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Effect of average refrigerant temperature on freezing-based combined seawater desalination

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ABSTRACT

The raw seawater taken from Bohai Bay was frozen into ice flakes by the patented experimental platform. The arithmetic average temperature of the intermediate refrigerant T_{ar} was regulated from –18.75°C to –50°C. The effect of T_{ar} on ice flakes desalination was investigated. Salinity, total dissolved solids and Cl^-, Ca^{2+}, Mg^{2+} concentrations of the product ice samples were measured. The experimental results showed a special phenomenon that the purity of ice flakes obtained from the employed freezing method was not improved with the increase of T_{ar}. We tried to explain this phenomenon through the ice formation process and the ice flake construction. Then the ice flakes were treated with gravity-induced, centrifugation, or gravity-induced plus centrifugation methods to ameliorate the ice purity. After the combined desalination treatments including freezing and gravity-induced desalination (FGD), freezing and centrifugal desalination (FCD) and freezing, gravity-induced and centrifuging desalination (FGCD), the purities of the ice flake samples increased with T_{ar}. It can be concluded that effects of the freezing-based combined desalination methods tested in this research could be listed in the descending order as FGCD, FCD and FGD and increasing T_{ar} would benefit to ameliorate desalination effect for these combined processes.

Keywords: Seawater desalination; Freezing desalination (FD); Combined desalination process; Liquefied natural gas (LNG) cold energy

1. Introduction

China is one of the water shortage countries and the fresh water resource per capita is about a quarter of the world average level. Nearly 400 cities in all more than 600 cities are facing water shortage problem and among them, more than 100 cities are in serious water crisis. More than 100 million people in China's rural areas drink brackish water and other unsafe water. Seawater desalination is a potential way to solve the water shortage problem, and China's long coastlines provide favorable conditions for the development of seawater desalination industry. For present seawater desalination technologies, reducing the cost is a very prominent problem.

More than 300 years ago, Danish physician Thomas Bartholinus found that fresh water can be obtained by melting ice formed in seawater [1]. From then on, freezing desalination has been investigated as one of desalination technologies. Theoretically, freezing method possesses the following advantages [2]. (1) From the perspective of energy consumption, the latent heat of ice melting is 334.7 kJ/kg under normal atmospheric pressure, which accounts for 1/7 of water evaporation heat (2,259.4 kJ/kg at 100°C). Therefore, freezing desalination (FD) is more energy saving than

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distillation desalination. (2) Compared with distillation method, freezing method operates in a low temperature, so that the corrosion of equipment is lightened. (3) Compared with reverse osmosis membrane method, freezing method does not need complex pretreatment process.

Because of strong hydrogen bonds, the water molecules are closely integrated and salt ions would be squeezed out in the freezing process. Therefore, freezing seawater should produce very high purity ice. In fact, the diffusion rates of salt ions in the progressive freezing process must be greater than the advance speed of ice front or else, salt ions will be wrapped in ice in the form of concentrated brine, which is so-called brine pocket [3]. The existence of brine pocket causes salinity increase of ice layer or ice block. For fine ice crystals produced in suspension freezing process, the cause to decrease their purity is that more brine are adhered to them because of their large specific surface areas.

To decrease ice product salinity, many reports were published on various possibilities of ice–brine separation for either ice block or fine ice crystals. Some research work focused on vacuum-freezing vapor-compression (VFVC) method which makes seawater evaporate and freeze simultaneously by creating the triple point through decreasing pressure and temperature [4]. As compressors and pumps have to be utilized in maintaining vacuum and operating the desalination system, the reported power consumption ranged from 48 kWh/1,000 gallons at the highest production rate of 100,750 gpd (gallons per day) to 64.4 kWh/1,000 gallons at the average capacity of 83,116 gpd. Condensing the vapor can obtain fresh water, whereas the individual ice crystal consisting pure water is coated with a layer of concentrated brine which must be removed by washing to get fresh water [5]. Study on counterwashers to wash ice crystals was presented in some articles [6,7].

There were many studies focusing on freezing desalination devices. Williams et al. [8] designed desalination equipment consisting of water distributor, refrigerated inclined plate, evaporator coils, re-circulating pump and collecting tank. Using this equipment, some experiments to freeze 3.5 wt% NaCl solutions were carried out from 15 to 90 min and the NaCl removal efficiency was maintained at about 50% and the highest yield rate was about 45%. Fujioka et al. [9] designed a progressive freezing desalination device with a stirring function to investigate desalination performance with NaCl solution. The authors focused on the effect of three parameters, namely the advance speed of the ice front, the circumferential velocity of the stirrer, and initial concentration by conducting the laboratory experiments, and proved that to the NaCl solution with initial concentration by 3.5 wt%, the optimal advance speed of the ice front and the circumferential velocity of the stirrer are 0.5 cm/h and 1.45 m/s, respectively. The highest NaCl removal efficiency under optimal conditions was about 50%, and the fraction of solid phase was less than 50% at this time. Similar progressive freezing device was also appeared in wastewater treatment [10] and concentration of liquid food [11,12]. Although the salinity of the seawater can be reduced by the progressive freezing method, the quality of the desalinated water can hardly achieve the requirements for industrial and domestic water [9,13,14], and therefore, further treatment processes are necessary.

