



Design and operation of membrane distillation with feed recirculation for high recovery brine concentration



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ABSTRACT

Thermal-energy-driven desalination processes such as membrane distillation (MD), humidification dehumidification (HDH), and multi-stage flash (MSF) can be used to concentrate water up to saturation, but are restricted to low per-pass recovery values. High recovery can be achieved in MD through feed recirculation. In this study, several recirculation strategies, namely batch, semibatch, continuous, and multistage, are compared and ranked based on flux and energy efficiency, which together influence overall cost. Batch has higher energy efficiency at a given flux than semibatch and continuous recirculation because it spends more operating time treating lower salinity water for the same value of overall recovery ratio. Multi-stage recirculation is a steady-state process that can approach batch-like performance, but only with a large number of stages. Feed salinity rises during the batch operating cycle, and as a result feed velocity may have to be increased to avoid operating above the critical specific area wherein both GOR and flux are low due to significant heat conduction loss through the membrane. Finally, the choice of optimal membrane thickness for batch operation is compared to that of continuous recirculation MD.

1. Introduction

Conventional brackish and seawater reverse osmosis systems are not readily applied for further concentration, towards zero-liquid-discharge, of desalination brines, produced water from hydraulic fracturing, and industrial effluents. Thermal-energy-driven technologies such as membrane distillation (MD) and humidification dehumidification (HDH) are considered to be promising for such brine concentration applications as they can operate at ambient pressure and relatively low temperatures. These brine concentration applications are characterized by a high recovery ratio requirement. However, MD (except multi-effect designs) is restricted to a low value of per-pass recovery ratio, necessitating some form of brine recirculation.

In this study, we will

1. compare energy efficiency and pure water flux of various recirculation designs (batch, semibatch, continuous and multi-stage) for brine concentration.
2. elucidate the value of a control scheme that avoids counter-productive conditions characterized by high heat conduction losses across the membrane towards the end of the batch cycle as feed salinity increases.
3. comment on the choice of optimal membrane thickness for a batch

recirculation system, and compare its performance against a similarly optimized continuous recirculation process.

1.1. Motivation for high product recovery

MD systems without recirculation have a low recovery ratio (RR). RR is the fraction of incoming feed water separated as pure water:

$$RR = \frac{M_{\text{permeate}}}{M_{\text{feed}}} \quad (1)$$

where M_{permeate} is the mass of permeate produced and M_{feed} is the mass of feed to be treated. Instantaneous or per-pass recovery ratio through the MD module (which is achieved without recirculation) can be defined in terms of the pure water production rate (\dot{m}_p) and feed inflow rate (\dot{m}_f) as $RR_{\text{per-pass}} = \dot{m}_p / \dot{m}_f$. If ΔT_c denotes the change in temperature along the length of the cold (preheat) stream, applying energy conservation for the preheated feed stream gives $\dot{m}_f c_p \Delta T_c = \dot{m}_p h_{fg} + \dot{Q}_{m,\text{cond}}$, where $\dot{Q}_{m,\text{cond}}$ is the heat transferred by conduction across the membrane. Since $\dot{Q}_{m,\text{cond}} > 0$ and $\Delta T_c < T_{h,\text{in}} - T_{c,\text{in}}$,

$$RR_{\text{per-pass}} < \frac{c_p (T_{h,\text{in}} - T_{c,\text{in}})}{h_{fg}} \quad (2)$$

