

REMOVAL OF FLUORIDE FROM DRINKING WATER BY FREEZING TECHNOLOGY

Sara Sadat Hosseini,^{a,b} Amir Hossein Mahvi ^{a,c,*}
Tehran, Iran

ABSTRACT: Many countries in the world are experiencing a severe water crisis with 1.1 billion people living without access to clean and safe drinking water. Fluoride is ubiquitous in nature and an excessive intake of fluoride via the drinking water results in significant health effects, particularly with exposure *in utero* and in early childhood. Most of the conventional methods for the removal of fluoride from drinking water in water treatment plants require high initial capital and ongoing maintenance costs. The aim of the present study was to investigate the freezing and melting process as an innovative, convenient, and low-cost technology to produce healthy drinking water for people living in areas with high drinking water fluoride concentrations. We found that the freezing and melting process had a proximate fluoride removal performance of 60% when the freezing of water with a high concentration of fluoride proceeded until 10–15% of the initial water volume was frozen. Nevertheless, in order to achieve drinking water with less than 1.5 mg/L of fluoride, from water with a high initial fluoride content, one or two re-freezing steps of the melted ice were required. In the end, it was proven that this method is able to produce enough healthy drinking water for children for households. This process could be used as a cost-effective large scale technology in the community or as a small scale method at home with a minimal capital expenditure and no need for advanced knowledge requirements. With application of liquid natural gas (LNG), solar energy, and wind power as clean and cheap energy sources, it will be possible to build large scale freezing systems for the removal of fluoride from drinking water for communities.

Keywords: Crystallization; Drinking water; Fluoride; Freezing process.

INTRODUCTION

We are currently experiencing a severe water crisis in many places around the world with 1.1 billion people living without access to clean drinking water. Water is scarce in numerous regions of the world and many people suffer from perpetual water shortages. According to estimates, without significant global policy changes, by the year 2030, the world will only have 60% of the water it needs.^{1, 2}

Fluoride, the ion of the element fluorine, is ubiquitous in nature. From abundant deposits in the lithosphere, fluoride has become widely distributed over the earth's surface and atmosphere, largely as the result of natural processes, such as erosion, hydraulic leaching, and volcanic activities.³ Fluoride is one of the very few chemicals that has been shown to cause significant effects in people through drinking-water.⁴ Fluoride may be present in groundwater aquifers.⁵ Fluoride levels exceeding 1.5 mg/L in the drinking water may lead to various diseases including dental fluorosis, skeletal fluorosis, and non-skeletal fluorosis.^{6,7} The current WHO guideline value for the upper limit of the drinking water fluoride concentration of 1.5 mg/L has been recommended to protect children from the adverse effects of fluoride exposure. These range, as the level and duration of the exposure increases, from mild dental

^aDepartment of Environmental Health Engineering, School of Public Health, Tehran University of Medical Sciences, Tehran, Iran; ^bCenter for Water Quality Research, Institute for Environmental Research, Tehran University of Medical Science, Tehran, Iran; ^cCenter for Solid Waste Research, Institute for Environmental Research, Tehran University of Medical Science, Tehran, Iran. *For correspondence: Amir Hossein Mahvi, Center for Solid Waste Research, Institute for Environmental Research, Tehran University of Medical Science, Tehran, Iran. Telephone: +989123211827; E-mail: ahmahvi@yahoo.com

fluorosis to crippling skeletal fluorosis.^{4,8} An inverse relationship has also been found between IQ and the urinary fluoride level.⁹ Crippling skeletal fluorosis is a significant cause of morbidity in a number of regions of the world.⁴ The main source of these effects is from fluoride-contaminated drinking water, which leads to 65% of endemic fluorosis diseases.⁶ Fluoride is ubiquitous in nature and the excessive intake of fluoride via drinking water may lead to significant health effects in children including impaired IQ and osteosarcoma. Many studies have considered the health risks of exposure to high levels of fluoride and the various methods for treating fluoride-contaminated water.¹⁰⁻²⁸ The removal of fluoride from high fluoride drinking water, so that it is safe for use, especially by children, can be accomplished by various processes including chemical precipitation, adsorption onto activated alumina, adsorption onto modified *Azolla filiculoides*, membrane process, and ion exchange.^{24,29} Among these processes, membrane and ion exchange processes are not very common due to their high installation and maintenance costs.²² Some of these methods could also be used for the simultaneous removal from aqueous solutions of multiple pollutants.³⁰⁻³¹

However, especially in rural and slum areas, most of these treatment methods and requesting expert technicians to provide a public source of potable water in communities are too expensive. Moreover, due to the lack of facilities available in these areas, most of these methods are not applicable. In these cases, in order to improve the quality of potable water, a low-cost intervention with a very simple household water purification process is required.³²⁻³⁵

The freezing of an aqueous solution and the melting of ice crystals are physical processes involving the matter of the differences in the freezing points of pure and concentrated water. There are two fundamental freezing mechanisms known as suspension crystallization and progressive freezing. Because of the small size of crystals, the inclusion of elements and compounds in the crystal lattice is different. Usually, it is much easier and simpler to separate the ice crystals from the main solution in the progressive freezing method than with the suspension crystallization method.