Many studies have shown that partial melting of ice under gravity contributes to desalination. Our previous experimental study also proved that for either brine ice made of water/NaCl solution frozen in refrigerator or raw seawater ice produced by our patented experimental platform, the salt removal efficiency increases with the increase of gravity-induced brine drainage proportion in both FGD and FGCD processes [15,16]. Rich et al. [17] did some experiments for water/NaCl solutions with different initial concentrations as well as seawater samples taken from Nice, Rabat and Marseille using a dynamic layer crystallizer which can produce layer ice and in which layer ice can also sweat at the studied temperatures. Their research focused on the effects of initial concentration, freezing temperature, freezing time, sweating time and sweating temperature on desalination. It was demonstrated that after freezing step, the ice salinity can't satisfy the drinking water standards. In their experiments, ice was further purified with sweating step to drain out the trapped solution pockets, and three of the experimental samples reached the requirement of drinking water standards with the whole duration including freezing and sweating processes from 8 to 28 h. They concluded that higher sweating temperature could lead to higher ice purity with more ice re-melted, that is to say, lower ice yield rate as well as more energy consumption. Therefore, trade-off should be conducted between process duration and ice yield rate. Gu et al. [18,19] did a lot of research on gravity-induced desalination of natural sea ice. They found when ambient temperature was between –4°C and 3°C, the drainage volume was very small but the concentration of the discharge was high, which is the most suitable brine discharge temperature range for natural sea ice desalination. They conducted gravity-induced desalination experiments using natural sea ice of 9,000 and 12,000 m^3 collected from Bohai Bay in winters of 2009 and 2010, respectively. The ice was put in a desalination pool. In 2009, the experiment lasted 80 d, and produced desalinated water of 4,500 m^3 with the salinity of 0.8‰. In 2010, the experiment lasted 90 d and yielded 6,700 m^3 water with the salinity of 1.4‰. Badawy [20] conducted freezing and melting experiments to desalinate sea water of Umluj beach, Red Sea. They used the freezer with indirect contact method to freeze sea water and changed crystallization ratio through regulating freezing time. It was presented that single-stage freezing could remove maximum 50% of the salt in the feed raw seawater. Continuous cycling of the freezing/watering/melting process and seawater ice gradual melting process could improve the purity of produced ice greatly. In their experiments, with eight freezing/ watering/ melting cycles or 6 h gradual melting process could finally make the TDS of melt water reach the level of drinking water. The mass loss of FM cycling method reached 66%, while the mass loss of gradual melting method reached 98%.

Some centrifugal desalination research was also conducted to improve ice purity. Chen et al. [21] demonstrated that the centrifugal rotation speed of 2,000 rpm can help most of the brine pockets to overcome the adhesive viscosity and surface tension and therefore to be separated from gray white ice. Wei et al. [22] found out that the salinity of natural sea ice decreases below 0.3 ‰ when separation time reaches 1 min with the separation factor of 1,100, which is defined as the ratio of centrifugal force and gravity force. Luo et al. [13] demonstrated that the method of crushing ice and centrifugation (CIAC) can help remove large amount of solute from ice. The brackish water samples with initial TDS range from 1,320 to 8,350 ppm were frozen to make the ice growing from top to bottom by unidirectional heat transfer method. Through freezing part of the brackish water, 57.88% to 48.38% of TDS in the ice was removed. Then CIAC method was used to further purify the ice samples and 30.91% to 47.28% more TDS was removed.

Some other ice purifying treatment methods such as washing [23] or soaking [24,25] were also studied and demonstrated effectively. Chang et al. [23] used Freon® gas to mimic the LNG regasification process in a LNG vaporizer to freeze NaCl solution with concentration of 3.5 wt%. They investigated the effect of step-by-step washing procedure of tap water on ice production and water quality, and got the ice product with salinity below 0.4 g/kg. Although the wastewater from the last step could be reused to wash the raw ice, the tap water consumed was still about 50 wt% of raw ice. Gu et al. [24] studied the soaking effect on natural sea ice. They found that the salinity of soaking solution, soaking time and the volume of soaking solution played an important role in desalination effect.

LNG occupies only about 1/600 of the volume of NG (natural gas) at atmospheric pressure under –162°C, which makes its transportation and storage conveniently. LNG is usually transported by ship to LNG receiving terminal constructed nearby the seaboard and generally has to be gasified before being used. During the gasification process, about 837 kJ/kg latent cold energy ranging from –162°C to ambient temperature can be released. From the theoretical perspective of improving thermodynamic efficiency, LNG cold energy should be used in cascade. For example, the lowest temperature cold energy can be used in air separation and electricity generation; the higher temperature cold energy can be used in food storage or air conditioning. Whereas, it is difficult to coordinate the product market, economic feasibility as well as technologic feasibility in real practices. The real situation is that most of the cold energy is dismissed to seawater near LNG receiving terminals, which causes both energy loss and negative influence on seawater environment. If the cold energy can be employed to desalinate seawater, the freezing desalination cost will be greatly reduced. 22 LNG receiving stations in China are put into operation now, with a total receiving capacity of nearly 70 million tons/year. This might bring a development opportunity to seawater freezing desalination. Some previous research was carried out on simulation and theoretical analysis of LNG cold energy transfer process including model establishment and process optimization other than experiments on desalination effect [26–30]. Some other research was concentrated on effects of parameters, such as refrigerant temperature, seawater flow rate and spraying nozzle number of the prototype system utilizing LNG cold energy on salinity of ice product [31,32].

