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Membrane Distillation - A New Approach for Treating Waste Water in Nano-Electronics Industries

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Abstract:

In this study, a novel industrial application of membrane distillation (MD) is presented for waste water treatment in the nano-electronics industries. Previously reported performance of a semi-commercial Air Gap Membrane Distillation (AGMD) module is employed to evaluate the system operation in the terms of thermal energy analysis. To comply with thermal power demand in the MD systems, different integration possibilities between the MD unit and waste heat sources namely condenser outlet water from heat recovery chiller, process cooling water exhaust from manufacturing tools and hot air from Volatile Organic Compounds (VOCs) abatement combustion system are identified. Along with the technical assessment, this feasibility study has also involved the economic evaluation of the industrial waste heat integrated MD systems including unit water treatment cost. Results show the techno-economic viability of the proposed MD system integrated with industrial waste heat sources.

Key Words:

Energy analysis, Industrial waste heat, Membrane distillation, Nano-electronics, Techno-economic, Waste water.

1. Introduction and objectives

In 1958 the very first patent on the principle of integrated circuits (ICs) was published by Jack Kilby of Texas Instruments labs. Since then, the concept of forming transistors on the silicon crystal wafer has been used to fabricate ICs (nano-chips) [1]. The manufacturing process of these ICs is quite complex and delicate which begins with 100% pure silicon (Si) to deliver chips that function as intended. The first stage is to create the silicon ingot which is further sliced into 200-300 mm diameter of silicon wafers. Then, the second stage is to diffuse the dopant on the wafer to alter its electrical characteristics. Later deposition of the desired insulating/conducting layers on the silicon substrate is done, depending on the application. In the next stage, the chip is printed with an image on the specific zones of the device via photolithography. In this process, a particular “mask” is employed to develop a precise IC pattern by exposing the wafer to the photoresist material. Then, etching is done to eliminate the hardened photoresist on selective parts of the pattern by using plasma. These steps are repeated several times to build layers of transistors. These transistor layers are then interconnected with different zones of the chips by creating bonding pads in the metallization stage. Further, a finishing conductive layer is placed on the entire wafer to protect the circuit from damage and contamination. At last, the wafer surfaces are smoothen with the hybrid of chemical etching and free abrasive polishing [2], [3]. The typical process steps for manufacturing nano-chips are presented in Fig. 1. These complex processes require large amounts of fresh water ~ 90-145 kg/s (8000-12000 m³/day), comparable to the water requirement of a small city [5], [6]. Consequently, the manufacturing units generate a corresponding amount of waste water (approx. 83% of required water) from different areas, including silicon growth, oxidation, doping, dicing, ion implantation,

photolithography, etching, stripping, metallization, chemical mechanical planarization, washing and cleaning, etc. [2], [7], [8].

