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Opinion

Dichotomy of Thermal Desalination: “To Evaporate or to Freeze, that is the Question”

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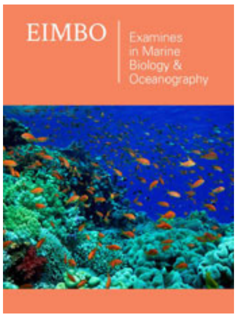

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Opinion

In this article, we overview the present technologies for freshwater production, such as membrane separation, thermal desalination, and water reuse/recycling. Specifically, we discuss the thermodynamic aspects of the desalination technologies to suggest freezing desalination as an alternative or supplement to mature desalination technologies.

Overview

Energy resources consist of non-renewable and renewable, based on their replenishing periods. Non-renewable energy, such as coal, oil, and natural gas, have higher production power than renewable energy consisting of solar, wind, hydro, biomass, and geothermal resources. Renewable energy resources are unlimited over the long term but are less efficient than non-renewable resources. Desalination technologies, primarily through seawater desalination, are known as energy-intensive processes because of the requirement to overcome feed osmotic pressure. Due to the continuous demand for high (electrical) energy consumption, desalination research often aims at achieving high rejection and production at low energy consumption. Freshwater production and water supply technologies can be categorized into thermal desalination, membrane separation, and water reuse/recycling [1-3]. Commercially available Reverse Osmosis (RO) membranes were designed in the early 1960s by Loeb and Souriranjan and proactively applied to salt rejection processes, from freshwater production by seawater desalination to ultra-pure water production in the semiconductor industry [4-7]. Although membrane technology became competitive with traditional thermal techniques, the membrane’s salt-rejection capability was often traded off with permeate flux, i.e., the production rate per unit membrane surface area [8,9].

Thermal energy utilization for freshwater production has a more than four-thousand-year history. Around 2000 B.C., Sanskrit writing records showed how to purify water through boiling, sunlight exposure, and charcoal filtering. From the 15th and 13th centuries B.C., Egyptian wall paintings depict the earliest siphoning apparatus for clarifying liquids. In 98 A.D., the first engineering report on water supply and treatment was written by Roman Senator Sextus Julius Frontinus in Rome. In modern times, thermal desalination primarily includes a series of efficient processes consisting of water evaporation followed by condensation.

Evaporative desalination

Distillation technologies have been developed to improve production efficiencies with smaller footprints. Vacuum distillation uses the basic thermodynamics principle of phase change [10,11], described by the Clausius-Clapeyron equation to link the evaporation enthalpy and vapor pressure [12]. Within a chamber, feed water evaporates at a pressure maintained lower than the vapor pressure at the chamber temperature. External condensers receive a vapor stream and produce distilled water with almost zero salinity, but thermal waste heat is unavoidable. Other processes are as follows. Vapor-compression distillation compresses the vapor present above the liquid, and the compressed vapor provides thermal energy higher than the evaporation enthalpy. Multi-Stage Flash Distillation (MSF) consists of a series of flash evaporators, fundamentally equivalent to vacuum distillation. Multiple-Effect Distillation (MED) is often used for seawater desalination, in which the incoming feed stream is sprayed and heated for efficient evaporation.

A hybrid technology of porous membrane and thermal desalination is Membrane Distillation (MD), consisting of various operation types [13-16], depending on the condensation mechanisms, such as direct contact [12,17,18], vacuum [19,20], sweep gas [21-23], air gap [24-26], and liquid gap [27-29] MD processes. Porous membranes, used in microfiltration and ultrafiltration, play the role of the contact barriers between the liquid and vapor phases of water, without any chemical reactions. After evaporation vapor molecules migrate through membrane pores by competing Brownian [30-35] and Knudsen [20,36] diffusion, and are either condensed or collected on the distillate side.