LNG cold energy can be used to freeze seawater through direct or indirect heat transfer method. Considering that ice formation on cold surfaces is relatively simple and peeling the flake ice from cold surface is easier than separating the granular ice from the solution, indirect heat transfer method was finally employed in our experimental system. In laboratory of Beijing University of Civil Engineering and Architecture (BUCEA), the patented experimental platform (with the authorized number ZL 2016208330324 in China), which can use LNG cold energy, was utilized to freeze seawater taken from Bohai Bay near an LNG receiving terminal into ice flakes. Many research as well as our own previous study [15,16,33] demonstrated that the salinity of ice produced only by freezing process is far more above the requirement of potable or industrial water standards and further treatment is necessary to purify the ice flakes. Different from consuming pure water to wash brine attached to fine ice crystals formed using VFVC method, washing is not the obligatory process to separate the contained brine pockets from ice flakes and instead, ice flakes can be treated further by other methods such as gravity-induced desalination, centrifugal desalination or the combination of above two methods. As the LNG cold energy distributed in wide temperature range, it is meaningful to study the influence of arithmetic average intermediate refrigerant temperature T_{ar} on seawater desalination. In this paper, T_{ar} was regulated from –18.75°C to –50°C and the effect of T_{ar} on FD, FGD, FCD and FGCD methods were investigated.

2. Materials and methods

2.1. Seawater freezing experimental platform

Fig. 1 shows the patented experimental platform which can use LNG cold energy to freeze seawater. In the experiment, liquid nitrogen having a similar gasification temperature was used instead of LNG for safety reasons. In order to better control the parameters of the experimental process, complete sensors and control devices as well as computer remote monitoring system were installed. The flow diagram of the freezing experimental platform is shown in Fig. 2. The raw seawater was frozen into ice flakes using indirect heat exchange method. The platform consists of liquid nitrogen gasification equipment, intermediate refrigerant circulation and flake ice maker. The liquid nitrogen exchanged heat with intermediate refrigerant in the cryogenic heat exchanger and then was gasified. The intermediate refrigerant cooled

Fig. 1. Seawater freezing experimental platform.

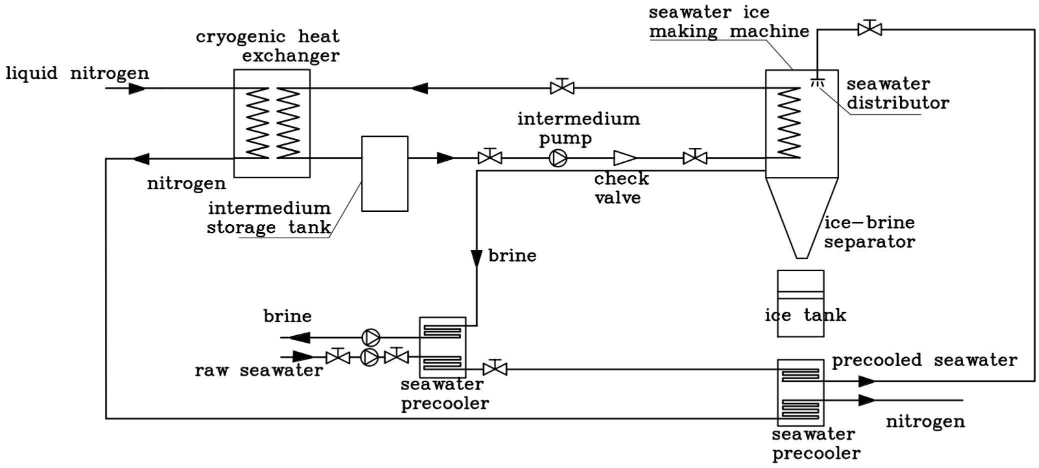

Fig. 2. Flow diagram of the platform.

by liquid nitrogen was circulated to ice maker by the intermedium pump. The flake ice maker, which is an indirect heat exchanger, was selected in the experiments to produce ice continuously. The system can assure the industrial seawater freezing process together with the LNG gasification process without interruption if it is used in real project. In LNG gasification process of receiving terminals, the flake ice maker can also be connected on parallel with the intermediate fluid vaporizers (IFVs) through which the LNG cold energy is released to seawater and the two kind of equipment can use the same intermediate fluid. In this way, no additional safety problem would be arisen by ice making system. The ice making equipment can also replace the backup IFVs depending on their capacities. In that case, parts of investment on desalination equipment can be recovered in advance by decreasing investment on IFVs.

2.2. Water quality test method

For both raw sea water and the ice product, salinity, TDS and the concentrations of Cl^-, Ca^{2+} and Mg^{2+} were tested as indicators of desalination effect. The salinity of the water samples was measured using the salinity meter of model HTATC 212 with the accuracy of 0.2% and the measuring range from 0 to 25%. The concentrations of the typical ions, i.e. Cl^-, Ca^{2+}, Mg^{2+}, in seawater and water products were measured using the same chemical titration as what used in the studies by Yang et al. [16,33]. The content of Cl^- was titrated using standard silver nitrate solution with potassium chromate as indicator. The total content of Ca^{2+} and Mg^{2+} was titrated with ethylene diamine tetraacetic acid disodium salt (EDTA) and eriochrome black T was used as indicator. The content of Ca^{2+} was titrated with EDTA and calconcarboxylic acid was used as indicator. Then with the total content of Ca^{2+} and Mg^{2+} minus the Ca^{2+} content to get the Mg^{2+} concentration. The TDS of the water samples was measured with the digital portable instrument of model Ultrameter II 4P (conductivity/TDS/resistivity/temp., Myron L Company, USA) with the measuring range from 0 to 200,000 mg/L, the accuracy of ±1% of reading and the auto temperature compensation.