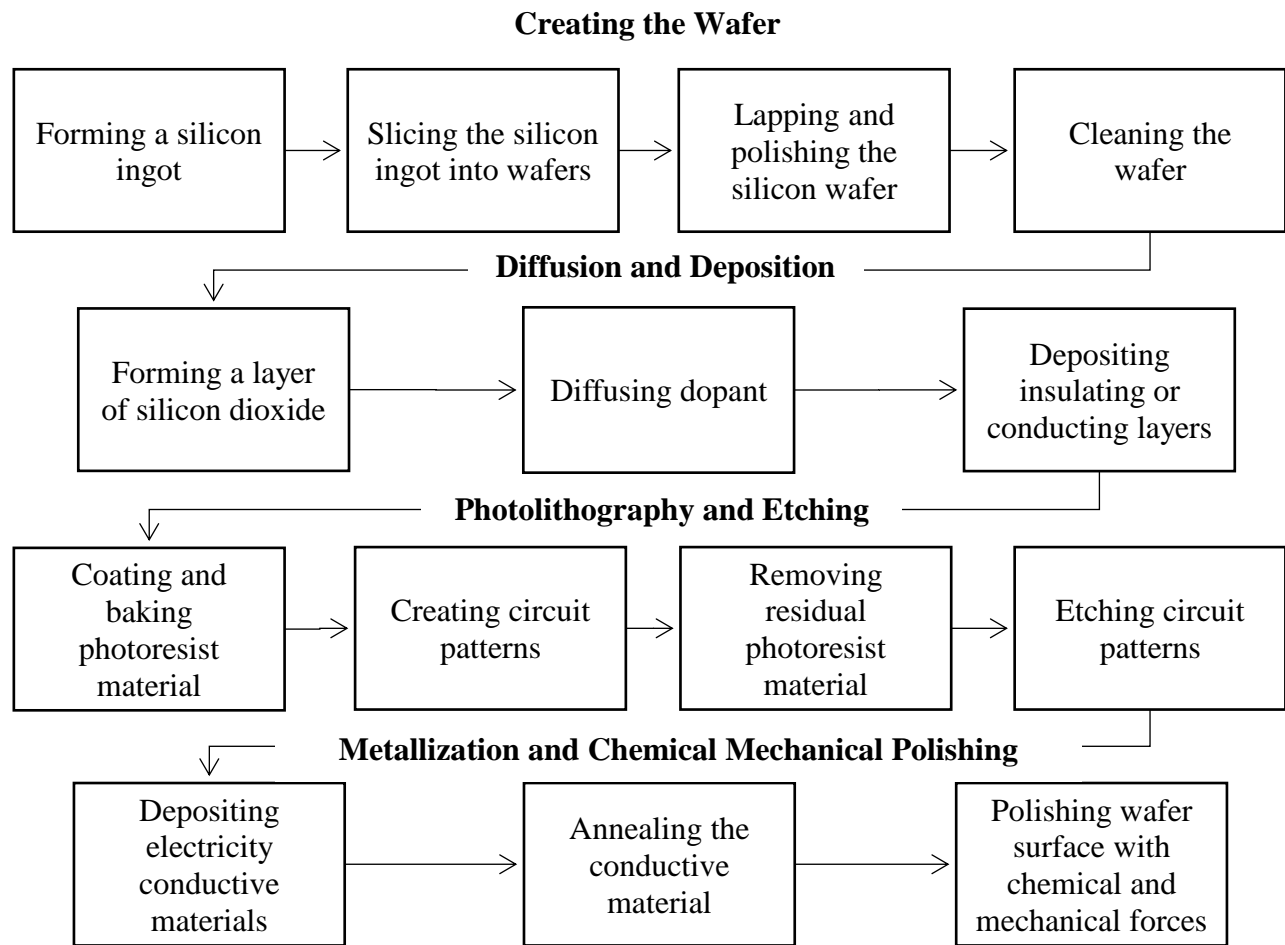


Fig. 1 Nano-chip manufacturing process steps [4]

Table 1 represents the wastes produced in aforementioned processes in a typical nano-electronics plant. Due to the push from EU urban waste water treatment directives, direct discharge of these waste waters into a water body is strictly regulated by water quality control authorities unless they are pretreated to international and local standards. Current emphasis is being placed on reducing the amount of waste produced. However, until a more suitable means of production is established, the waste streams must be treated.

Table 1 Major contaminants in waste water streams of the nano-electronics industries

Stream Characterization	Major Contaminants	Concentration, mg/L
Metallization	Copper, Chromium, Lead, Nickel, Iron, Magnesium and Calcium [14]	1-250
Etching Process	Acetic acid, Nitric acid, Sulfuric acid, Hydrochloric acid, Phosphoric acid and Hydrofluoric acid [3]	100000-250000
Photoresist Stripping	Dimethyl sulfoxide, Dimethyl sulfide, Dimethyl disulfide, Toluene, N,N-dimethyl acetamide, Acetone, Isopropanol and Dimethyl sulfide [15]	0.023-0.65
Chemical Mechanical Planarization (CMP)	Silica, Alumina, Ceria, Oxalic acid, Citric acid and Polyacrylic acid [16]	1-4000

There are many different technologies in use or under development for treating waste water, involving either mechanical or thermal separation [9], [10]. Membrane technologies are gaining more attention owing to their high separation efficiency and other positive attributes. Reverse osmosis, micro-filtration, nano-filtration and pervaporation are traditional membrane processes which are used for this purpose [11]–[13]. These approaches are highly recognized and generally effective, but there are some technical issues involved in these technologies, especially organic and inorganic fouling/scaling along with handling and disposal of the resulting brine solutions. In addition, these processes often involve significant chemical pre- and post-treatment and may exhibit high specific electricity demand. In this setting, membrane distillation (MD) is a promising membrane process for water treatment. Membrane distillation, a thermally driven process, provides a double barrier during purification: separation due to differentiation in contaminant boiling point, and membrane hydrophobicity that delivers high recovery ratios with pure distillate production. The main driving force is the temperature difference between the feed and cold side of the MD module. This results in water evaporation in the feed side which forms the liquid/vapor interface at the membrane surface. In air gap membrane distillation (AGMD), the vapor is transported through a thin air layer before condensing on a cold surface, with indirect heat removal provided by a coolant on the opposing side of the surface. The process normally takes place at temperatures below 100°C and at ambient pressure [17], [18]. Fig. 2 represents a schematic diagram of the basic AGMD purification method. As compared to other water purification methods, MD technology can theoretically achieve 100% rejection at relatively mild operating temperature and pressure; it is insensitive to feed concentration and pH; it utilizes low-grade heat; it needs less pretreatment procedures as compared to pressure-based membrane processes; and it requires less mechanical properties of membrane and lower capital costs than reverse osmosis and conventional distillation process [19], [20].