Water reuse/recycling has been less popular since it requires new development of or significant changes in social infrastructure while using conventional water treatment methods [37,38]. In reality, RO pretreatment is as essential as the RO process itself because imperfect pretreatment causes rapid fouling on membrane surfaces, potentially resulting in the process ceasing [39]. Thermal and membrane-based desalination produces warmer saline water than ambient water or highly concentrated brine stream, which may cause unexpected environmental impacts. Zero Discharge Desalination (ZDD) started as a critical research topic, which has become the future universal requirement at the city or county level [40-42]. In conventional thermal or membrane-based desalination, the continuous salt rejection produces either dense liquid or dry salts. At the theoretical limit of perfect recovery, the dry salt production rate is calculated using the seawater salinity, i.e., 35kg per ton or 103,200lb per million gallons of product water. Steady disposal of this large salt amount can include direct ocean disposal, incineration, or reuse as a defrosting agent on traffic roads. In an efficient water reuse/recycling system, zero discharge can be more feasible if the sources and types of water utilization are better understood.

Thermal desalination has focused on process optimization for effective water evaporation, followed by its condensation with minimum energy consumption. The ultimate obstacle to thermal desalination is the vapor pressure at operating temperature. For example, the gas pressure should be maintained below 0.03 atm in the chamber to evaporate water at 25 °C. Maintaining the low pressure state, closer to the vacuum phase, requires steady electrical consumption. Costs for the vacuum process are comparable to electricity consumption for high-pressure pumps for reverse osmosis processes. Nevertheless, the specific advantage of evaporation-based desalination is excellent salt rejection (even if seawater is used as a feed stream) due to water's intrinsic material properties.

In principle, thermal desalination is the traditional method that effectively excludes salt ions during the water evaporation process. Excessive thermal energy, higher than the latent heat demand for the phase change, breaks hydrogen bonds between two adjacent water molecules at the water-air interface. The latent heat of liquid water is represented as a function of absolute temperature T, such as $H(T) = l_0 - l_1 T$, where $l_0 = 57.075 kJ/mol$ and $l_1 = 4.3856 \times 10^{-2} kJ/mol$ K are estimated assuming the water is incompressible [12]. An accurate estimation of the evaporation enthalpy is 40.65kJ/mol or 2256.4kJ/kg at 100 °C. The vapor pressure is derived as $p(T) = p_0 \exp\left(\frac{-(l_0 + l_1 T \ln T)}{RT}\right)$, where R is the gas constant and p_0 =2.71690×1024mmHg or 3.5749×10^{21}atm. Note that the latent heat coefficient l_0 and l_1 also determines the vapor pressure. The required enthalpy to change the water phase from 0 °C ice to 100 °C vapor is the sum of 6.00kJ/mol for fusion, 7.54kJ/mol as sensible heat change, and 40.65kJ/mol for vaporization. The magnitude of freezing enthalpy is lower than the enthalpy required to increase water temperature from 0 °C to 100 °C and only 1/7 of the evaporation enthalpy. Thermal desalination technologies have reached their fundamental water phase-change limit in terms of vapor pressure, determined by the evaporation enthalpy and temperature, specifically by two constants p_0 and l_1. If a technology can reduce p_0 or l_1 noticeably, even for a short time, it would be a groundbreaking achievement in distillation-based thermal desalination. Besides, when ice is formed from a liquid phase, each water molecule forms a crystal-like structure, effectively pushing impurities away.

