



Freeze desalination of seawater using LNG cold energy



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ABSTRACT

With the aid of cold energy from regasification of liquefied natural gas (LNG), freeze desalination (FD) is an emerging technology for seawater desalination because of its low energy characteristics and insensitivities to fouling problems. This work aims to investigate the major operating parameters of FD such as coolant temperature, freezing duration, supercooling, seeding, agitation, crystallizer material and subsequent washing procedure on ice production and water quality. It was found that the optimal freezing duration per batch was 1 h for an iron crystallizer and 1.5 h for a glass crystallizer. The optimal coolant temperature should be around $-8\text{ }^{\circ}\text{C}$. The optimal amount of washing water to clean the raw ice was about 50 wt% of the raw ice. Over 50 wt% of the feed could be recovered as raw ice within 1 h, which means an overall ice recovery rate of higher than 25% (of the original seawater), considering the consumption of washing water. Both artificial and real seawater were tested under the optimized conditions. The total dissolved solid in the product ice was around 300 ppm, which met the World Health Organization (WHO) potable water salinity standard of 500 ppm. Therefore, the process parameters optimized in this study can be directly used for the freeze desalination of seawater.

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1. Introduction

Alleviating water scarcity is one of the major challenges facing the 21st century (Gude, 2016; Subramani and Jacangelo, 2015; Roberts et al., 2010). In light of the vast amount of seawater, much research has been conducted to desalinate seawater via processes such as reverse osmosis (RO) (Fritzmann et al., 2007; Sirkar et al., 1982; Kang and Cao, 2012), multi-effect distillation (MED) (Al-Shammiri and Safar, 1999), membrane distillation (MD) (Khayet and Matsuura, 2011; Wang and Chung, 2015; Zuo et al., 2016) and forward osmosis (FO) (Zhao et al., 2012; Valladares Linares et al., 2014; McGovern and Lienhard V, 2014). However, these processes also face challenges of either high-energy consumption or severe membrane fouling. Freeze desalination (FD) is an emerging desalination technology because of its insensitivities to fouling problems and the provision of cold energy from processes such as regasification of liquefied natural gas (LNG) in recent

years (Nebbia and Menozzi, 1968; Johnson, 1976; Shirai et al., 1999; Wang and Chung, 2012). The cold energy is the heat absorbed from ambient surrounding when converting LNG ($-162\text{ }^{\circ}\text{C}$) back to natural gas at LNG terminals (Wang and Chung, 2012; Miyazaki et al., 2000). It is one of high quality energy sources to cool other media, but most cold energy during LNG regasification has been wasted until today. Because the latent heat of ice fusion is only 1/7 of the latent heat of water vaporization (Rahman et al., 2006), it is much easier and more cost-effective to induce and harvest ice crystals from seawater by the cold energy in FD than to produce potable water by means of evaporation in many distillation processes (Wang and Chung, 2012; Cao et al., 2015).

There are two categories of freezing processes in FD; namely, direct and indirect contact freezing (Lu and Xu, 2010). The former mixes seawater with a liquid refrigerant directly. The refrigerant vaporizes and escapes from the top of the freezer. At the same time, seawater is cooled and then freezes. This process has a high production rate, but the requirements of the refrigerant are stringent. It should be nontoxic, chemically inactive, immiscible in water, and resistant to forming hydrates (Rahman et al., 2006). In contrast, indirect freezing uses walls to isolate the refrigerant from seawater (Zvinowanda et al., 2014). Heat exchange happens through the crystallizer walls with adjustable cooling conditions (Wang and Chung, 2012; Mandri et al., 2011). An intermediate coolant may

List of abbreviation: FD, freeze desalination; LNG, liquefied natural gas; MED, multi-effect distillation; RO, reverse osmosis; TDS, total dissolved solid; WHO, world health organization.

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be utilized to pass the cold energy from LNG to seawater at an appropriate temperature to facilitate ice production without incurring salts. Some previous studies used $-10\text{ }^{\circ}\text{C}$ or $-15\text{ }^{\circ}\text{C}$ for simplicity (Wang and Chung, 2012; Badawy, 2016; Efrat, 2011), while this work will find the optimal coolant temperature.

Various crystallizer configurations such as static layer crystallizer (Rich et al., 2010), progressive crystallizer (Fujioka et al., 2013), dynamic layer crystallizer (Rich et al., 2012) have been employed to investigate the ice formation in FD. In a static layer growth system, the seawater stays stagnant on a surface, the driving forces for heat and mass transfers are free convection and gravity. Thus, it would take a long time, usually more than 10 h per batch, to form a reasonable amount of ice crystals. In a progressive crystallizer, only one single ice crystal is present, which is at one end of the vessel. The ice-solution interface gradually moves towards the other end. However, the speed of the freezing front is highly constrained. A fast moving freezing front would inevitably cause concentration polarization. In a dynamic growth system, seawater flows down along a cold surface and ice crystals form underneath the flowing film. One of the major obstacles in this process is the salt entrainment (Reddy et al., 2009). The brine tends to be trapped in the interstices of ice crystals due to capillary force. Different from aforementioned approaches, the current study uses a simple batch reactor. We aim to have a deeper understanding of each basic parameter that would benefit FD development in years to come.