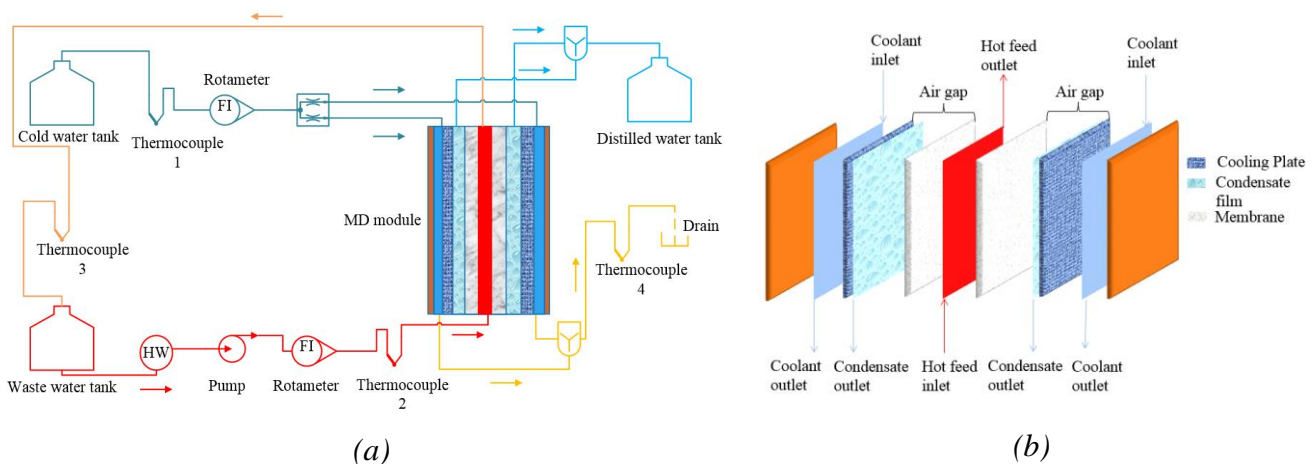


Fig. 2 (a) Schematic diagram of AGMD purification process, (b) Exploded view of AGMD module

Several researchers have characterized MD as an efficient technique for treating aqueous streams either for dehydration purposes or for water purification. Some relevant applications include recovery of heavy metals (arsenic, chromium, gold, copper, cobalt, lead, nickel, iron, magnesium, calcium, zinc and sodium) [21]–[23], dehydration of organic compounds (aromatic compounds and halogenated VOCs) [24], [25], concentration of acids (hydrochloric acid, phosphoric acid and sulfuric acid) [26]–[29] and separation of pharmaceutical residues at very low to moderate concentrations (ibuprofen, dibucaine, acetaminophen and diphenhydramine) [30], [31]. Such applications clearly indicate that MD has potential to be successfully employed in the nano-electronics industries for treating waste water streams which are typically contaminated with heavy metals, organic compounds, acids and nano-scale oxides. Nevertheless, waste water treatment through MD is accountable for relatively higher thermal energy consumption than other water purification processes [32]. Still, due to the fact that the system can be operated using low grade thermal energy sources, there are many opportunities available as heat sources including solar energy [33], geothermal energy

[34], district heating [31] and industrial waste heat [35] to facilitate the thermal energy requirement of the MD system. These integrated systems provide sustainable, cost effective and energy efficient methods for waste water treatment. Moreover, these systems play a vital role in reducing carbon footprints and in promoting sustainability.