MD is operated at a top temperature below 100° C, often in

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Nomenclature*Roman symbols*

A	Area, m ²
AGMD	air gap membrane distillation
B	membrane permeability, kg/m ² ·s·Pa
B_0	membrane permeability coefficient, kg/m·s·Pa
c_w	specific cost of pure water, \$/m ³
c_f	specific cost per unit of feed treated, \$/m ³
C	cost factor, \$/m ³ (heating, cooling) or \$·kg/m ⁵ ·s (flux)
c_p	specific heat capacity, J/kg·K
d	depth or thickness, m
CGMD	conductive gap membrane distillation
DCMD	direct contact membrane distillation
GOR	gained output ratio = $\dot{m}_p h_{fg} / \dot{Q}_h$
h	heat transfer coefficient, W/m ² ·K
h_{fg}	enthalpy of vaporization, J/kg
HDH	humidification dehumidification
HX	heat exchanger
J	permeate flux, kg/m ² ·s
k	thermal conductivity, W/m·K
L	length of module, m
M	mass, kg
MD	membrane distillation
MVC	mechanical vapor compression
\dot{m}	mass flow rate, kg/s
N_{stages}	number of stages
NTU	number of transfer units
Nu	Nusselt number
Pr	Prandtl number
\dot{Q}	heat transfer, W
Re	Reynolds number
RR	recovery ratio of cycle
$RR_{\text{per-pass}}$	recovery ratio per-pass through system
s	salt concentration, g/kg
t	time, s
t^*	non-dimensional time
T	temperature, °C
TTD	terminal temperature difference, °C

U	overall heat transfer coefficient, W/m ² ·K
v	velocity, m/s
V_0	volume of recirculation loop, m ³
\dot{V}	volume rate, m ³ /s
w	width, m

Greek symbols

α	dp/ds , kg ² /g·m ³
δ_m	membrane thickness, μm
Δ	difference
μ	viscosity, Pa·s
ρ	density, kg/m ³
τ	cycle time, s

Subscripts, superscripts

b	brine
c	cold
ch	feed cold channel
cond	conduction
crit	critical value
eff,m	effective membrane property
f	feed channel
h	hot/heater
i	initial
HX	heat exchanger
in	inlet
m	membrane
max	maximum
MD	membrane distillation module
min	minimum
mu	make up
out	outlet
p	permeate
pw	pure water
($\bar{\quad}$)	average over a cycle, see Eqs. (4)–(5)

combination with low-temperature heat sources. If the ambient temperature is 25 °C, $RR_{\text{per-pass}} < 13\%$. In practice (e.g., Ref. [1]) the recovery with single-pass MD without recirculation is much lower, around 8% due to a lower $T_{f,\text{in}}$ and boiling point elevation of the salty feed stream leading to higher $\dot{Q}_{m,\text{cond}}$.

In contrast, in order to achieve zero-liquid-discharge, the desalination process would have to concentrate the salt solution up to saturation concentration (260 g/kg for NaCl), at which point the solution could be passed to a crystallizer. In this study, we will focus on desalinating a NaCl feed solution at 70 g/kg up to 260 g/kg. The corresponding required recovery ratio is $1-70/260 = 72.1\%$, much higher than the limiting value for a single-pass system. In order to implement such a high RR in a hypothetical single-pass MD process, the feed stream would have to be heated up to 500 °C, after being pressurized to prevent boiling. Our focus is on more practical, alternatives configurations with feed recirculation.

1.2. Options for high recovery with MD

The following operation strategies enable high overall pure water recovery employing a low-recovery single stage process:

(a) batch recirculation

(b) semibatch recirculation

(c) continuous recirculation

(d) continuous multi-stage recirculation

Fig. 1 shows a schematic representation of these alternatives.

Continuous recirculation [Fig. 1 (c)] is operated such that the brine leaving the MD module is at the required final brine salinity, and is continuously bled out of the system. In order to produce this output brine salinity, the inlet salinity to the desalination process has to be: $s_{h,\text{in}} = s_{b,\text{out}} \times (1 - RR_{\text{per-pass}}) = s_{f,\text{in}} \times \frac{1 - RR_{\text{per-pass}}}{1 - RR}$. For the brine concentration application considered in this study, since $RR > RR_{\text{per-pass}}$, the recirculated feed entering the MD module is at a higher salinity than the original feed stream. The remaining brine flow after bleed out is mixed with an appropriate quantity of incoming make-up feed to reach this inlet salinity at the module inlet.

In the MD literature, continuous recirculation has been a popular method for achieving high product recovery ratio because it is a steady-state process that is easy to implement and evaluate [2-5]. Recently, Lokare et al. [6] identified and evaluated the negative impact of continuous recirculation on both energy consumption and flux.

The multi-stage recirculation process illustrated in Fig. 1 (d) combines several single stage recirculation systems in series. Multi-stage recirculation also operates at steady-state and achieves a high overall

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