Any saline water desalination by the freezing mechanism is based upon the fact that the ice crystals are essentially produced from pure water as a result of lowering the temperature of the saline water solution to below its freezing point by removing heat from the water body. The impurities are excluded naturally from the ice crystals as the crystals grow. In the freezing process, the water phase changes from liquid to solid as ice, which is in contrast to the situation with thermal desalination where water changes from the liquid phase to vapor. In theory, to freeze 1 kg of water, 80 kcal of energy is required, which is in contrast to the energy requirement for the evaporation of 1 kg of water, when approximately 600 kcal of energy is consumed. Therefore, the total power to run the process is much lower for the freezing process. Since, this freezing process is takes place below 0°C (-3°C) and no heating is applied, no scale formation and corrosion occur.²¹

In theory, the freezing process of liquids for the separation of impurities has a lower energy consumption in comparison to other thermal methods.³⁶ It is obvious that the energy consumption of the sea water desalination processes, such as evaporative and reverse osmosis, is much higher than that required for the freezing process. Currently,

with energy costs being high, renewable energy sources, such as wind and solar energy, are receiving consideration and becoming more popular as power sources for saline water desalination plants.³⁷

Although a lower energy is utilized in the freezing process compared to other processes, the application of this process by refrigerators usually utilizes much more electricity, which is known as a high class of energy in contrast to heating energy. If cold energy could be obtained at a lower price, saline water desalination plants would be much more interesting and cost effective. A few researchers have shown the consumption of liquefied natural gas as an energy source can be much more beneficial and cost effective.³⁸

The most important advantages of the freezing method for saline water desalination, compared to the other thermal technologies, are the lower theoretical energy requirement and the lower operating temperature. In addition, the freezing and melting of ice crystals process faces much less scaling, fouling, and corrosion difficulties. Also, this process deals with the application of much cheaper materials and equipment. It should be noted that in the freezing process, there is almost no need for pretreatment.³⁹ In the case of small scale sea water desalination, such as conducting the work in a freezer at home, no expert technician would be required to produce the low fluoride water.

The aim of the present study was to investigate the performance of the freezing and melting process as a simple, convenient, and low-cost defluoridation method to produce a healthy source of water for poor people living in rural and slum areas. The range of the fluoride concentrations used in this study was 1.5–10 mg/L and included five different concentrations (1.5, 2, 3, 5, and 10 mg/L) which were chosen based on the reported levels of this pollutant in natural groundwater around the world.^{4,20}

In a small scale freezing process one uses a refrigerator and the same technology can be extended for use on a larger scale. Experts and engineers are currently working to scale up the freezing technology to set up saline water desalination plants for large communities. The flow chart of the freezing and melting process to produce fresh water from saline sea water is presented in Figure 1.

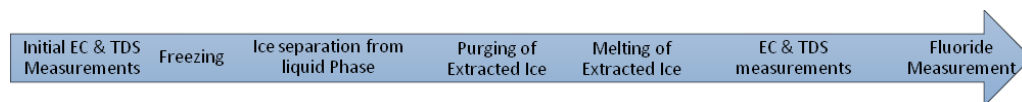


Figure 1. The process flow chart

MATERIALS AND METHODS

1.1. REAGENTS:

a. Standard fluoride solution: Standard stock solutions of 100 mg/L fluoride were prepared by dissolving 0.221 g of formerly dried (in an oven for 24 hours at 105°C) analytical grade sodium fluoride (Merck, Germany) in double-distilled water. The fluoride solutions for the batch experiments were prepared from fresh stock fluoride solution by appropriate dilution. A series of the samples and standard solutions were prepared by pipetting suitable volumes of stock fluoride solution by means of a Scienceware[®] Roxy M[™] Repeating Pipettor. All the experiments were examined in a 250 mL Erlenmeyer Flask.

b. SPADNS solution: The SPADNS (2-(4-sulphophenylazo)-1,8-dihydroxy-3,6-naphthalenedisulphonic acid) solution was prepared in accordance with the 4500-F-D method of *Standard methods for the examination of water and wastewater*⁴⁰ using analytical grade chemicals (Merck, Germany) and used while fresh.