Liu and Martin [36] performed pilot scale experiments for power, cooling and ultrapure water production (from fresh water) using AGMD module integrated with gas engines and heat recovery chiller in nano-electronics industries. However, to date, there is no study that has covered the application of MD in nano-electronics industries for waste water treatment, according to the authors' knowledge. Therefore, the objective of the present work is to introduce MD technology for the waste water treatment in these industries. Furthermore, the concept of integration of the MD system with industrial waste heat sources in nano-electronics industries i.e., condenser outlet water from heat recovery chillers, process cooling water exhaust from manufacturing tools and hot air from Volatile Organic Compounds (VOCs) abatement combustion systems is examined as well. By considering energy efficiency as the key parameter for the economical waste water treatment, this paper is focused on the techno-economic evaluation of industrial waste heat integrated MD systems for the treatment of CMP effluent (mainly SiO₂) in nano-electronics industries. This study specifically aims at finding out the optimal integration option in the view of water unit treatment cost, as the sole economic criterion. Typically, 4.17 kg/s (15 m³/h) of CMP waste water is released from an average nano-electronics industry having 10 CMP tools [37]. Therefore, the integration systems in this study are designed for waste water flowrates of 4.17 kg/s (15 m³/h).

2. Methodology

The applied method involves MD and waste heat integration system analysis based on energy and material balances with directions obtained from the nano-electronics industries, coupled with published results from AGMD experiments performed for treating municipal water contaminated with silica (SiO₂) [21]. Kullab and Martin [21] conducted pilot scale investigations and demonstrated that the achieved SiO₂ concentration in distilled water was <5 µg/L (below detection limit). These results made MD a very strong prospective candidate for removal of the most important constituent of CMP waste water i.e., silica (SiO₂) in nano-electronics industries. On this basis, it is suggested that for case of CMP waste water the separation efficiency would be similar. (In a follow-on study, tests with pharmaceutical residues [31] also demonstrated very high separation efficiencies.) This work provides a detailed parametric study of a semi-commercial Xzero MD module, including temperature and flow rate dependencies on permeate yield and thermal energy demand. Table 2 presents the characteristics of Xzero AGMD module that have been considered as reference for this study.

Table 2 Properties of reference AGMD unit [31]

Modules in series	2	Membrane thickness, mm	0.2
Membrane material	Polytetrafluoroethylene	Membrane support material	Polypropylene
Membrane area, m ²	4.6	Capacity, kg/s	0.33

2.1. Industrial waste heat sources

There are three waste heat sources which have been selected for this study in order to provide thermal power to the MD feed water.

- The condenser outlet water stream (~35°C) from heat recovery chiller [38].
- Process cooling water exhaust stream (~85°C) releases from the manufacturing tools [39].
- Hot air stream (~350°C) from VOCs combustion abatement systems [40].

The above mentioned heat sources can either be used individually, or they can be combined with other heat sources in order to realize the thermal power requirement for achieving the target temperature of 80°C. In this regard, two cases have been analyzed in this study as shown in Fig. 3.

