

Applying Membrane Distillation in High-Purity Water Production for Semiconductor Industry

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Abstract

While modern ultrapure water (UPW) systems are relatively well developed for semiconductor manufacturing, there is still room for improvement with regards to enhanced reliability, reduced environmental impact, and cost reductions. Membrane distillation (MD) is one promising alternative for high-purity water production that has several advantages compared to reverse osmosis (RO) and other technologies. In short, MD is a thermally driven process utilizing a hydrophobic membrane to produce high-purity water from a contaminated feedstock. Previous studies have shown that MD produces water with equal or superior quality to RO; in addition MD is relatively insensitive to process variations (e.g. pH, TDS levels, etc.), thus opening up the possibility of recycling rinse water. Low-temperature heat sources (i.e. under 100 °C) may be employed, thus allowing MD to be readily integrated into existing processes or even on-site cogeneration.

This paper explores the viability of MD for high-purity water production in a typical semiconductor chip fabrication plant. A brief literature review is included along with water quality analysis reports on typical MD product water. System simulations are employed to highlight possible scenarios with cogeneration. Results show that the specific energy consumption in the case study is around 440 kWh/m³ (thermal) and 1.9 kWh/m³ (electrical). The specific capital cost is around \$1.2/ m³ for the MD facility. These findings provide a strong impetus towards demonstration trials and other follow-on research and development activities.

Keywords: Membrane Distillation; Polygeneration; High-purity Water Production; Water treatment

1. Introduction

The manufacture of semiconductor components is a highly complex process with intensive demands on water and other feedstocks. According to statistics more than 6,000 liters of ultrapure water (UPW) are required for the production of one 200 mm wafer (1); this amount is expected to rise significantly when standard wafer diameters increase up to 300 mm in the future (2). Moreover, water purity levels are becoming more stringent with improvements in manufacturing techniques. These and other issues – namely environmental impact and energy costs – represent challenges that must be met by competitive fabs in the near term.

Most of today's commercial UPW systems rely heavily on membrane technology, and relevant membrane-based water purification techniques utilized in the semiconductor industry include reverse osmosis (RO) and ultrafiltration/nanofiltration. RO systems, **which are at the**

heart of most current UPW plants, have enjoyed several decades of successful research and development activities. While RO is proven and reasonably cost-effective, there are several drawbacks that have been difficult to overcome, e.g. elaborate pre- and post-treatment, high operation and maintenance requirements, high electricity consumption, and low recovery rates. Alternative technologies that retain the advantages of traditional membrane technology but have added robustness, lower costs, and better environmental performance therefore remain a priority.

Membrane distillation (MD) is a promising technology in this context. In short, MD utilizes differences in vapor pressure to purify water via a hydrophobic membrane. The process occurs typically at atmospheric pressure and can be driven with heat sources under 100°C, which offers advantages in robustness and energy efficiency as compared to RO. Previous research has shown that MD is capable of producing high-quality water with a variety of feedstocks. This opens up the possibility of utilizing chip rinse water in new recycling schemes. MD is ideal for integration with process heat recovery and/or on-site cogeneration of heat and electricity, since thermal energy that is normally wasted can now be employed for producing high-quality water. Hence this technology presents a wide spectrum of benefits for fab water purification systems.

The objective of this investigation is to explore MD as a promising technology for water purification in the semiconductor industry. The present work focuses on a prototype MD unit that is geared for mass production. Results from laboratory trials are scaled up for use in system simulations featuring MD integrated with distributed heat and power generation from natural gas-fired engines. A discussion on water quality issues and integration with existing process stages is also included. Finally cost estimates are presented for the system.

2. Membrane distillation technology

2.1 Background

Membrane distillation (MD) is a novel water purification being investigated worldwide as an alternative to conventional separation processes. The potential benefits of MD can be summarized as follows: [1] 100% (theoretical) rejection of ions, macromolecules, colloids, cells, and other nonvolatiles; [2] lower operating temperatures than conventional distillation; [3] lower operating pressure than conventional pressure-driven membrane separation processes; [4] reduced chemical interaction between membrane mechanical property requirements; [5] reduced vapor spaces compared to conventional distillation processes (3); [6] high resistance to the contamination ;and [7] recovery rates up to 77% (4). This thermally driven process employs a hydrophobic microporous membrane to support a vapor-liquid interface. If a temperature difference is maintained across the membrane, a partial 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. A variety of methods may be employed to impose this vapor pressure difference, and Figure 1 shows the four major types of MD system configurations: direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), sweep gas membrane distillation (SGMD) and vacuum membrane distillation (VMD). In the present work, air gap membrane distillation (AGMD) alone is investigated. (Air would be replaced by nitrogen or other inert gas in the present context, however this does not have an appreciable impact on operating principles.) The advantage of AGMD against other configurations of membrane distillation arises from the possibility of condensing the permeate vapors on a cold surface rather than directly in a

cold liquid (i.e. in the case of DCMD). In this configuration, the mass transfer steps involve movement within the liquid feed toward the membrane surface, evaporation at the membrane interface and transport of the vapor through the membrane pores and air gap prior to condensation (5).