c. Zirconyl-acid reagent: The zirconyl-acid reagent was prepared by dissolving 133 mg zirconyl chloride octahydrate, $ZrOCl_2 \cdot 8H_2O$, in about 25 mL of distilled water and the addition of 350 mL concentrated HCl. This solution diluted to 500 mL with distilled water. This reagent was prepared by means of analytical grade chemicals (Merck, Germany) and used while fresh.

d. Acid zirconyl-SPADNS reagent: The acid zirconyl-SPADNS reagent was prepared with equal volumes of SPADNS solution and zirconyl-acid reagent. This reagent was prepared by analytical grade chemicals (Merck, Germany) and used while fresh.

e. Reference solution: The reference solution was prepared in accordance with the 4500-F-D method of the *Standard methods for the examination of water and wastewater*⁴⁰ using analytical grade chemicals (Merck, Germany) and used while fresh.

f. Sodium arsenite solution: The sodium arsenite solution was prepared by the dissolution of 5.0 g $NaAsO_2$ which was successively diluted to 1 L with double distilled water.

1.2. EC AND TDS PRE-MEASUREMENT: The electrical-conductivity and the total dissolved solids were measured for every sample at the beginning in compliance with the American Society for Testing and Materials Standard Methods⁴¹ and the *Standard Methods for the Examination of Water and Wastewater*.⁴⁰

1.3. CRYSTALLIZATION: Equal amounts of the samples were allocated among identical flasks which were subsequently stored in a Pars-PAMCO-FRZNF170 freezer with an energy consumption of $0.1 \text{ kW}\cdot\text{hr}^{-1}$ to be crystallized by indirect freezing. Frequent observations took place to determine the amounts of ice produced. Samples with different amounts of ice production were obtained from the freezer and the layers and crystals of ice were extracted from the solution. These ice layers and crystals were coated with a thin layer of residual fluoride-polluted water which were purged by means of double-distilled water, prior to letting them melt at room temperature in an airtight jar to mitigate dry deposition and the adsorption of gases and particles.

1.4. EC AND TDS RE-MEASUREMENT: The residual solutions and the melted ice water were examined to investigate their electrical-conductivity and total dissolved solids using methods ASTM D1125 - 14 and 2540 C in compliance with the American Society for Testing and Materials Standard Methods,⁴¹ and the *Standard methods for the examination of water and wastewater*.⁴⁰

1.5. MEASUREMENT OF FLUORIDE CONCENTRATION: The fluoride ion concentration in both the melted ice and the residual solution was analyzed using the US EPA-method-13A method (SPADNS Zirconium Lake Method)¹⁷ which is in compliance with method 4500-F-D of the *Standard methods for the examination of water and wastewater*⁴⁰ by means of a DR 5000™ UV-Vis Spectrophotometer in accordance

with the manual for this device.⁴² All the measurements were performed three times to mitigate measurement errors.

The fluoride removal, as well as the TDS and EC reduction performance of the freezing-melting process, were calculated by means of the following equation:

$$\text{Removal \%} = \frac{P_1 - P_2}{P_1} \times 100$$

Where:

P_1 : The initial value of the parameter

P_2 : The ultimate value of the parameter

Removal %: Removal percentage as a performance indicator.

RESULTS AND DISCUSSION

3.1. REDUCTION OF FLUORIDE CONCENTRATION BY MEANS OF FREEZING AND MELTING PROCESS:

The results show how the freezing of various proportions of a fluoride-containing solution can separate purified ice crystals from the fluoride-containing water and that when this ice melts it turns to water with a lower fluoride concentration compared to the initial fluoride concentration of the solution.

The initial fluoride concentrations in the sample waters were 1.5, 2, 3, 5, and 10 mg/L. Figures 2 to 8 illustrate how various proportional volumes of freezing, from 10% up to 50% of the initial solution volume, can affect the degree of fluoride removal.

It was found that the freezing and melting process had a proximate fluoride removal performance of 60% when the freezing occurred for 10% up to 15% of the total volume (Figures 2–4).

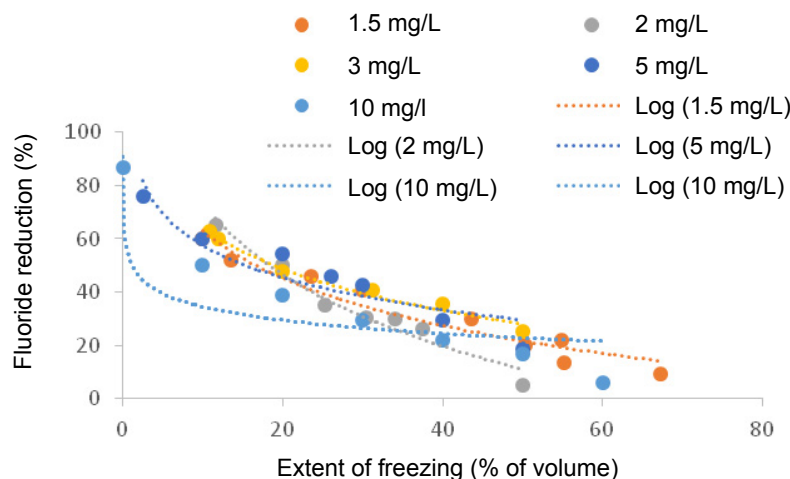


Figure 2. The effect of the extent of freezing, as a percentage of the volume, on the fluoride removal rate. The initial fluoride concentrations were 1.5, 2, 3, 5, and 10 mg/L.