3. Experiments and results

Our previous research proved that higher salt removal efficiency would be induced by higher freezing temperature when the same mass fraction of NaCl/water solution was frozen into ice in a refrigerator [15], which was in accordance with some other research aiming at ice layer formation process [13,14]. It seems that above conclusion was also demonstrated by some research on artificial seawater ice flakes using LNG cold energy with the refrigerant temperature from –15.4°C to –20.4°C [31,32]. Considering of wider available temperature range that could be created by LNG cold energy, it is meaningful to conduct more experiments in exploring the effects of lower refrigerant temperature or wider refrigerant temperature range on seawater desalination.

Raw seawater was taken from Bohai Bay with initial salinity, TDS and ion concentrations as shown in Table 1 and then was frozen into ice flakes using above-mentioned patented experimental platform. Keeping the other operational conditions of the experimental platform constant, the average temperature of the intermediate refrigerant was changed by adjusting the liquid nitrogen flow rate, and thereby the ice formation velocity was changed along with the surface temperature of the ice maker. The intermediate refrigerant temperature passing through the ice maker was calculated by the arithmetic average of the inlet and outlet temperatures and regulated from –18.75°C to –50°C. The arithmetic average refrigerant temperature T_{ar} conditions are shown in Table 2 and 500 g seawater ice sample was fetched under every constant operational condition to be melted for measuring the salinity and TDS. The freezing

Table 1
Salinity, TDS and the ion concentrations of raw seawater

C_0 (%)	3.3
C_{0TDS} (mg/L)	32,325
C_{0Cl^-} (mg/L)	17,355.95
$C_{0Ca^{2+}}$ (mg/L)	421.33
$C_{0mg^{2+}}$ (mg/L)	1,129.6

Table 2
Freezing salt and TDS removal efficiencies under different T_{ar}

Cases	T_{ar} (°C)	R(FD) (%)	R_{TDS}(FD) (%)
Case I–(1)	–18.75	6.06	6.37
Case I–(2)	–20.85	9.38	6.14
Case I–(3)	–23.5	6.25	6.42
Case I–(4)	–27.5	9.09	6.52
Case I–(5)	–30.2	9.38	8.08
Case I–(6)	–31.3	9.09	9.2
Case I–(7)	–34.65	9.38	9.41
Case I–(8)	–37.95	6.06	9.89
Case I–(9)	–40	8.09	10.2
Case I–(10)	–40.2	9.09	9.45
Case I–(11)	–41.05	9.09	10.33
Case I–(12)	–42.15	10.61	10.68
Case I–(13)	–43.2	11.09	11.04
Case I–(14)	–45.1	12.12	11.08
Case I–(15)	–50	16.18	15.34

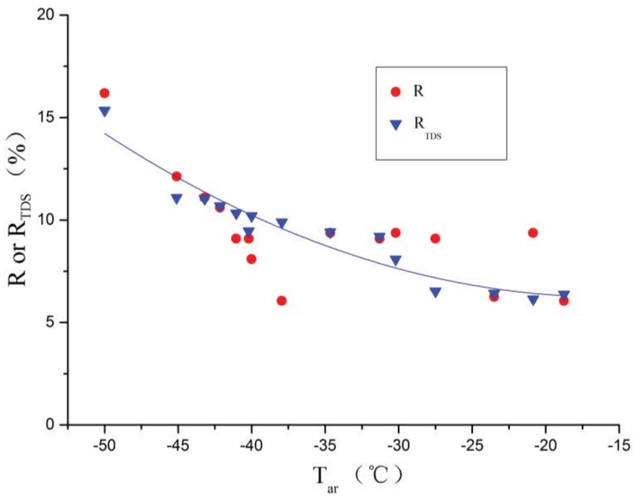

Fig. 3. Relationships between R(FD), R_{TDS}(FD) and T_{ar}.

removal efficiencies of salt R(FD) and TDS R_{TDS}(FD) were calculated using Eqs. (1) and (2) as shown in Table 2, and the relationships between R(FD), R_{TDS}(FD) and T_{ar} are presented as Fig. 3.

$$R = \left(1 - \frac{C_i}{C_0}\right) \times 100\% \quad (1)$$

$$R_{\text{ion}} = \left(1 - \frac{C_{\text{ion}}}{C_{0\text{ion}}}\right) \times 100\% \quad (2)$$

Obviously, the conclusion obtained by much previous research that the salt removal efficiency increases with the freezing temperature [13,14,30,31] is not adaptable here. On the contrary, it seems that higher salt and TDS removal efficiencies could be got under the lower T_{ar}, for example, R(FD) and R_{TDS}(FD) reach the maximum values of 16.18% and 15.34% when T_{ar} is –50°C. And when T_{ar} are –18.75°C and –20.85°C, respectively, R(FD) and R_{TDS}(FD) reach their minimum values of 6.06% and 6.14%. The special phenomenon might be explained from the freezing process of flake ice maker as well as the construction characteristics of the ice flakes. The ice flakes obtained at different T_{ar} are shown in Fig. 4. It can be observed that the ice flakes produced under –26.5°C have loose texture with smaller size,