The most important advantages of the MD process are that it does not need to operate at high temperatures as compared to traditional distillation, nor are high pressures required as compared to reverse osmosis. Thus low-grade heat (theoretically temperatures in the range from 30 to 70°C) can be employed to drive the MD process under atmospheric conditions. Such temperatures are available from a wide variety of waste heat streams, either in-plant or via external sources like cogeneration facilities, solar thermal collectors, etc. Previously various MD test beds have been employed to conduct experimental work. Alkalibi and Lior (6) have reviewed the state-of-art in this field for desalination applications. In nearly all cases these facilities are confined to bench-scale tests that are not readily applicable to commercialization (i.e. large-scale production of durable, scalable, and low-cost units). Lawson and Lloyd have reviewed MD and summarized transport theory, membrane properties, and module design (3). They pointed out the advantages mentioned above but also stated that the lack of experience in long term performance and uncertainties in cost are obstacles towards applying membrane distillation in industry.

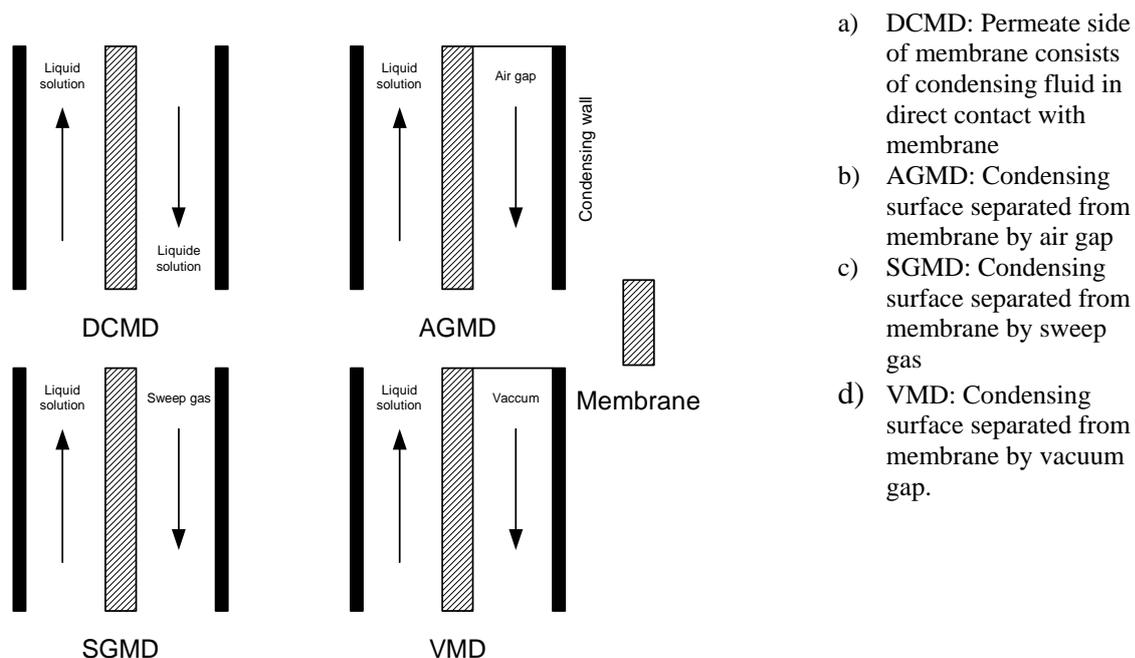
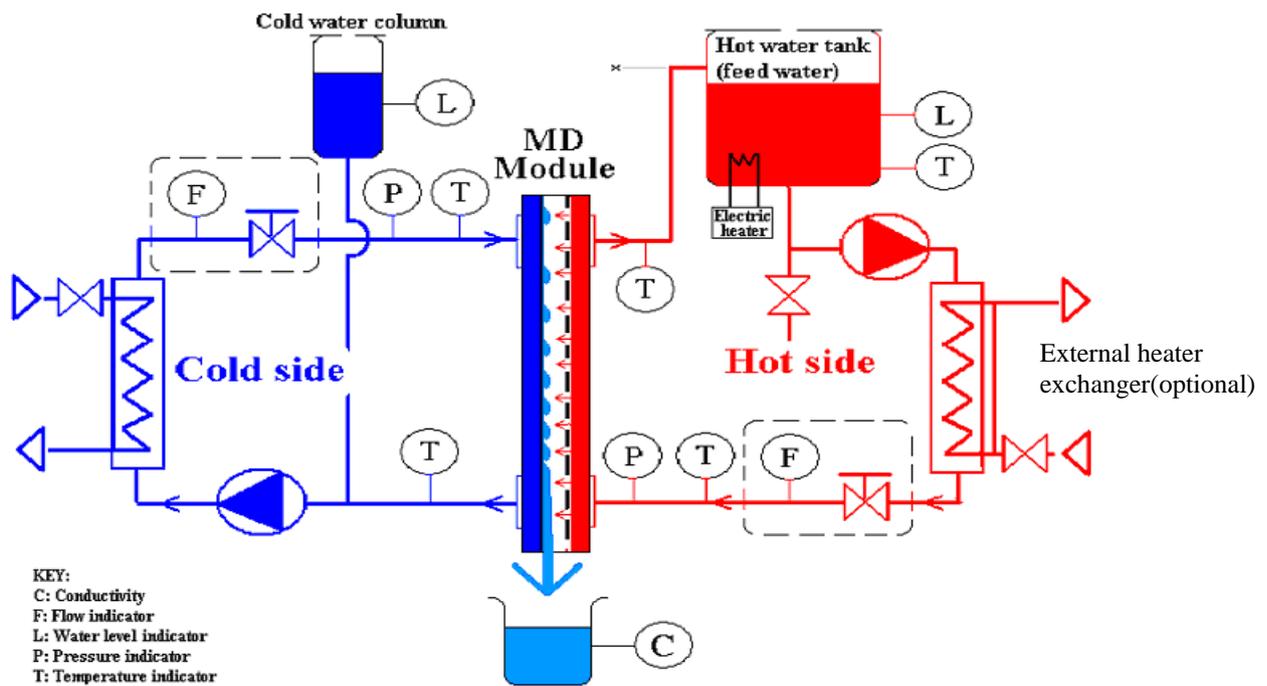