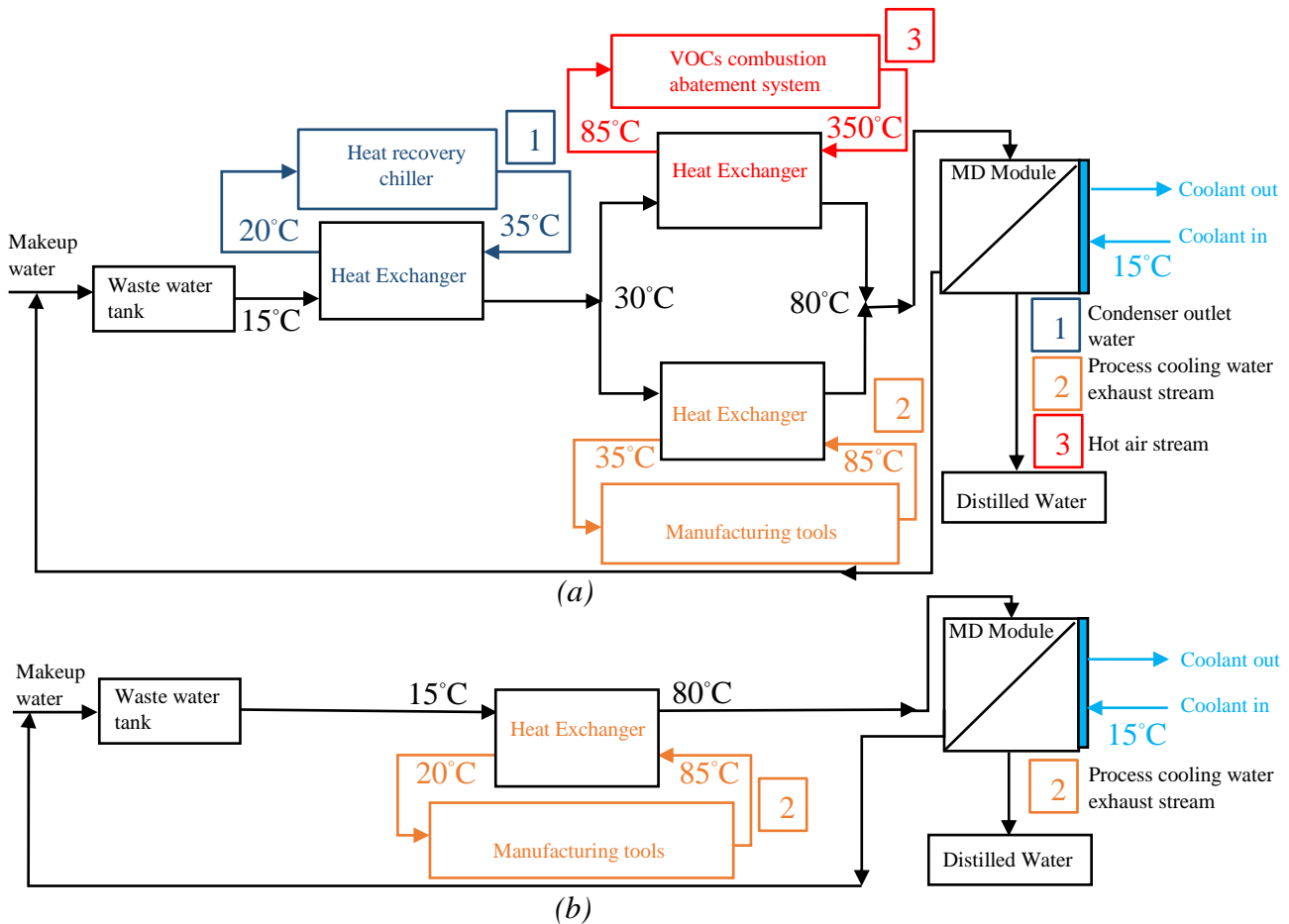


Fig. 3 Waste heat integration cases with MD: (a) Case 1, (b) Case 2

Case 1 – In this integration option, the MD feed water with flowrate 4.17 kg/s (15 m³/h) and inlet temperature 15°C, was preheated to 30°C with recovered heat from the outlet water of chiller’s condenser. Further, the energy recuperated from VOCs abatement combustion exhaust has been used to increase the temperature of partially divided preheated waste heat stream up to 80°C. The other part of the preheated waste stream was heated with the process cooling water exhaust stream having inlet temperature of 85°C.

Case 2 – In this scheme, process cooling water exhaust stream has been used individually as a thermal energy source to heat up the MD feed water up to 80°C.

Table 3 represents the characteristics of the available low grade heat sources/industrial waste heat sources in nano-electronics industries.

Table 3 Characteristics of low grade energy sources

	Condenser outlet water [41], [42]	Process cooling water exhaust [43]	Hot air [40]
Thermal power, MW	3.5*	2.44*	0.4**
Fluid temperature, °C	35	85	350
Fluid flowrate, kg/s	49.5	9***	1.5

* Reference to 20°C (room temperature)

** Reference to 85°C, considering the dew point of VOCs exhaust (hot air)

*** 10% of the typical water consumption in nano-electronics industries

2.2. Energy balance across MD module

The system involves substantial latent heat transfer across the membrane in order to facilitate the phase change of the feed water. This causes the change in enthalpy of hot stream (feed) since part of its heat is interchanged in latent heat that is then collected on the other side of the membrane by the

cold stream (coolant water) indirectly. Resultantly, the enthalpy change of the cold stream can be observed. The enthalpy changes of hot and cold streams were calculated using (1) and (2), respectively.

$$\dot{Q}_h = \dot{m}_h \times c_p \times (T_{h,i} - T_{h,o}), \quad (1)$$

$$\dot{Q}_c = \dot{m}_c \times c_p \times (T_{c,o} - T_{c,i}), \quad (2)$$

Where \dot{Q}_h and \dot{Q}_c are rate of enthalpy change (kW) for hot and cold streams, and \dot{m}_h and \dot{m}_c are the mass flow rate (kg/s) of hot and cold streams. $T_{h,i}$ and $T_{h,o}$ are hot feed inlet and outlet temperatures (°C) while cold water inlet and outlet temperatures are denoted by $T_{c,i}$ and $T_{c,o}$. The specific heat of water is termed as c_p (4180 J/kg K).