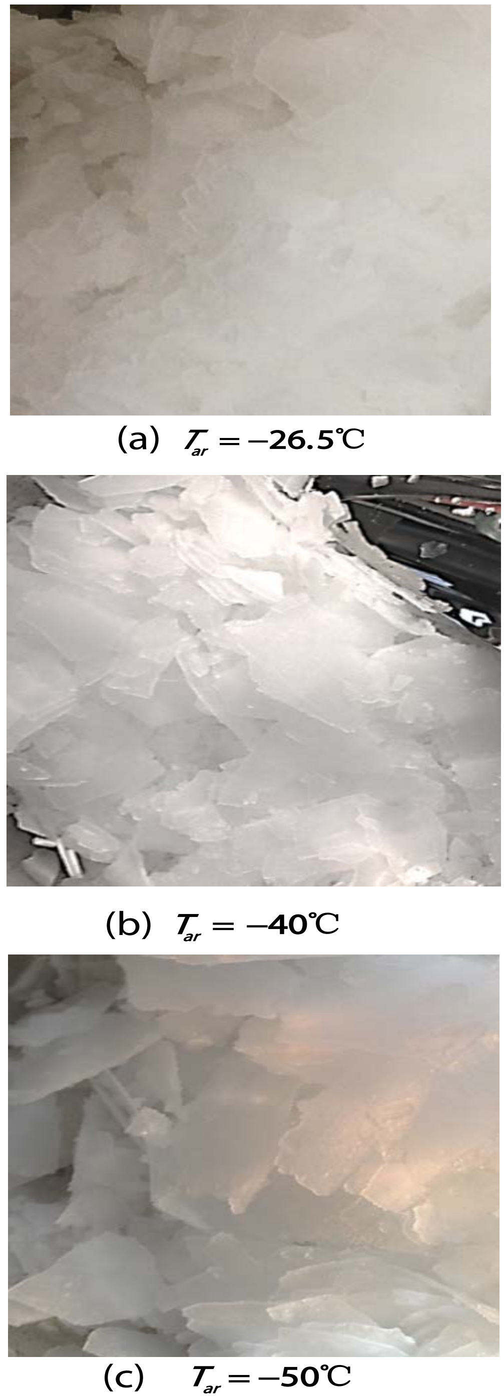

Fig. 4. Ice flakes obtained at different T_{ar}.

whereas the ice flakes produced under –50°C are much more compact and harder with larger pieces. The ice flakes produced under –40°C are medium in size and density. We guess that loose texture and small size ice flakes with more porous or gaps could contain more raw seawater, which was sprayed on the ice flakes during the ice maker rotation process before the ice flakes been separated from the ice maker. Limited by the experimental conditions, we did not carry out further study on the ice flake texture. Therefore this hypothesis is still waiting for proof and study on micro-construction of ice flakes should be conducted in the future. Besides that the experimental results showed that the salt removal efficiencies of the ice flake samples are still too low and merely artificial fast and continuous freezing process can only produce ice with the salinity far more above the requirement of potable or other industrial water standard. Further treatments are necessary to improve the purity of the ice flakes.

In this paper, seawater ice flakes produced under every T_{ar} as shown in Table 2 were taken as experimental samples and three combined desalination processes including FGD, FCD and FGCD were utilized in the experiments. For each T_{ar} condition, three 500 g ice samples were taken to be treated by gravity-induced process, centrifugal process as well as gravity-induced plus centrifugal process, respectively. In order to decrease the process duration, for the experiments with gravity-induced stage, ice samples were put on filters in ambient with temperature of 18°C and when the brine drainage proportion reached 10%, the gravity-induced process stopped. For the experiments with centrifugal stage, ice samples were treated using filtering centrifuge of type TD5F with the rotation rate of 3,000 rpm for 2 min. The gravity-induced drainage device and centrifuge are shown in Figs. 5 and 6. The effects of T_{ar} on seawater ice flake desalination using the three processes were compared.

After FGD, FCD or FGCD treatment process, the ice product for every sample was melted and the salinity, TDS together with the concentrations of ions including Cl^-, Ca^{2+} and Mg^{2+} were measured. The removal efficiencies of salt, TDS and measured ions as well as ice yield rates R_{iy} were calculated using Eqs. (1)–(3), and the results were listed in Tables 3–5.

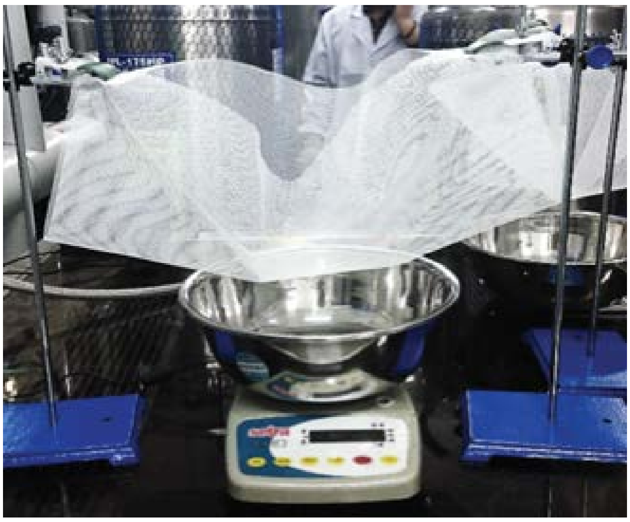

Fig. 5. Gravity-induced melting device.