Figure 1. Common configurations of membrane distillation (3)

2.2 Experimental facility and results

The MD test facility was supplied by Xzero AB (our industrial partner) and was originally intended for process water treatment in industrial applications. The facility includes one membrane module consisting of ten plastic cassettes stacked together, resulting in nine feed channels and nine permeate channels. Module size is 63 cm wide and 73 cm high with a stack thickness of 17.5 cm, and the total membrane area is 2.3 m². PTFE membranes are employed with a porosity of 80% and a thickness of 0.2 μm. The width of air gap of AGMD is 2 mm.

Roughly 10000 hr of operation time have been logged with the original equipment (earlier testing was performed in Karlstad, Sweden, and current testing has been performed at KTH since March 2003). The experimental flow diagram and photo of MD module can be seen in Figure 2.2. When running the experiment, two taps are used to measure the water flow rate from cold and hot sides. During the experimental period, the pilot plant was operated in three kinds of flow conditions, e.g. 1.36 m³/h, 2.04 m³/h, and 2.72 m³/h. For simplicity identical flow rates were maintained for both hot and cold streams.



(a)



(b)

Figure 2. (a) Experimental flow diagram of MD test facility; (b) MD module

In the test facility the feedstock (normally municipal water) fills a 1-m³ tank equipped with three electrical heating elements. Pumps maintain the flow rates to the desired levels. On the cold side, an expansion tank is used to accommodate the expansion of heated cooling water. Tap water was used to cool the cold side through a heat exchanger, so the cold side water is in a closed system. In the summer time, the tap water temperature is very stable at around 25°C,

so the cold side temperature can be readily maintained in the range of 25-30°C. When conducting the experiments, run-time data included pure water production rate, cold and hot side temperatures, conductivity of pure water, TDS, and pH. Measurements related to water quality were designed to provide qualitative information on unit performance; more rigorous testing was included in separate laboratory trials (see below). Recent tests have shown that the conductivity of the product water is around 1.5 $\mu\text{S}/\text{cm}$ at steady state, even for cases where the feedstock was spiked with table salt at concentrations up to 1100 ppm. The pH value of the produced water is around 6.5-7.0, and TDS is always under detection limits (4 mg/L). Additional information may be found in Liu and Martin (7).