The net enthalpy change \dot{Q}_n across the MD module in kW was calculated from (3).

$$\dot{Q}_n = \dot{m}_h \times c_p \times (T_{h,i} - T_{h,o}) - \dot{m}_c \times c_p \times (T_{c,o} - T_{c,i}), \quad (3)$$

2.3. MD capacity requirement

For large scale processes, the capacity of the MD system was scaled up by increasing the area of the membrane. In this regard, some assumptions were made including similar operating conditions and specific performance parameters as in the reference case. The total membrane area (A_T) can be calculated for waste water flowrate of 4.17 kg/s (15m³/h) while considering the reference membrane area (A_m) and the corresponding mass flowrate (\dot{m}) using (4).

$$A_T = \dot{m}_h \times \left(\frac{A_m}{\dot{m}} \right), \quad (4)$$

2.4. Economic evaluation

Apart from technical assessment, economic feasibility of the MD module was also considered. Capital expenditures (CAPEX), and operational and maintenance expenditures (OPMEX) were calculated for cases 1 and 2 when the feed temperature was 80°C. For the purpose, different assumptions have been considered; such as (1) the annual interest rate and plant lifetime were selected at 5% and 20 years respectively; (2) membrane life was considered 5 years; (3) retrofitting cost was taken at 4% of the CAPEX and (4) annual OPMEX was determined as 20 % of the annual CAPEX.

CAPEX were distributed among all the processing/manufacturing equipment's cost which contained MD modules, heat exchangers, water tanks, pumps, temperature and pressure sensors, control system, piping, fittings, valves and retrofitting cost. However, OPMEX covers expenses such as utilities (heat/electricity), chemicals and disposal, operating supplies and services, technical assistance and insurance. Moreover, water treatment unit cost was calculated by adding the annual CAPEX and OPMEX and then dividing them with annual waste water production rate. Table 4 presents the cost of equipment for building a MD module setup in reference unit.

Table 4 CAPEX contributions of different components for a MD module setup in reference unit [31]

Equipment	Cost, \$	Equipment	Cost, \$	Equipment	Cost, \$
MD module	6100	Pressure indicators	48	Water tank	671
Heat exchanger	854	Security system	610	Piping	1025
Pump	366	Other costs	3660	Valves	24
Construction	1208	Temperature indicators	101		

3. Results and discussion

3.1. Thermal power demand and integration options

Owing to the temperature limitations of condenser outlet water stream from heat recovery chiller and thermal power limitation of VOCs exhaust stream, two different integration cases (as mentioned above) have been selected to fulfil the power requirement in order to achieve the target temperature.

It was determined that power requirement can be fully satisfied in case 2 with process cooling water exhaust stream individually, where the required flowrate of that was 4.17 kg/s (available: 9 kg/s) and estimated required power was 1.1 MW (available: 2.4 MW). However, in case 1, the waste water of 4.17 kg/s was preheated from 15°C to 30°C, accounting for thermal power demand of 0.26 MW which was fulfilled by the condenser outlet water stream coming from heat recovery chiller. Further, 1.5 kg/s of hot air from VOCs abatement system has been used to heat the fragment of waste water having flowrate of 1.91 kg/s (out of 4.17 kg/s) up to 80°C. In parallel, for the other part of the waste water, 2.26 kg/s of process cooling water exhaust stream has been employed to reach the target level. Thus both cases have the potential to fulfil the thermal energy demand while achieving the desired temperature (80°C). Table 5 and Fig. 4 show the characteristics of different low grade heat sources in the specific integration cases to provide thermal energy to the MD feed of flowrate 4.17 kg/s (15 m³/h).