Fig. 6. TD5F centrifuge.

$$R_{iy} = \frac{M_{ip}}{M_0} \times 100\% \quad (3)$$

According to the data of FD, FGD, FCD and FGCD process experiments in Tables 2–5, the relationships between R and T_{ar} and the relationships between R_{TDS} and T_{ar} are plotted in Figs. 7 and 8, respectively. The relationships between R_{iy} and T_{ar} of FCD and FGCD processes are plotted in Fig. 9. It can be observed that different from above-mentioned relationships between R(FD), R_{TDS}(FD) and T_{ar}, removal efficiencies of salt, TDS and ions after FGD, FCD and FGCD processes all increase with T_{ar}. It might indicate that for the ice flakes which were produced under higher T_{ar} with loose texture and smaller size, brine pockets contained in these ice flakes as well as the raw seawater adhered to them are easier to be separated, no matter how their initial salt concentrations are. On the contrary, for the ice flakes with more compact and harder texture and larger pieces, which were produced under lower temperature, it is more difficult to separate brine from ice. It can be concluded that although LNG cold energy has wide available temperature range and relative higher freezing salt removal efficiency can be obtained to the ice flakes produced under lower T_{ar}, after FGD, FCD or FGCD processes, it is still the ice flakes produced under higher T_{ar} that can arrive higher purity with higher salt removal efficiency. Centrifugal process is more efficient than the gravity-induced process of 10% brine drainage proportion and the range of R increases from 19.7%–34.34% for FGD to 62.5%–84.38% for FCD meanwhile, the range of R_{TDS} increases from 19.46%–33.23% for FGD to 63.59%–81.39% for FCD. The experiments also proved that additional gravity-induced process with brine drainage proportion of 10% is helpful for improving the desalination effect of centrifugal process. R was increased from 62.5%–84.38% for FCD to 70.79%–90.63% for FGCD and R_{TDS} was increased from 63.59%–81.39% for FCD to 73.01%–87.35% for FGCD. Meanwhile, the ice yield rate was decreased from 44.09%–66.2% for FCD to 35.61%–46.02% for FGCD. These experiments demonstrated that

Table 3
Salt, TDS and ion removal efficiencies after FGD process

Cases	*R*(*FGD*) (%)	R_{TDS}(*FGD*) (%)	$R_{Ca^{2+}}$(*FGD*)(%)	$R_{Mg^{2+}}$(*FGD*)(%)	R_{Cl^-}(*FGD*)(%)
Case II–(1)	34.34	33.23	27.85	23.51	81.18
Case II–(2)	33.33	32.74	27.85	21.81	80.64
Case II–(3)	30.30	26.98	27.85	20.96	79.41
Case II–(4)	30.30	27.18	27.85	19.26	76.31
Case II–(5)	24.24	25.18	27.85	18.41	73.82
Case II–(6)	24.24	27.4	27.85	18.62	76.31
Case II–(7)	22.62	23.62	27.85	18.41	64.55
Case II–(8)	21.30	22.86	27.85	17.56	63.77
Case II–(9)	22.86	23.51	27.85	17.96	62.41
Case II–(10)	23.11	23.25	27.85	17.23	62.64
Case II–(11)	20.61	21.91	27.85	16.99	61.55
Case II–(12)	21.21	22.63	27.85	16.99	60.59
Case II–(13)	19.70	20.17	27.85	16.56	59.32
Case II–(14)	19.70	19.46	27.85	16.11	58.19
Case II–(15)	22.36	23.44	27.85	15.76	53.64

Table 4
Salt, TDS and ion removal efficiencies and ice yield rate after FCD process

Cases	*R*(FCD) (%)	R_{TDS}(FCD) (%)	$R_{Ca^{2+}}$(FCD) (%)	$R_{Mg^{2+}}$(FCD) (%)	R_{Cl^-}(FCD) (%)	R_{iy} (FCD) (%)
Case III–(1)	84.38	81.39	82.91	81.73	84.00	44.09
Case III–(2)	81.25	76.74	77.22	81.30	82.77	42.73
Case III–(3)	75.22	75.22	81.01	72.80	81.95	42.37
Case III–(4)	75.76	74.7	77.22	72.80	81.55	39.28
Case III–(5)	81.82	74.63	73.42	71.95	81.13	49.99
Case III–(6)	72.73	75.45	65.82	69.41	80.18	48.79
Case III–(7)	75.76	69.64	73.42	64.31	79.5	44.48
Case III–(8)	75.00	76.44	62.03	55.81	77.59	50.23
Case III–(9)	75.76	71.66	61.17	61.84	76.36	55.04
Case III–(10)	75.00	72.17	60.57	61.70	76.19	63.10
Case III–(11)	72.73	72.38	59.91	60.55	75.32	57.88
Case III–(12)	69.70	71.52	58.97	58.69	80.50	63.19
Case III–(13)	72.73	67.39	56.71	57.41	74.30	64.70
Case III–(14)	62.50	65.92	54.79	56.40	74.77	65.98
Case III–(15)	65.38	63.59	52.34	51.79	72.82	66.20

Table 5
Salt, TDS and ion removal efficiencies and ice yield rate after FGCD process