Figure 3 shows a plot of pure water production as a function of hot water temperature for the three internal recirculation flow rates tested. The performance improved at higher flow rates where the temperature drop in the channel direction is lowest. Comparisons to theoretical results are also included. ‘Unadj’ theoretical results refer to predictions obtained from Jönsson et al. (8), which considers a one-dimensional mass and energy balance coupled to experimental correlations. An attempt was made to account for non-ideal deviations from this model, shown by the ‘adj’ curve. This model was reasonably good for low temperature levels only. Both models overpredicted performance at higher temperatures, so output was simply extrapolated in the range 80-95°C. (This finding points to the need for additional fundamental investigations.)

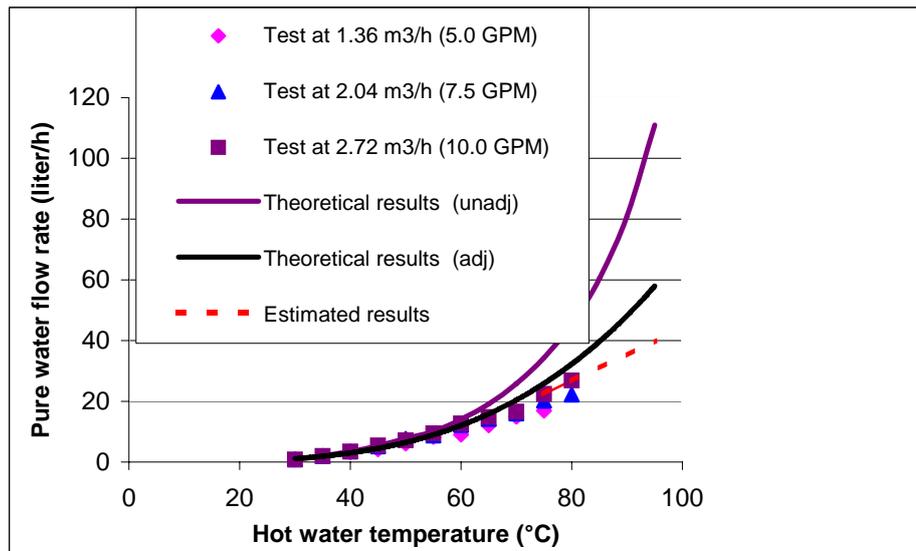


Figure 3. Pure water production versus hot side temperature for three different feed stock flow rates.

A more detailed water quality analysis has been performed by Sandia National Laboratories (9) using a smaller bench-scale MD unit. Three types of feed water were considered: rinse water collected over the weekend; rinse water representative of the daytime fab operations; and nonpotable city water. As shown in Table A, the MD unit was capable of producing high-quality water from the two feedstocks of primary interest. MD displayed superior performance to RO especially in its ability to reduce chloride, NVR, and silica levels. These results, in particular silica removal, indicate that MD can handle tough rinse water contaminants like boron and arsenic (10). This in turn implies that MD-purified spent rinse water could be supplied directly to the polishing loop. RO systems would require significant pretreatment in such a scenario. More details are on this point can be found in a subsequent section.

		Feed Water	Product Water From MD
Feed Water 1*	Chloride, ppb	108	3.8
	NVR, ppb	30	10
	Silica, ppb	17	3
	Resistivity, MΩ-cm	0.7	3
Feed Water 2**	Chloride, ppb	108	3.8
	NVR, ppb	500	20
	Silica, ppb	19	5
	Resistivity, MΩ-cm	0.002	0.7

* rinse water collected over the weekend; ** rinse water representative of daytime fab operations

Table A. Key results from Sandia National Laboratories tests (9)

3. Combined power generation and purified water production

MD systems on the scale required for fabs require substantial amounts of low-grade heat, and the combination of various fab processes consume electricity on the order of several MW or more. Thus it is logical to consider the integration of an MD water purification system with on-site power generation. Gas engines have been selected in this study due to their modularity and relatively low cost (gas turbines could also be a viable alternative for larger fabs).

3.1 Gas engine parameters and cooling water data

In general, the cooling system of a gas engine includes flue gas boiler, high temperature charging air (HTCA) cooling, jacket water (JW) cooling, lubrication oil (LO) cooling, and low temperature charging air (LTCA) cooling, as shown in Figure 4. For selected gas engine (11) the cooling water into gas engine is 50°C and is heated up to 95°C. The thermal power output is proportional to the engine's load; at full load the output is 5700 KW. In addition the number of engines can be varied to best match fab electricity needs.