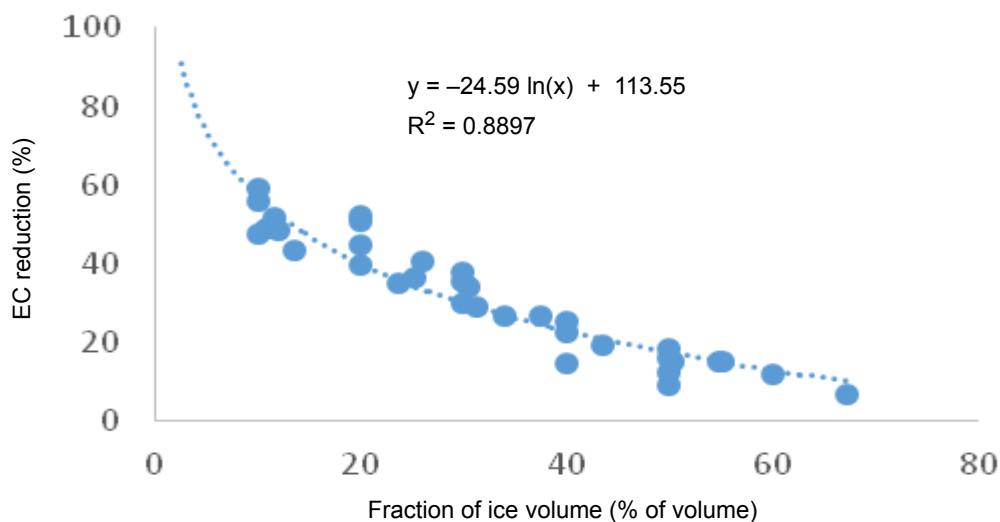


Figure 3. The effect of the proportional volumes of freezing on electrical conductivity (EC) reduction.

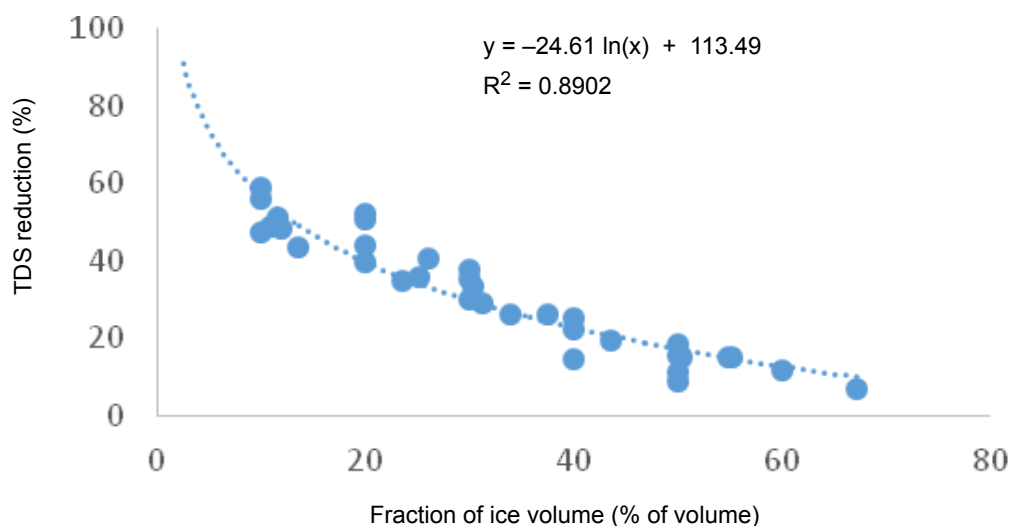


Figure 4. The effect of the proportional volumes of freezing on the total dissolved solids (TDS) reduction.

This amount of reduction is enough to create an acceptable source of potable water, with a fluoride concentration of less than 1.5 mg/L, for an initial fluoride concentration of less than 3 mg/L with a single freezing. Nevertheless, in order to achieve water, with less than 2 mg/L fluoride, from a more concentrated fluoride solution, with 5 and 10 mg/L, one or two re-freezings of the melted ice was required. It was also found that fluoride always tends to be more concentrated in the residual solution compared to the concentration in the solid phase.