Freezing desalination

As briefly explained above, the freezing process removes salt ions in formed ice, consuming much less thermal energy. Freezing Desalination (FD) includes direct contact and indirect contact FD processes. In direct contact freeze desalination, the cold refrigerant directly contacts with the feed solution to vaporize the refrigerant at a lower pressure. The vaporization causes ice formation through heat removal from the feed solution. This method has a high ice production rate, but the created ice can be potentially contaminated due to volatile refrigerants. Refrigerants such as Butane have been studied due to its water-immiscible properties, but its flammability raises safety issues. Recently, Xie et al. enhanced heat transfer and phase separation in the freeze desalination process [43] by investigating the effects of initial refrigerant temperature, the ratio of produced ice to feed seawater mass, and ice production. In the spray freezing desalination studied by Liu et al. [44] warm seawater is sprayed at the top of a freezing tower and cold air (from the atmosphere or re-gasification of liquefied natural gas) is injected at the bottom of the tower. As the sprayed seawater droplets descend, heat transfer results in ice particle formation. A 200m high tower can yield 27.7kg/s fresh water (of 0.5% NaCl concentration) in an atmospheric temperature of -26 °C. An earlier study by Gao et al. [45] demonstrated the possibility of ~60% impurity removal using spray freezing [45]. Indirect contact FD consists of Suspension Freeze Crystallization (SFC) and Layer Freeze Crystallization (LFC) [46,47]. In contrast to direct contact FD, salt water and refrigerant/coolant are separated by thermally conducting walls.

In SFC, seed ice crystals are produced in salt water and grow larger via the Ostwald ripening mechanism in suspension [48]. Shin et al. [49] introduced surface-scraped freeze-crystallizer process to remove the ice crystal seeds creation step using a U-shaped agitator. The two-staged freeze desalination process resulted in 40% water productivity with 0.18 wt% salinity (with feed salinity of 3.5 wt%) [49]. Erlbeck et al. [50-52] used various SFC crystallizers to treat salt water with a 3.75 wt% NaCl concentration to produce a potable drinking water level of 0.01wt% with 22% water productivity [50-52].They used ice pressing as a post-treatment with a force as large as 37.4kN (or 100 bar), which significantly decreased the NaCl concentration to ~0.01wt%. Sahu et al. [53] used a U-shaped crystallizer in the SFC process to enable continuous ice production. Their method integrated cooling, chilling, and freezing processes to result in a water productivity of 15% and 0.8 wt% salinity of product water (with feed salinity ~3.5 wt%) [53].

LFC employs a cold surface to form a large ice crystal and facilitates efficient salt removal due to a smaller ice surface area. However, heat transfer between the plate and feed stream becomes less effective when ice layers are created, hindering further ice formation. Chen et al. [54] developed a supercooled water dynamic ice making method to achieve 50% water productivity with 0.045wt% salinity using a 3.5wt% feed stream with lower energy consumption due to smaller ice powder sizes [54,55]. Rich et al. [55] developed a dynamic layer crystallizer capable of obtaining a water productivity of 19% with 0.03wt% of product water salinity with a sweating post-treatment. Vertical freezing apparatus were operated in a batch-wise mode, where ice was forcefully grown in one direction [56,57] to demonstrate a water productivity of 51% and 0.91 wt% salinity of product water [57] and a ~25% water productivity and ~0.1wt% salinity [56].

Future perspective: Enthalpy, blessing or obstacle?

As one freshwater production technology, desalination aims to remove salt ions and impurities with low energy consumption. During with the desalination process, saline water enters a different phase from the liquid phase. The transition phases are membrane materials in the membrane separation and vapor phase in evaporative desalination, respectively. Evaporative desalination requires high energy consumption for the high rejection ratio; on the other hand, freezing desalination consumes a function of the thermal energy required for water evaporation but the salt rejection ratio is not high enough. The evaporation and melting enthalpies form energy barriers for water phase changes. The fixed enthalpy is a blessing under which materials and their phases in nature are balanced, but this is an obstacle to overcome for advanced thermal desalination. In freezing desalination, the temperature is adjusted for ice crystal formation, and pressure is maintained accordingly. Both evaporative and freezing desalination are subject to latent heat and vapor pressure, respectively, at the transition temperatures. Conventional thermal desalination aims to develop more energy-efficient processes, but this is limited by enthalpies for melting and vaporization. Novel technologies that can reduce latent heat and vapor pressure during a short period (i.e., of an order of seconds or shorter) can significantly reduce energy consumption and enhance phase change efficiencies.

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