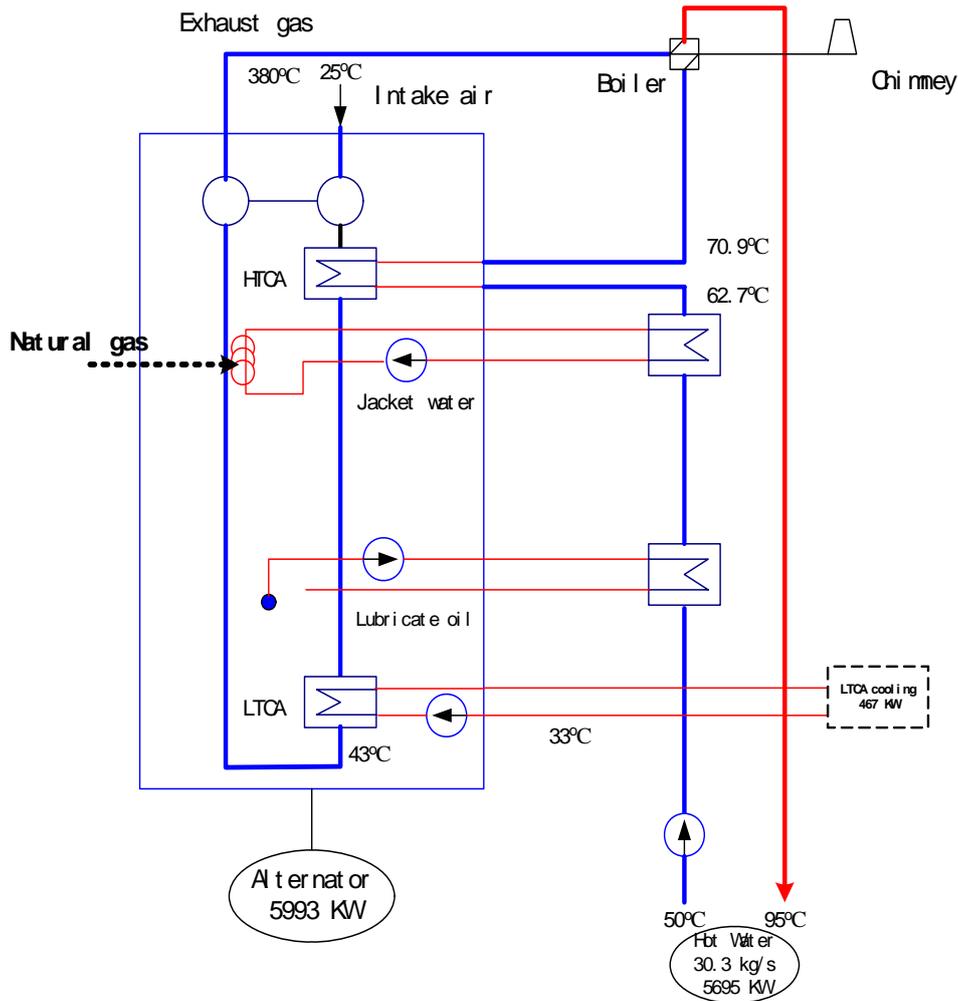


Figure 4. Gas engine hot water production system (11)

3.2 MD system description and gas engine integration

As shown in Figure 5, the MD system is assembled in one main stage and two sub-stages. The main stage comprises nine serial MD cascading stages, and the first and the second sub-stages comprise three and one serial MD cascading stage(s), respectively. Each MD stage consists of 36 MD modules assembled in parallel, and the size of each module is two times larger than those considered in laboratory testing. The system configuration was optimized from energy saving perspective in order to improve the efficiency of energy utilization. Heat exchanged from the gas engine is transferred into the hot side of MD main stage, and is then fed back to the hot side heat exchanger. The cold side of the MD main stage is used as the feed water of the first MD sub-stage, and the same configuration is repeated for the second MD sub-stage. Since the temperature of the last two main stages is low, additional sub-stages are deemed to be uneconomical. Cooling water is fed to the last two stages of main stage and the last stage of first sub-stage and the entire second sub-stage. System circulation was carried out in closed-loop principle, thus blow-down operations are necessary once critical contaminant concentrations are attained (such aspects are not accounted for here).

This integrated process was simulated with Aspen Utilities, with primary results including number of MD modules, pure water production, net energy consumption, and net electricity

consumption. A total of three gas engines are employed to supply the fab with 18 MW electricity; here the UPW demand is assumed to be 50 m³/hr.