Theoretically, when salt water starts freezing, ice crystals are essentially salt-free. This phenomenon is due to the earlier crystallization of pure water prior to the crystallization of dissolved salts. However, as the freezing extends, the crystallization of the dissolved salts will also take place since the solubility of the salts reduces as a result of the thermodynamic limitations of dissolution. Therefore, when using freezing as a treatment method for the desalination of water; it is highly important to control the time and the temperature in order to retard the crystallization of the dissolved salts and to enhance the pure water production. In fact, ice production is a function of the temperature and the freezing time. When the freezing time is prolonged and/or the freezing temperature is reduced, more ice will be formed. However, this rise in the amount of ice formed is synchronous with a fall in quality.

Fluoride removal, as a percentage of the initial concentration, was plotted against the proportional volumes of freezing. Different percentage reductions in the fluoride concentrations were plotted against their proportional volumes of freezing for all the initial concentrations of fluoride. There was a negative coefficient for the logarithmic relationship between fluoride removal % and the proportional volumes of freezing. It was inferred from this fact that the previous paragraph's statement, that a rise in the amount of ice formed is synchronous with a fall in quality, was proven (Figure 2).

Electroconductivity and total dissolved solids were also plotted against the proportional volumes of freezing and the resulted charts for these two parameters were similar to each other. The reduction of EC and TDS because of water purification due to freezing had a logarithmic relationship with the proportional volumes of freezing (Figures 3 and 4).

Figure 2 describes the effect of the volume of liquid frozen on the degree of fluoride removal. For an initial fluoride concentration of 1.5 mg/L, when the amount of the frozen volume was less than 30% of the total raw water volume, fluoride concentrations of less than 1 mg/L could be achieved. For an initial fluoride concentration of 2 mg/L, when the amount of freezing was less than 20% of the total raw water volume, acceptable standard levels of fluoride, not exceeding the WHO upper limit of 1.5 mg/L, could be achieved. For an initial fluoride concentration of 3 mg/L, when the amount of freezing was about 10%, the fluoride concentration was reduced from the initial value by more than 62% and therefore the guideline value for fluoride of 1.5 mg/L could be achieved. For an initial fluoride concentration of 5 mg/L, when the amount of freezing was less than 2.5%, standard levels of fluoride could be achieved. It worth mentioning that for this solution the freezing-melting process was done in two stages. The freezing amount of the first step was 10% and water from the melting of the ice in this stage was used for the next stage. The freezing amount of the second stage was 25%. For an initial fluoride concentration of 10 mg/L, when the amount of freezing is less than 0.25%, standard levels of fluoride could be achieved. For this solution, the freezing-melting process was triple stage. The freezing amount of the first step and second step was 10% and water from the melting of the first step was used for the second stage and water from melting of the second step was used for the third stage. The freezing amount of the third stage was 25% of the last volume.

Figure 3 describes the effect of the freezing amount on the electrical conductivity reduction. The relationships between EC reduction and the freezing amount were comparable to the relationships between the fluoride removal rates and the freezing amounts.

Figure 4 describes the effect of the freezing amount on the total dissolved solids concentration reduction. The relationships between the TDS reduction and the freezing amount were comparable to the relationships between the fluoride removal rates and the freezing amounts.

Figure 5 describes the additive effects of every stage of freezing on the fluoride concentration compared to an initial fluoride concentration of 10 mg/L.

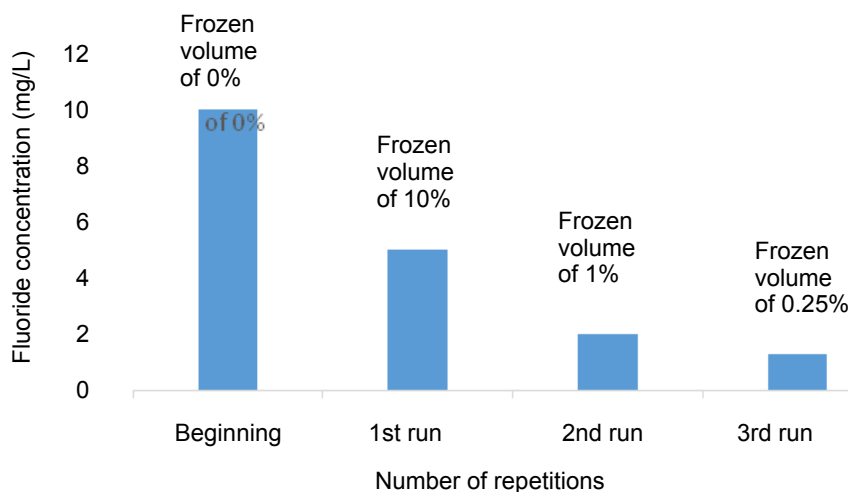


Figure 5. The effect of each treatment stage on the fluoride content with an initial concentration of 10 mg/L (triple stage).