Cases	*R* (FGCD) (%)	R_{TDS} (FGCD) (%)	$R_{Ca^{2+}}$ (FGCD) (%)	$R_{Mg^{2+}}$ (FGCD) (%)	R_{Cl^-}(FGCD) (%)	R_{iy} (FGCD) (%)
Case IV–(1)	90.63	87.35	86.71	88.53	88.55	36.81
Case IV–(2)	87.50	87.10	86.71	88.53	87.32	36.52
Case IV–(3)	87.88	83.09	84.81	87.25	87.18	37.42
Case IV–(4)	89.39	85.23	83.01	86.40	86.77	35.09
Case IV–(5)	87.50	85.97	84.81	85.55	86.50	35.61
Case IV–(6)	86.36	84.59	84.81	84.7	86.36	42.12
Case IV–(7)	84.38	85.70	84.81	83.85	86.23	36.14
Case IV–(8)	84.85	84.29	81.01	83.00	85.14	36.36
Case IV–(9)	81.82	83.30	80.42	82.55	85.00	42.61
Case IV–(10)	84.85	83.15	80.18	82.24	84.82	38.93
Case IV–(11)	81.82	84.08	77.22	82.00	84.64	38.93
Case IV–(12)	80.30	82.66	79.22	81.85	84.50	44.28
Case IV–(13)	81.82	83.24	78.97	81.55	84.23	45.20
Case IV–(14)	78.79	81.00	78.97	81.00	84.07	45.96
Case IV–(15)	70.79	73.01	77.22	79.53	83.52	46.20

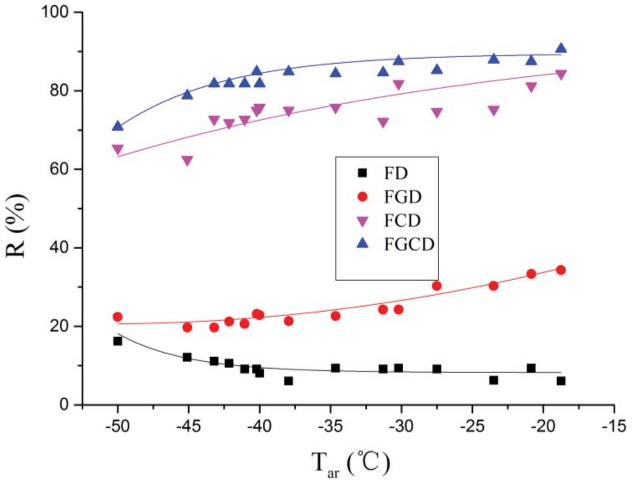

Fig. 7. Relationships between R and T_{ar} for FD, FGD, FCD and FGCD processes.

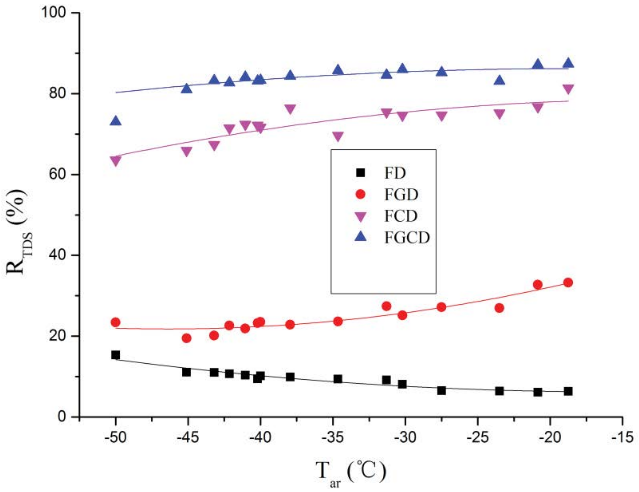

Fig. 8. Relationships between R_{TDS} and T_{ar} for FD, FGD, FCD and FGCD processes.

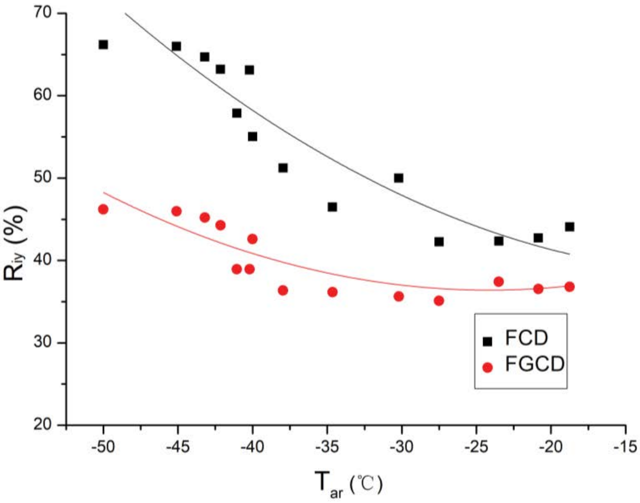

Fig. 9. Relationships between R_{iy} and T_{ar} for FCD and FGCD processes.

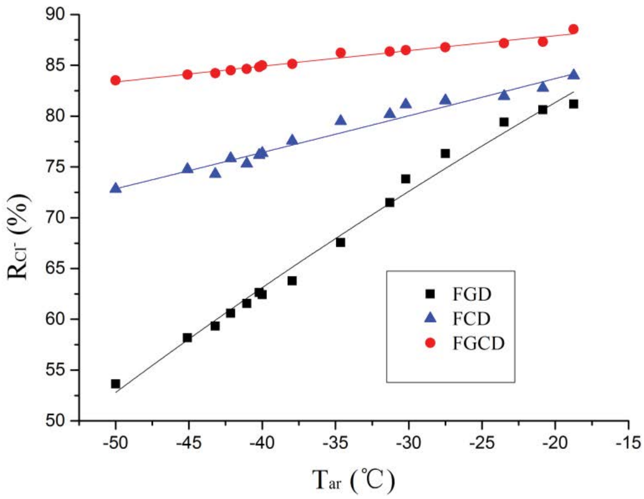

Fig. 10. Relationships between R_{Cl^-} and T_{ar} for FGD, FCD and FGCD processes.