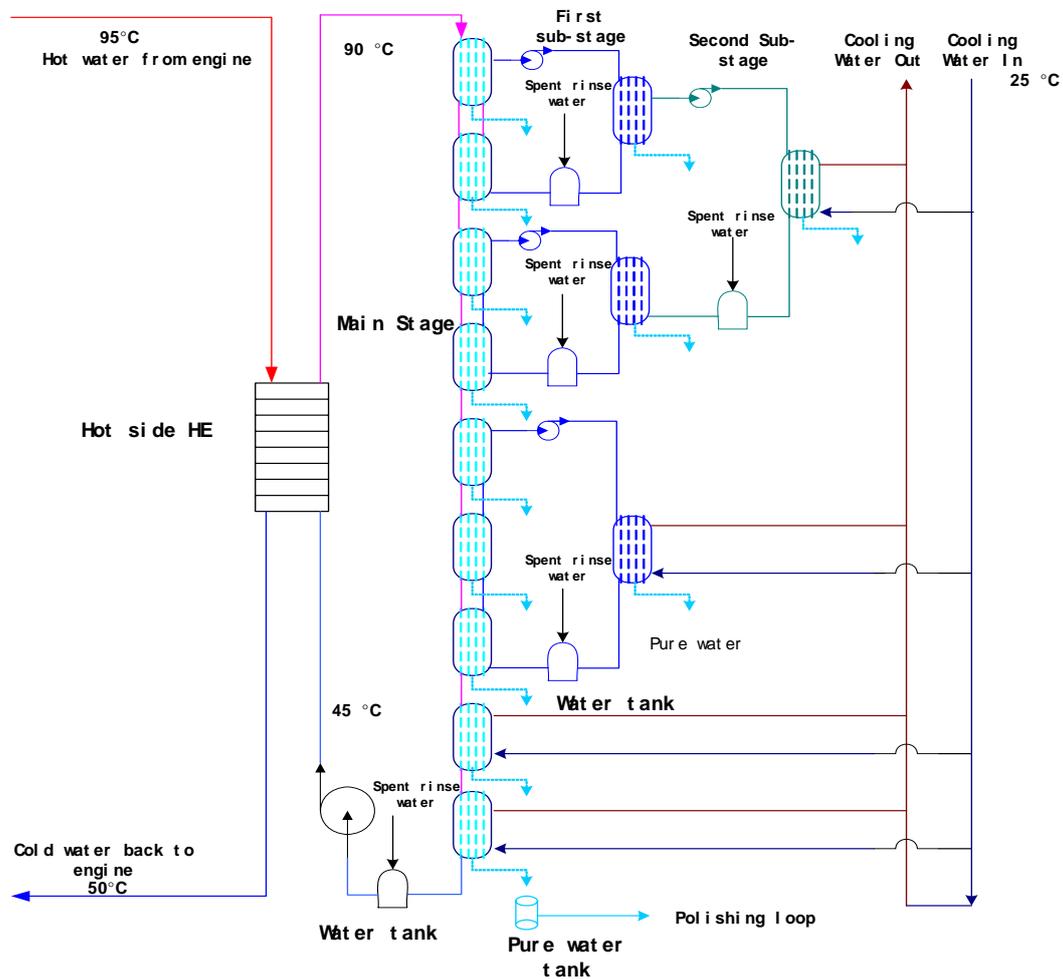


Figure 5 Simulation layout of one cogeneration/MD integrated unit.

4. Results and discussion

4.1 Technical results

The main results from simulation are presented in Table B.

Total number of MD modules	1404
MD pure water production, m ³ /hr	36.6
Fab high-purity water demand, m ³ /hr	50
Electricity production, MW	18 (combined total from 3 engines)
Fab electricity demand, MW	18
MD thermal energy consumption, kWh/m ³	438
MD electrical energy consumption*, kWh/m ³	1.9

*includes pumps, cooling tower, and auxilliary equipment

Table B. Simulation results

The selected integrated system with three gas engines covers all electrical needs of the fab, and the MD unit is capable of supplying over 70% of the pure water demand. (Remaining water could be provided by additional MD units driven by waste heat recovered from various fab processes.) The high specific thermal energy consumption demonstrates a limitation in recovering heat fully from the gas engines. Although the dual sub-stage configuration improves output by about 20%, around 80% of the total heat input is dissipated to the cooling towers. Adding more MD modules would increase water production marginally since the underutilized temperature levels are relatively low. The gas engine also has restrictions on temperatures that contribute to this situation. However, electrical energy consumption is relatively low, especially when compared to RO systems treating highly contaminated water.

4.2 Cost estimate

A first-order estimate on costs related to the MD system and cogeneration plant has also been considered. We assumed the thermal energy from gas engine is free for MD water production system. The following are key assumptions for the analysis:

- Cogeneration facility operated 8000 h/yr.
- The net present value and internal rate of return is 7 %.
- The lifetime of facility is 15 years.