Figure 6 describes the additive effects of every stage of freezing on the fluoride concentration compared with an initial fluoride concentration of 5 mg/L.

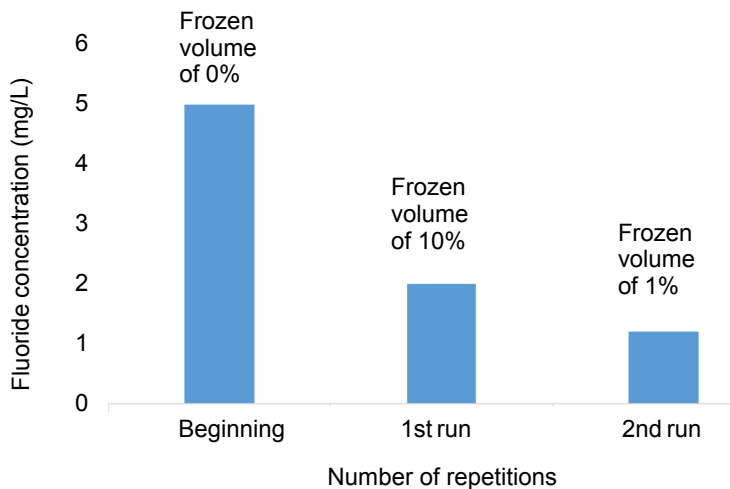


Figure 6. The effect of each treatment stage on the fluoride content with an initial concentration of 5 mg/L (double stage).

Figure 7 compares the fluoride concentration of the residual solution and the solid phase for the different fluoride concentrations following by the first stage of the freezing-melting process.

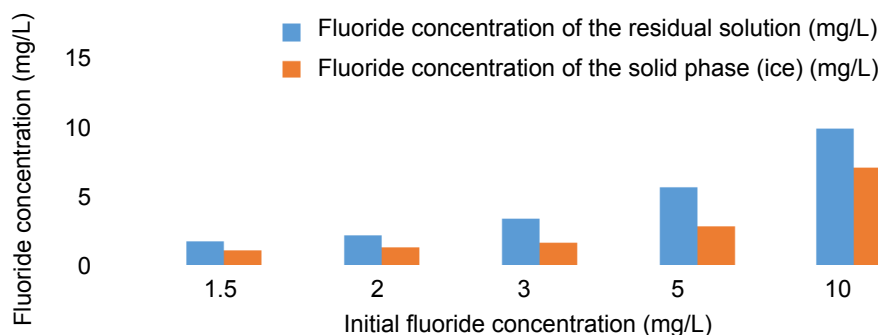


Figure 7. Comparison of the fluoride concentration in the solid phase (ice) and the residual solution.

Different normality tests (Anderson-Darling, Lilliefors, Jarque-Bera, and Shapiro-Wilk) were used to determine the appropriate correlation method. Since, the data distribution was normal, the Pearson correlation method was preferred. The two-sample Kolmogorov-Smirnov test was also used to make an illustrative comparison between the distributions of the variables. Identical distribution trends were inferred as a result of the correspondence between the variables. The following charts and tables exhibit the results of these statistical analyses. All the parameters of the normality tests were significant and the normality hypotheses was assumed to be true. Probability-Probability (P-P) plots were also used to demonstrate these findings by plotting the theoretical cumulative distribution against the empirical cumulative distribution. A P-P plot compares the empirical cumulative distribution function of a variable with a specified theoretical cumulative distribution function. Table 1 presents the p values for the different normality tests. Since the p values are greater than $\alpha=0.05$, the null hypothesis, that the sample was derived from a population with a normal distribution, could not be rejected.

Table 1. Summary of normality test results (p value), (EC = electrical conductivity, TDS = total dissolved solids)

Variable	Normality test			
	Shapiro-Wilk (p value)	Anderson-Darling (p value)	Lilliefors (p value)	Jarque-Bera (p value)
Fluoride reduction (%)	0.440	0.701	0.614	0.511
EC reduction (%)	0.129	0.162	0.320	0.336
TDS reduction (%)	0.133	0.170	0.348	0.337

Figure 8 compares the fluoride removal distribution with an absolute normal distribution by plotting their quantiles against each other. As the points in the fluoride removal Q–Q plot lie approximately on the absolute normality line $y=x$, this distribution has an approximately normal distribution.

