L/min, coolant flow 1.9 L/min, feedwater temperature ca 80°C, coolant temperature varying).

The slight increase at higher ΔT is attributed mainly to the fact that the enthalpy drop across the hot feedstock side rises at a faster rate than the concomitant augmentation in product water yield. The opposite trend can be seen in the specific thermal energy consumption defined as Net Energy Input (NEI=Total Energy Input – Energy recovery from cooling water), i.e. as the feedstock-coolant temperature difference is raised, the rate of increase in product water yield dominates over the difference in net feedstock and coolant enthalpy change. Moreover, heat recovery is enhanced with higher driving forces, which is reflected in a reduction of Net Energy Input at higher feedstock-coolant temperature differences. Figure 19 also shows the specific energy consumption in the feed water side of both MD modules follow same pattern though the feed thermal energy consumption is comparatively higher for AGMD-HVR unit. On the other hand, it can be said that the net specific energy consumptions have significant difference between two AGMD modules at especially lowtemperature differences (Δ°C) and for the case AGMD-Elixir unit, net thermal energy consumption is much lower. It also can be concluded that the new MD module (AGMD-Elixir) is highly energy efficient when heat recovery is deployed. The results indicate that the better heat transfer occur in the AGMD-Elixir module due to cooling plate thickness and high condensation rate of product water due to better geometry of cooling plate surface. It also shows that narrowing air-gap and decreasing the flow channel thickness, increases the Reynolds number, increases the linear velocity and convective heat transfer coefficient, decreasing the heat transfer resistance in the channel. However, heat of conduction losses might be higher for AGMD-Elixir module cooling plates due to the plate's surface geometry, reduced air-gap and condensate film thickness.

When we compare the performances of the two tested modules with that of Xzero, as summarised in Table 7, both the HVR and Elixir500 modules have better performance in terms permeate flux and net specific heat demand parameters. The net specific heat demand for the Elixir500 module is the least of the other two modules. This low net heat demand could be due to the high heat transfer across the cooling plates to the module's outside surface. Hence, the HVR and Elixir500 modules have better performance in minimizing the heat losses and so heat recovery by the cooling water is higher. Due to its high permeate flux, comparably lower specific heat input than HVR and the least net specific heat demand, Elixir500 module could be the preferred than HVR when heat recovery is possible in a given process.

Table 7. Comparison of the performances of the two modules in terms of permeate flux and specific heat demands

Module	**Permeate flux, L/m^2h**	**Specific heat input, kWh/m^3**	**Net specific heat, kWh/m^3**
HVR	14.2	972	70
Elixir500	17.4	990	36

*The feed water used in Xzero's test was treated wastewater from municipal WWTP, Henriksdal; whereas tap water is used in the other two modules' tests.

6.3. Thermoeconomic performance of developed modules for the large scale applications

For the large scale applications considered, scaling up simulations were done to calculate the energy demands and different costs related to capital investment, operation and maintenance costs which in this case is mainly for cleaning and replacing of membranes, heating and electric costs in annual basis and then the total cost for cubic meter of treated product. As the specific heat demand by Elixir modules is lower than Xzero modules, the annual heat demand for the MD system becomes lower as well. The number of MD modules or membrane area is also calculated for the particular industrial application. Essential assumptions have been considered in previous studies

(Kullab & Martin, 2011).

6.4. Test results from experimental analysis – ethanol/water mixtures as feedstock

Experiments were conducted with the Elixir module for 5% ethanol/95% water solutions. A full series of parametric data could not be obtained within the scope of this project, so instead preliminary tests were conducted in order to determine the specific heat demand and yield. Table 3 in the previous section lists the main results. It was not possible to achieve ethanol concentrations higher than 77% in this testing. Moreover the ethanol concentration in the permeate was found to be highly sensitive to the feed temperature, an expected result given that both ethanol and water are volatile.

7. CONCLUSION AND RECOMMENDATIONS FOR FUTURE STUDIES

This project was successful in addressing the overall goal – highlighting options for district heat-driven membrane distillation – and was also successful in treating sub-goals related to mapping of potential industrial applications, system analyses of selected case studies, and experimental investigations of MD prototypes.

Of the three case studies the potential of MD technology for scrubber water treatment at Agroetanol is clearly the most promising. Analyses of distillation column heat demand, MD system sizing (including detailed capital cost calculation), and process integration study should be conducted. Moreover pilot plant trials would be extremely helpful in confirming the viability of MD technology for the particular process stream. Discussions with the neighboring CHP facility are necessary to obtain the energy supplier's view on reducing steam delivery in favor of district heating.

For Astra Zeneca the potential benefits of MD technology are also positive, as this approach can lead to sizeable reductions in water-borne pollutants. Such gains need to be put into perspective of the high level of integration required (i.e. placement of MD system between district heating and entire facility heating needs). Future studies should focus on such aspects. Pilot plant trials are also justified, in particular to determine long-term performance and limitations on reconcentration levels.

MD technology as applied to Arla Foods was not viable as the required district heating demand was large, leading to high costs.

Experimental investigations of MD prototypes have shown that the technology is improving. Follow-on laboratory testing should be conducted on ethanol/water solutions in order to fully characterize heat demand and yield as a function of the process variables. Additional development work is needed to resolve key commercialization issues such as scale-up to multi-cassette modules featuring the tested cooling plate approach.

Beyond these aspects future investigations should consider promising applications that were not studied in this project, namely district heat-driven MD for low-volume wastewater treatment in hospitals along with water purification in swimming halls. Other potential areas for exploration include combinations featuring district heating with solar energy or industrial waste heat.

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DISTRICT HEAT-DRIVEN WATER PURIFICATION VIA MEMBRANE DISTILLATION

Här redovisas undersökningar av hur fjärrvärmedriven membrandestillering kan användas för att rena vatten i industriprocesser. Membrandestillering, också kallad MD-teknik, är en värmedriven teknik som utnyttjar partialtryck-differenser över ett mikroporöst, hydrofobiskt membran för att rena vatten.

Tekniken kan användas i temperaturer mellan 50 och 90 grader Celcius och är därför intressant för fjärrvärmetillämpningar eftersom det innebär en lägre el- och värmeanvändning jämfört med befintliga processer för vattenrening. Industrins egna miljö- och hållbarhetsmål bidrar till att intresset för fjärrvärme ökar. Tre fallstudier har gjorts där resultaten visar på fördelar och kostnadsbesparingar med tekniken, men även situationer där MD-teknik inte är lämplig att använda.