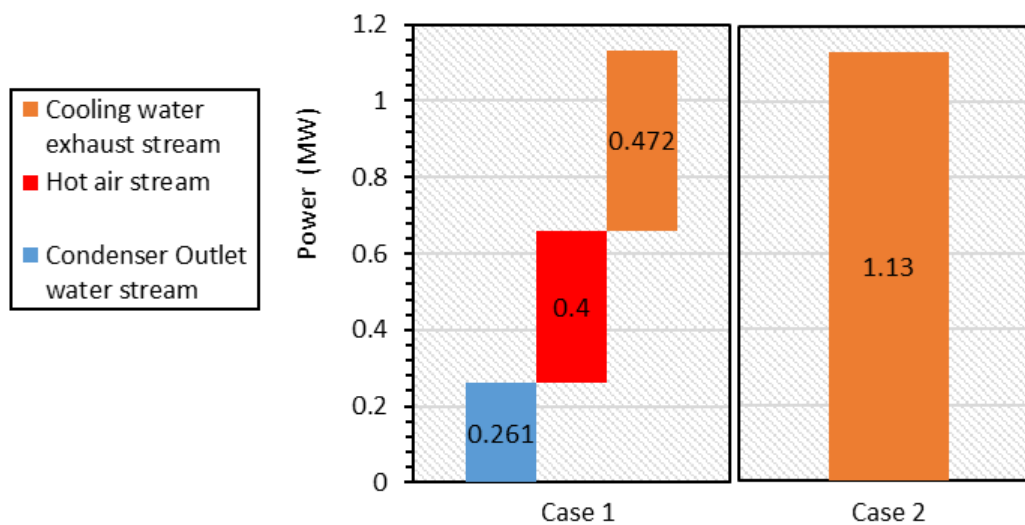


Fig. 4 Thermal power distribution of industrial waste heat sources for the integration cases

Table 5 Characteristics of energy sources and MD feed streams in the two integration cases

Cases	Industrial waste heat source	Flowrate of heat source, kg/s	Corresponding flowrate of MD feed, kg/s	Max. achieved temperature by MD feed, °C
1	Condenser outlet water stream	4.17	4.17	30
	Hot air stream	1.5	1.91	80
	Process cooling water exhaust stream	2.26	2.26	80
2	Process cooling water exhaust stream	4.17	4.17	80

3.2. Performance of the AGMD module

For the large scale MD system, the new total membrane area calculated was 58 m² based on the waste water flowrate of 4.17 kg/s (15 m³/h), while considering 2.3 m² of active membrane area of each module. In this case, obtained distillate yield was 0.106 kg/s (0.384 m³/h) when the feed temperature was 80°C. The simulated results were further applied for economic analysis of the industrial waste heat integrated MD system. Table 6 presents the performance of large scale case AGMD system based on reference case.

Table 6 Performance of AGMD large scale system

Xzero Module	Feed Temperature, °C	Cooling water Temperature, °C	Feed Flowrate, kg/s	Membrane Area, m ²	Distillate yield, 10 ⁻³ kg/s	Enthalpy change, kW	
						Feed	Net
Large scale	80	15	4.17	58	106.8	282	31.6

3.3. Economic analysis

Fig. 5 presents the CAPEX distribution and unit water cost for the integration cases 1 and 2 for full scale MD system (25 MD modules) having capacity of 4.17 kg/s (15m³/h) while considering MD feed target temperature as 80°C.