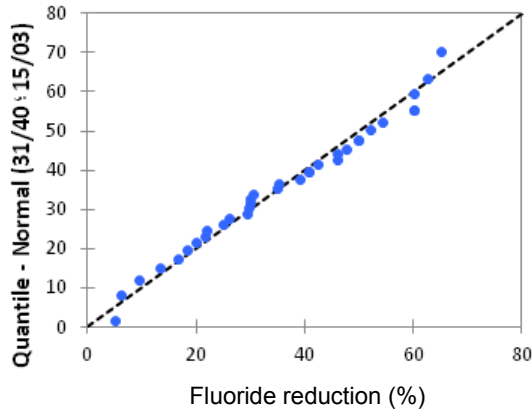


Figure 8. Normal Q-Q plot for fluoride reduction.

Figure 9 compares the EC reduction distribution with an absolute normal distribution by plotting their quantiles against each other. As the points in the EC reduction Q–Q plot lie approximately on the absolute normality line $y=x$, this distribution has an approximately normal distribution.

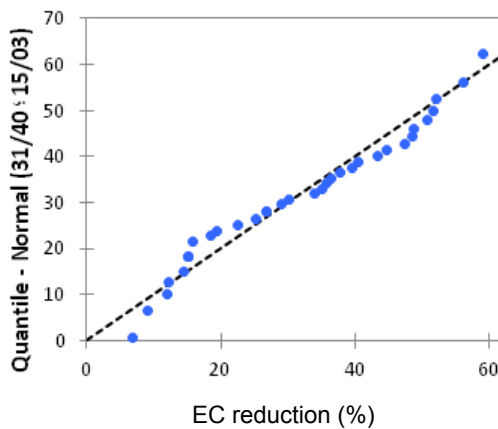


Figure 9. Normal Q-Q plot for total dissolved solids (TDS) reduction.

Figure 10 compares the TDS reduction distribution with an absolute normal distribution by plotting their quantiles against each other. As the points in the TDS reduction Q–Q plot lie approximately on the absolute normality line $y= x$, this distribution has an approximately normal distribution.

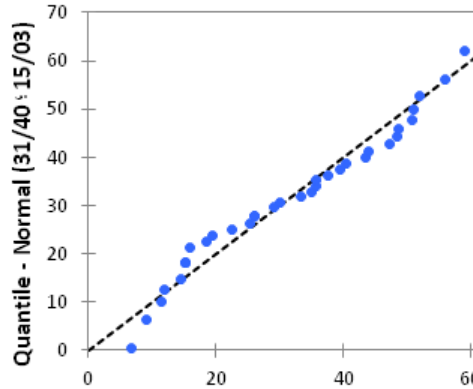


Figure 10. Normal Q-Q plot for total dissolved solids (TDS) reduction.

Pearson’s correlation method was used to analyze the inter-relationships of the variables. These values can vary between -1 and $+1$ with respect to the direction of the correlation (reverse or direct) while a value of “0” reveals that there was no linear correlation. Table 2 presents the Pearson correlation coefficients for fluoride removal, EC reduction, and TDS reduction. Since these values are around $+1$, it could be considered that there were some strong inter-relationships between these variables.

Table 2. Correlation matrix (based on Pearson's method), (EC = electrical conductivity, TDS = total dissolved solids)

Correlation matrix (based on Pearson's Method)			
	Fluoride reduction (%)	EC reduction (%)	TDS reduction (%)
Fluoride reduction (%)	1	0.896	0.898
EC reduction (%)	0.896	1	1.000
TDS reduction (%)	0.898	1.000	1

Table 3 presents the coefficients of determination (R^2) for the variables of fluoride removal, EC reduction, and TDS reduction for the Pearson’s correlation test. Since these values are around 1, it can be considered that the Pearson correlation test results have a high probability of trueness or goodness of fit.

Table 3. Coefficient of determination (R^2) matrix for the different variables of the Pearson's correlation test: fluoride reduction (%), electrical conductivity (EC) reduction (%), and total dissolved solids (TDS) reduction (%)

Coefficient of determination (R^2) matrix			
	Fluoride reduction (%)	EC reduction (%)	TDS reduction (%)
Fluoride reduction (%)	1	0.802	0.806
EC reduction (%)	0.802	1	1.000
TDS reduction (%)	0.806	1.000	1

Table 4 the presents the p values for the variables of fluoride removal, EC reduction, and TDS reduction for the Pearson's correlation test. Since these values are around 0, it can be considered that Pearson correlation test results have a high probability of trueness or goodness of fit.

Table 4. p value matrix for the different variables of the Pearson's correlation test: fluoride reduction (%), electrical conductivity (EC) reduction (%), and total dissolved solids (TDS) reduction (%)

p value matrix			
	Fluoride reduction (%)	EC reduction (%)	TDS reduction (%)
Fluoride reduction (%)	0	0.000	0.000
EC reduction (%)	<0.0001	0	<0.0001
TDS reduction (%)	<0.0001	<0.0001	0

Table 5 presents the results of applying the two-sample Kolmogorov-Smirnov test, which is used for investigation of relationships between different variables, to the variables of TDS reduction and EC reduction. With regard to the small but contrasting degrees of freedom (D) values it was found that there were strong relationships between the variables, but that these strengths were not identical.