For power plant:

- The installation cost is \$758/kW (see Table C for cost break-down)
- The overall O&M cost is 0.93/MWh
- The price for natural gas is \$6/Million BTU (13)

	Cost, \$/kW
Equipment	
Gen Set Package	400
Heat Recovery	40
Interconnect/electrical	12
Total Equipment	452
Labor/material	140
Total process capital	592
Project and construction management	75
Engineering and fees	31
Project contingency	15
Project financing	45
Total plant cost	758

Table C. Installation cost of gas engine system (12)

For MD water production system:

- The annual maintenance cost is 5% of the installed cost
- Membrane price is \$36/m²

- Electricity price is \$0.06/kWh
- Construction price is \$44 SEK/hour
- Security and control system is \$11250
- System heat losses are 5%
- Zero land cost

Capital Cost	
Item	
Membrane	\$90 000
MD modules	\$675 000
Heat Exchangers PH1(hot side)	\$150 000
Heat Exchangers PH2 (cold side)	\$112 500
Pump	\$225 000
Water tank	\$7 500
Piping	\$112 500
Valves	\$750 000
Measurement equipment	\$45 000
Security&control system	\$15 000
Construction	\$105 000
Cooling tower	\$10 000
Others	\$112 500
Subtotal	\$1 725 000
50% margin of error	\$612 500
Total investment	\$2 602 500
Operating and Maintenance	
Electricity	\$25 034
Maintenance	\$18 750
Subtotal	\$43 784

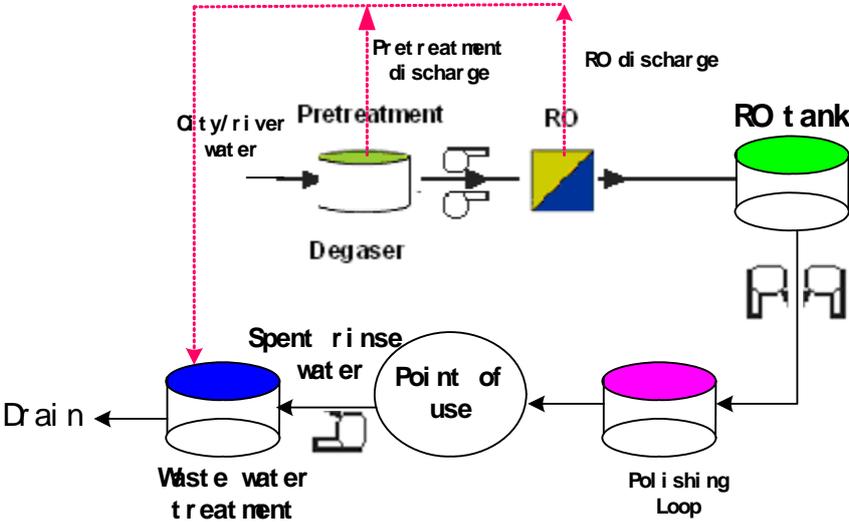
Table D. Cost Estimate for MD system

The specific cost for electricity production is \$0.064/kWh, which is less than typical grid prices of around \$0.10/kWh (13). In addition, with the own distributed power plant, the chip fab can achieve a more secure electricity supply. The specific cost of product water is \$1.13/m³, which is relative more expensive than the present cost for RO system. However this comparison should be weighed against the level of development for each respective technology. The present cost is calculated based on the laboratory level, and the price is expected to be reduced definitely with commercialization development. Moreover, MD has other inherent advantages that may partially offset the higher costs, namely the potential to utilize spent rinse water (see next section).