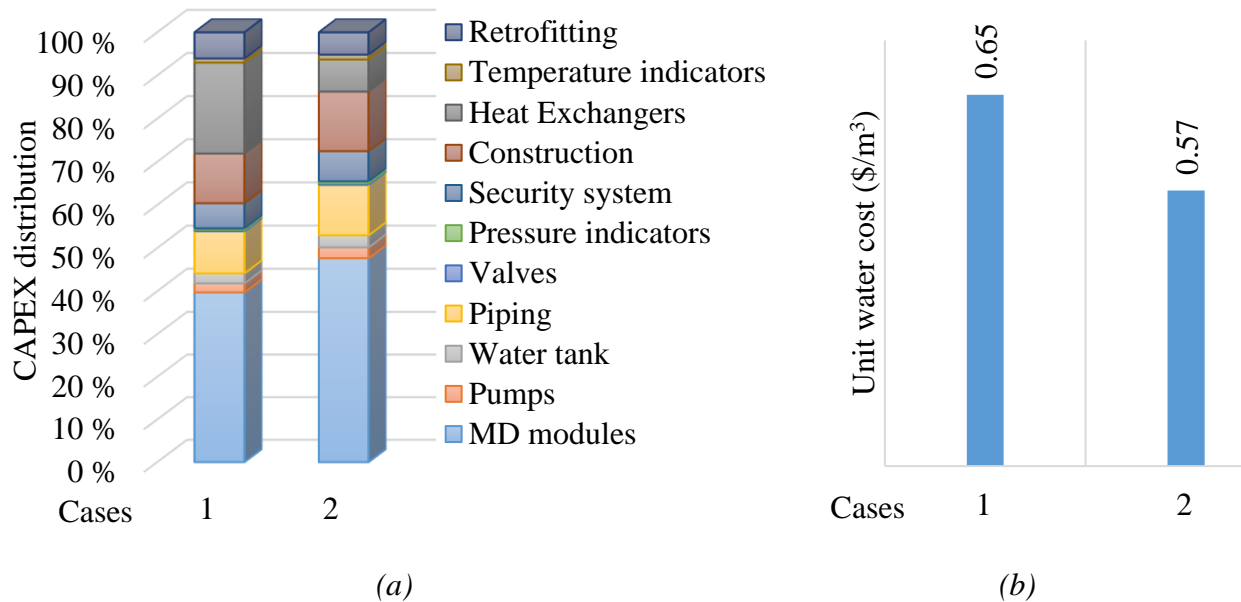


Fig. 5 (a)CAPEX distribution, (b)Unit water treatment cost

CAPEX distribution varies in these cases due to different low grade heat sources. In each case, MD modules have the highest cost, i.e., 39% and 47 % for case 1 and 2 respectively, followed by construction/assembly and piping cost in case 2. Heat exchangers as accounted for 21% of the CAPEX in case 1, mainly, due to low heat transfer coefficient of air to water in case of heat recovery from VOCs abatement combustion exhaust. Annual CAPEX was also determined which was approximately 60000 \$/year for case 1 and 50000 \$/year for case 2. OPMEX has been calculated accordingly with the specified assumption, that became 12000 \$/year and 10000 \$/year for the first and second case, correspondingly. Furthermore, unit water treatment cost has been calculated for these cases which was 0.65 \$/m³ for case 1 and 0.57 \$/m³ for case 2 as shown in Fig. 5b. The unit water treatment cost found in the present study is in agreement to the literature [44], [45] and also comparable to the cost presented for other related water treatment technologies [44], [46] as shown in Table 7.

Table 7 Comparison of unit water costs from literature and present study

Water treatment technologies	Unit water cost, \$/m ³	Literature
Reverse Osmosis (RO)	0.69-0.8	[44]
Multi-effect distillation (MED)	0.81-1.48	[44]
Multi stage Flash (MSF)	1.1-1.27	[46]
Membrane Distillation (MD)	0.57-2.2	[44], [45]
	0.57-0.65	Present study

Table 8 summarizes the techno-economic advantages and disadvantages of the available industrial waste heat sources and the integration cases. It shows that the case 2 is the preferred arrangement of the low grade heat integrated MD setup due to minimum system requirement which was complemented with very low CAPEX and OPMEX. Resultantly, it has also provided with lower cost of water purification per cubic meter.

Table 8 Summary of potentials and pitfalls of low grade heat sources and integration cases

Heat Sources	Potentials	Pitfalls
Condenser outlet water stream	Reasonable flowrate for industrial scale plant. It can be used for preheating the MD feed up to 30°C	Insufficient temperature level so it cannot be used as standalone option
Hot air stream	Highest temperature level compared to other options. Reasonable option for heating up to temperature which comes under the technical limitations of the MD integrated system	Low energy content so it cannot be used as standalone option, large heat transfer surface area required to achieve the target temperature
Process cooling water exhaust stream	Reasonable flowrate, temperature level and energy content so it can be used as standalone option to heat the MD feed (4.17 kg/s) up to 80°C	-
Integration Cases		
Case 1	No external heat source required, reasonable unit water treatment cost	Higher cost of heat exchangers, higher retrofitting cost and resultantly higher CAPEX as compared to case 2
Case 2	Low CAPEX, no external thermal energy cost, lower unit water treatment cost compared to case 1	-

4. Conclusion

This study was focused on proposing a novel industrial application of MD system in nano-electronics industries for waste water treatment. Since energy has been considered as the most important parameter for economic feasibility of the water treatment process, two MD integration options with industrial waste heat have been studied. The investigation involved the thermal energy analysis and economic evaluation for large scale MD unit having capacity of 4.17 kg/s (15 m³/h). In the economic evaluation, comparison has also been made between the possible integrations with industrial waste heat. It is found out that 1.1 MW thermal power was required to preheat the MD feed up to 80 °C which was accompanied with enthalpy change of 0.28 MW across MD unit. Calculated annual permeate production was 3077 m³. The estimated water treatment cost was less than 1 \$/m³. Case 2, integration of process cooling water exhaust with MD, was found as the most optimized integrated system with reasonable annual CAPEX (50000 US dollars) and the least unit cost (57 US cents) for water purification per cubic meter.

Further study is recommended in order to address the issues including heat losses, thermal efficiency and energy efficiency since it will greatly affect the energy balance and cost of such integrations. Detailed economic assessment should also be considered to determine more about operational and maintenance cost.

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Nomenclature

A area, m²

c_p specific heat, J/ (kg K)

\dot{m} mass flowrate, kg/s

\dot{Q} enthalpy change, kW

T temperature, °C

Subscripts

a annual

c cold

d distillate

h hot

i inlet

o outlet

T total

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