Table 5. Two-sample Kolmogorov-Smirnov test for total dissolved solids (TDS) and electrical conductivity (EC) reduction

D	0.059
Asymptomatic p value	1.000
Alpha	0.05

Two-sample comparisons of distributions were also used in order to illustrate these relationships. Table 6 presents the degrees of freedom (D), asymptotic p value, and the predefined significance level (alpha) for the two-sample Kolmogorov-Smirnov test between the variables of EC reduction and fluoride. This high p value, as well as the small number of degrees of freedom, show that

Table 6. Two-sample Kolmogorov-Smirnov test for fluoride and electrical conductivity (EC) reduction

D	0.147
Asymptomatic p value	0.856
Alpha	0.05

Table 7 presents the degrees of freedom (D), asymptotic p value, and the predefined significance level (alpha) for the two-sample Kolmogorov-Smirnov test between TDS and fluoride. This high p value, as well as the small number of degrees of freedom, show that there were strong relationships between the variables.

Table 7. Two-sample Kolmogorov-Smirnov test for fluoride and total dissolved solids (TDS) reduction

D	0.147
Asymptomatic p value	0.856
Alpha	0.05

The results from this study are in agreement with similar studies reported in the desalination literature which show an inverse relationship between the proportional volume of freezing and the efficiency of the desalination process.⁴³⁻⁴⁶ This process does not require any experimental apparatus and it does not add any other substances to the water, unlike the ion-exchange and the electrocoagulation methods. Moreover, there was no need for electrode consumption or resin capacity reduction. The power consumption for this method was about $1 \text{ kW}\cdot\text{h}^{-1}\cdot\text{L}^{-1}$, for a freezing time of 1 hour and a freezer loading of 4 liters of an aqueous solution with an initial fluoride concentration of 5 mg/L and a 2.5% proportional volume of freezing. For comparison, the power consumption for an electrocoagulation process with aluminum electrodes is about $0.058 \text{ kW}\cdot\text{h}^{-1}\cdot\text{L}^{-1}$ for the treatment of a similar solution.³¹ Activated alumina is a suitable adsorbent of fluoride, but it requires a set of equipment and basins for the process to proceed. This need for chemicals for an electrocoagulation process is a disadvantage in comparison to the freezing process.³ The operating capacity of alumina is 2.94 g fluoride/kg alumina. This means that 588 liters of the aforementioned solution could be treated with a kilogram of alumina. All these methods have their own pros and cons and the best available technology may vary based on practical and local experience.

One of the most important challenges for this process is to maintain cheap and available sources of energy. With the help of sustainable and renewable sources of energy, such as solar energy or wind power in many areas in the world, technology promises much for many communities. Similarly, the use of LNG as a clean and accessible source of energy also has considerable potential, especially in the Middle East where the need for desalination of saline sea water is very considerable. With the use of LNG as a cheap source of energy, the operating and maintenance costs of this freezing technology could be reduced.

CONCLUSIONS

The present study aimed to evaluate the performance of the freezing and melting process for the production of high-quality water. As it is shown by the results, this process has a good functionality for the removal of fluoride from water. In spite of only small amounts of water being produced in some settings, as with its use as a home water treatment method, this process is of interest because of the low capital expenditure involved, as nowadays there is a refrigerator in almost every home, and the simplicity of the process makes it really easy for it to be performed by a non-expert person living in a developing country in order to produce a convenient supply of healthy potable water, especially for infants. The results from this study could also be used by ice companies for the optimization their manufacturing process in order to reduce the fluoride content of their products. This process has been used for water desalination for many years, but there were not enough feasibility studies regarding its use for the removal of different pollutants from water. Since this process does not produce any by-products (except for the production of water with a higher concentration of the particular pollutant which could be used for the irrigation of tolerant plants, etc.), does not produce any solid wastes (such as ion-exchange resins, etc.), and has an energy consumption which is lower than required for some other methods, it can be considered as a cleaner and more eco-friendly water treatment alternative, both industrially and domestically. One of the major limitations of this

process (except for automatic industrial units) is its demand for appropriate timing and an awareness of the ice and water proportions in order to produce a healthy source of water cost effectively. Moreover, it is time consuming. One of the most important challenges for this process is to maintain cheap and available sources of energy. With the help of sustainable and renewable sources of energy, such as solar energy or wind power which are now available in many areas in the world, this technology has much to offer for many communities for producing safe drinking water. Similarly, the use of Liquid Natural Gas (LNG) as a clean and relatively inexpensive source of energy would also allow the operating and maintenance costs of this freezing technology to be reduced.

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