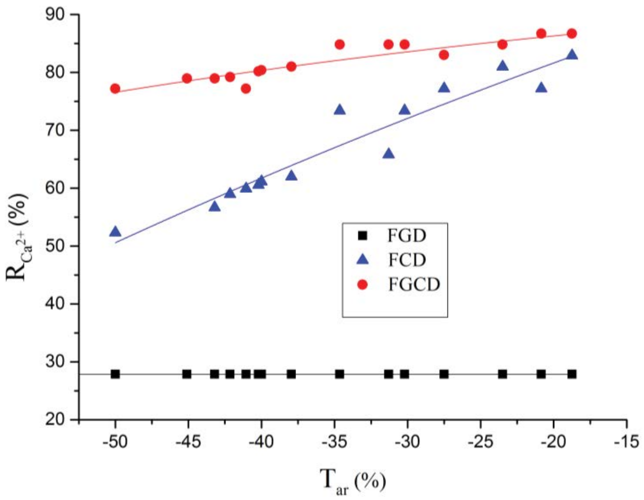

Fig. 11. Relationships between $R_{Ca^{2+}}$ and T_{ar} for FGD, FCD and FGCD processes.

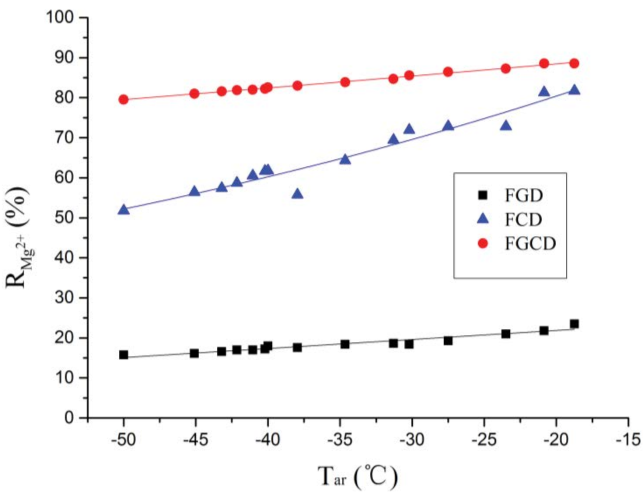

Fig. 12. Relationships between $R_{Mg^{2+}}$ and T_{ar} for FGD, FCD and FGCD processes.

for the seawater ice flakes produced under the same T_{ar}, the effects of freezing-based combined desalination methods tested in present research can be listed in the descending order as FGCD, FCD and FGD. The relationships between the removal efficiencies of the measured ions and T_{ar} shown in Figs. 10–12 are in accordance with this result. The changes of ion removal efficiencies also support the conclusion that increasing refrigerant temperature would benefit to ameliorate desalination effect, especially for separating Ca^{2+}, Mg^{2+} in FCD and FGCD processes and for separating Cl^{-} in FGD, FCD and FGCD processes.

4. Conclusion

In this paper, the patented experimental platform, which could use LNG cold energy, was employed to study the effects of the arithmetic average temperature of the intermediate refrigerant T_{ar} on the freezing-based combined desalination processes. Based on freezing process, gravity-induced desalination with 10% brine mass drainage proportion, centrifugal desalination were selected to form FGD, FCD and FGCD processes to deal with ice flakes. The main conclusions are obtained as follows:

- A special phenomenon was found for seawater ice flakes that the freezing salt and TDS removal efficiencies did not increase with T_{ar} and on the contrary, higher ice purity could be got under lower T_{ar}. It seems can be explained from the special freezing process and the construction characteristics of the ice flakes that loose texture and smaller size of the ice flakes produced under high T_{ar} could contain more raw seawater that was sprayed on them before they were separated from the ice maker. But further study on micro-structure of ice flakes should be conducted to prove this hypothec. Besides that, experimental results demonstrated again that merely artificial fast and continuous freezing process cannot produce high purity ice and further treatments are necessary to decrease the salinity of the ice flakes.
- After FGD, FCD and FGCD process, ice flakes samples produced under higher T_{ar} arrived higher purity with higher removal efficiencies of salty, TDS and ions. For the seawater ice flakes produced under the same T_{ar}, the effects of freezing-based combined desalination methods tested in this research can be listed in the descending order as FGCD, FCD and FGD.
- Centrifugal process is more efficient than the gravity-induced process of 10% brine drainage proportion, but additional gravity-induced process is helpful for improving the desalination effect of centrifugal process. That is why FGCD is the most efficient combined desalination process in this study. But it has to take the cost that the ice yield rate decreases at the same time.

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Symbols

T_{ar} — Arithmetic average temperature of the intermediate refrigerant, °C
R — Salt removal efficiency, %
C_0 — Salinity of the raw seawater (%), 3.3% as a constant in this research
C_i — Salinity of the ice product, %
R_{ion} — Ions or TDS removal efficiency, %
C_{0ion} — Mass concentration of the measured ions or TDS in raw sea water, mg/L
C_{ion} — Mass concentration of the measured ions or TDS in ice product, mg/L
M_{ip} — Mass of ice product, g
M_0 — Initial mass of seawater ice sample (g), 500 g as a constant in this research
R_{iy} — Ice yield rate, %

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