There are two major challenges in the FD technology. One is the amount of salts adsorbed on the ice crystals, while the other is the amount of washing water to remove these salts. Since ice crystals generated in FD have very small sizes, they have an enormous specific surface area. Their surfaces are usually covered by a slew of salts due to the strong adhesive force between ice and brine. Therefore, one must wash ice crystals to remove these salts. The amount of washing water has been overlooked in most of past studies, but this study would revisit this crucial issue and optimize the amount of washing water. Therefore, this study may provide useful insights and operational data for the entire FD process. In addition to these challenges, FD also faces the difficulties of being accepted as a new technology from industries, especially when it is perceived as costly and mechanically complex (Randall and Nathoo, 2015).

2. Background and theory

2.1. Pivotal role of washing in FD

Freezing pure water and saline water are distinctly different. Without the disturbance of soluble ions, the ice crystals grow on top of one another. Their sizes can be very large. Visibly, the ice could form an entire chunk in the cylinder. However, water does not freeze in the same manner when many ions are present. Instead, dendritic ice crystals (Teimat, 1980; Lorain et al., 2001) will occupy the space and form a slurry.

Brine adheres to ice crystals considerably due to the massive interfacial tension (Shwartz and Probst, 1968). A thorough separation of ice and brine require accessory procedures other than simply draining off or normal filtration. The design of washing columns has been actively investigated (Shwartz and Probst, 1968; Harriott et al., 1969) and used in pilot plants (Efrat, 2011). The ice slurry is introduced to the bottom of the washing column that is filled with clean water. Ice crystals float up by the buoyancy force and the brine trapped in interstices would be displaced. Clean ice can then be harvested at the top of the washing column. However, there exists a serious dilemma regarding whether to use agitation and pulsing. Without them, the mass transfer would be extremely slow because the buoyant movement is severely

inhibited by the adherence force between ice and water. If agitation and pulsing are adopted, their contributions to longitudinal mixing would disrupt the concentration gradient distribution (Sherwood and Brian, 1966). Once the upper portion of the column contains a high amount of salts, the column is no longer able to purify ice. In contrast, washing ice crystals with the aid of vacuum filtration can drain out salty water within seconds. Not only can it lower the salt impurity on the ice crystals to be almost negligible but also significantly reduce the energy consumption for the washing procedures.

2.2. Supercooling and nucleation

Supercooling is the process where a liquid is cooled below its freezing point without crystallizing. For instance, pure water has to be cooled below $0\text{ }^{\circ}\text{C}$ to initiate freezing. Although $0\text{ }^{\circ}\text{C}$ is widely understood as its freezing point, it is rather an equilibrium point during the freezing process. The real temperature for water to start freezing depends on when nucleation happens. According to crystallization kinetics, a trace amount of ice lattice larger than the critical size must be generated (Tiller, 1992). In other words, there is a minimum embryo-crystal size (i.e., the critical radius) for the embryo to grow spontaneously. In fact, for a given temperature there will be a critical radius. The relationship among them is reflected by the Kelvin equation (Teimat, 1980):

$$r^* = \frac{2\sigma T_f}{L_f \Delta T} \quad (1)$$

where r^* is the critical radius, T_f is the freezing temperature, L_f is the latent heat of fusion, ΔT is the temperature difference departed from the freezing point, and σ is the interfacial tension between water and ice that is estimated to be 22 erg/cm^2 . The critical radius is then plotted relative to water temperature in Fig. 1(a).

Once ice has the critical radius, nucleation may take place by either homogeneous or heterogeneous nucleation. Theoretically, a cluster of about 25 water molecules would form ice at $-41\text{ }^{\circ}\text{C}$ via homogeneous nucleation. In reality, it is difficult to cool the water down to near this temperature without forming ice because the nucleation barrier is largely reduced by the existence of impurities that lead to heterogeneous nucleation. As depicted in Fig. 1(b), when water molecules wet an impurity surface, a small portion can actually correspond to a much larger radius (Muldrew and McGann, 1999). As a result, this cluster of water molecules is likely to stabilize themselves at a much higher temperature and spontaneously set off the heterogeneous nucleation. The ice crystal would then grow irreversibly as long as the cooling condition is maintained.

Additionally, the supercooling condition can be interrupted when nucleation is triggered by external nucleus (Randall et al., 2009). If any seed ice is added into the supercooled solution, the status of supercooling would cease immediately and a large amount of ice crystals would form shortly. This process releases so much heat that the system temperature would rise shortly to the equilibrium point, i.e., freezing point. In the absence of a seed, the temperature at which ice crystallization will occur is highly stochastic (random). Seeding helps to overcome this variability because ice crystallization will occur at the same temperature with seeding (Randall et al., 2012).

2.3. Freezing-point depression

Freezing-point depression is a phenomenon where the freezing point of a solvent is decreased by the presence of solutes (Gooch, 2011; Halde, 1980). It is noted that the lowering of the freezing point does not depend on the nature of the solutes. Instead, it is

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