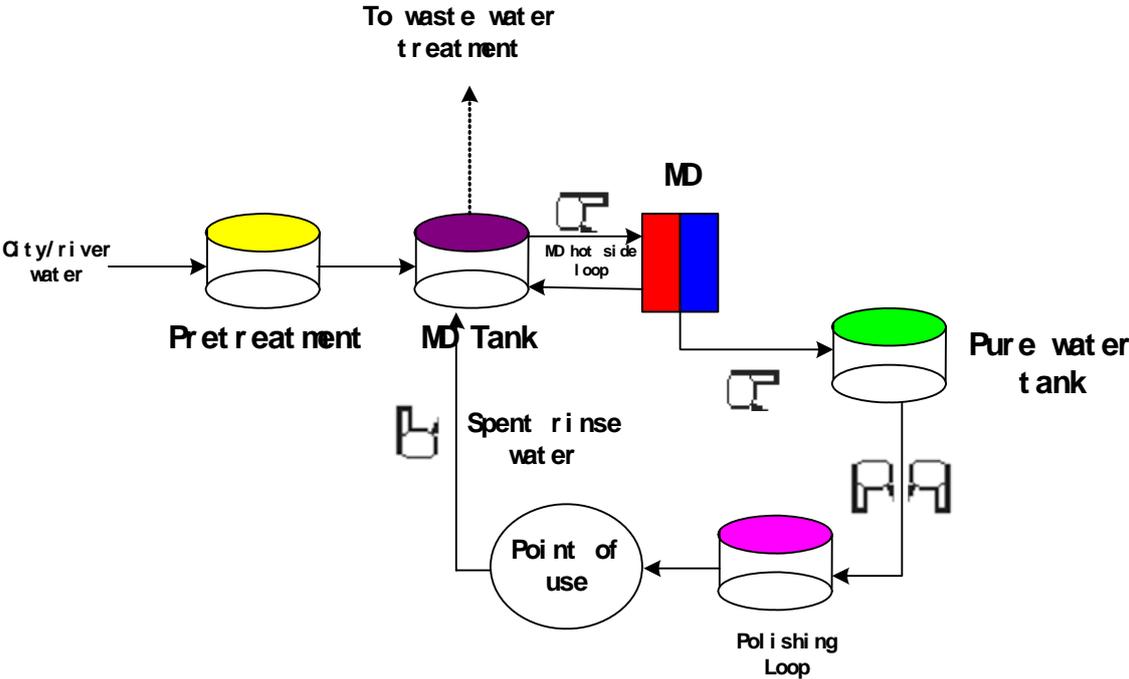
4.3 Water recycling with MD

Today's fab water purification systems cannot be easily designed for recycling since the introduction of contaminated water is problematic for RO. This difficulty is not as much an issue with MD as it can deliver high purity water even with highly fluctuating feedstock quality. Figure 5 illustrates a standard fab water purification system along with an advanced MD-

based layout with spent rinse water recycling. Although it is premature to say how far MD can go with regards to recycling, this configuration represents a significant step towards the ideal zero liquid discharge (ZLD) fab.



(a)



(b)

Figure 6. (a) Simplified flow diagram of current UPW system; (b) advanced UPW system with MD and spent rinse water recycling.

5. Conclusion

This case study shows that producing high-purity water from waste heat using MD integrated with a distributed power plant is a promising technology for future implementation. Nevertheless, more effort should be done in efficiency enhancement in the performance, water quality analysis, and heat recovery. Areas for further study include:

- Experimental investigations considering rigorous testing with respect to water quality especially for the semiconductor case investigation, including different parametric variations.
- Looking into different system arrangements, possible integration with other types of heat-driven processes or technology like heat pump or waste heat from chip fab.
- Exploring new module designs for optimizing output.
- Detailed economic analysis.

6. Acknowledgment

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Reference:

1. IONICS Pure Water Solutions, Fred Wiesler. *How to Meet Today's Dissolved Oxygen Specifications with Degassification Membranes*. (2004).
2. The McILVAINE, *Ultrapure Water Market*. (2002)
3. Kevin W. Lawson and Douglas R. Lloyd. "Review of Membrane Distillation", *Journal of Membrane Science*. (1997)
4. E. Drioli, F. Laganá, A. Criscuoli, and G. Barbieri. "Integrated Membrane Operations in Desalination Process", *Desalination*, 122. (1999)
5. S. Bouguecha, R. Chouikh, and M. Dhahbi. "Numerical study of the coupled heat and mass transfer in membrane distillation", *Desalination*, 152. (2003)
6. A. M. Alklaibi and Noam Lior. "Membrane distillation desalination: status and potential", *Desalination*, 171. (2004).
7. C. Liu and A. Martin. "Membrane Distillation and Application for Water Purification in Thermal Cogeneration-A Pre-study", VÄRMEFORSK Service AB, Stockholm. (2005)
8. A.-S. Jönsson, R. Wimmerstedt, A.-C. Harrysson. "Membrane Distillation – A Theoretical Study of Evaporation through microporous membrane," *Desalination*, 56 (1985) 237-249

9. R. P. Donovan and D. J. Morrison, "Evaluation of membrane distillation (MDU) prototypes for use in semiconductor manufacturing", Sandia National Laboratories. (1998)
10. Roy Hango, personal communication in response to Donovan and Morrison report, April 16, 1999.
11. Anders Lindberg, personal communication, Wärtsilä AB (Finland), 2003.
12. Wärtsilä AB, "Reciprocating Engines in Distributed Energy and CHP Applications" 3rd Annual National CHP Roadmap Workshop, DER&CHP in Federal Facilities, Boston, October 23-25, USA
13. Energy Information Administration, September 30, 2005.