



Application of freeze desalination for chromium (VI) removal from water



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HIGHLIGHTS

- High Cr(VI) removal efficiency with low water rejection can be obtained by freezing
- Freeze desalination is useful for Cr removal in a wide range of Cr concentrations
- Removal efficiency decreases with increasing salinity

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ABSTRACT

Chromium (VI) is a highly toxic metal ion found in the priority list of pollutants, which is also often abundant in areas having low access to high-tech solutions for chromium (Cr) removal. This study was initiated to investigate the capacity of home-use refrigerators to generate fresh water free from Cr(VI) by melting ice produced from Cr(VI) polluted water. Simulated tap water samples as well as deionized water to which different concentrations of Cr(VI) were added were frozen in a closed freezer unit. The effects of initial concentration, time of ice nucleation, fraction of ice volume, and influence of co-occurring ions were evaluated in relation to the quality of the produced ice. The physicochemical characteristics of the produced ice cubes were also evaluated. A high total water recovery of up to 85% was achieved. Chromium removal ranged from 57.4 to 80% for simulated tap and from 93 to 97% for deionized water spiked with Cr. The energy consumption estimation basing from the refrigerator amounted to 0.076 kWh. Freeze desalination was found to be relatively viable desalination technology in terms of quality of water produced, easiness for safe water production, and amount of energy consumed, especially where high-tech solutions for Cr(VI) removal are not available.

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1. Introduction

Chromium is one of the most common pollutants introduced into natural waters due to the discharge of a variety of industrial wastewaters containing Cr, including wastewater generated by leather tanning, electroplating, textile, and metal finishing industries. Although tanneries represent an important economic field both in developed and in developing countries, many of the compounds used in the tanning process release chromium to the environment and induce negative effects [1,2]. Among the various tanning methods, more than 90% of the leathers tanned globally, contain chromium, with 30–50% of the Cr used in the process leaching into the environment [3,4]. According to the Blacksmith institute report (2007), various drinking water sources in developing countries are highly polluted with Cr(VI) [5]. In Ethiopia, literatures are pinpointing towards the presence of hexavalent

chromium in surface waters such as streams and rivers nearby leather industries. In the water bodies of Ethiopian rift-valley lakes, in six rivers (their inflows), and in effluents from two factories Cr was reported to occur in ranges between 0.104 and 0.121 mg/L [6]. Cr(VI) concentrations have been shown to exceed the Ethiopian drinking water standard (ES261:2001 or CES 58-2013) of 0.05 mg/L and the maximum permissible level set by the WHO [7,8]. An important example is the pollution of surface water in Addis Ababa reported by Alemayehu [9], where chromium is present above background level due to the uncontrolled disposal of wastes, especially tannery wastes, and inadequate waste management strategies.

Chromium exists in several forms, including trivalent and hexavalent states, and the toxicity of those species differs significantly. The occurrence of Cr(III) or Cr(VI) has a significant effect on the transport and fate of chromium in the environment. Chromium (III) is an essential compound for humans and animals and plays an important role as the glucose-tolerance factor in insulin metabolism. On the other hand, Cr(VI) is relatively mobile in the environment and due to its powerful oxidizing nature, it is even evidenced as being mutagenic and

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carcinogenic [10,11]. Considerable efforts, therefore, have been made to treat mostly Cr(VI) containing water/wastewater.

Several desalination technologies exist for the production of fresh water involving either separation processes or thermal processes. Separation processes include electro dialysis (ED) and reverse osmosis (RO). The main drawbacks for membrane separation processes, however, are clogging, adsorption and cake layer formation on the membrane by the pollutants. Thermal processes which involve phase changes, such as membrane distillation and freezing processes, are employed frequently [12–14]. Recently extensive efforts have been focused on reducing the bio-fouling. One such mitigation is the hybrid membrane bioreactor, where membrane technology is integrated with various other treatments such as the conventional activated sludge process and reverse osmosis with plane membrane hybrid techniques [15]. However, such treatments are handicapped by high treatment cost. Treating industrial wastewater by segregation of waste streams is a very important step in tannery pollution prevention although it is not widely applied [16]. Moreover, it is often not feasible to apply high-tech membrane-based solutions at individual household levels in rural areas of developing countries.

Freezing and subsequent removal and melting of the ice is an alternative physical process which can be used for desalting, based on the different freezing points of fresh and salt waters. It has been reported as being effective to remove various organic and inorganic impurities from water/wastewater [17,18]. When freeze concentration is used to purify water or liquid waste, impurities are separated from the ice phase during formation of the ice crystals. Two basic freeze desalination methods are available: suspension and progressive freeze crystallization [19]. In both processes, inclusion of most compounds in the ice crystal lattice is impossible due to the small dimensions of ice crystal lattice. In progressive freeze crystallization, the separation of ice crystals formed from the concentrated mother liquor is much easier than in the conventional suspension crystallization, in which many small ice crystals are formed [12,20]. In theory, freeze desalination has a lower energy requirement compared to other thermal processes [21]. In the current study, we focused on assessing the potential of home-use refrigerators to generate drinking water from Cr-contaminated water at the point of use.

2. Materials and methods

2.1. Reagents

All chemicals were of analytical reagent grade. Standard stock solutions of 1000 mg/L Cr(VI) were prepared by dissolving 2.829 g of potassium dichromate (Riedel-deHaen, Germany) in double-distilled water. Working solutions were prepared by diluting the Cr stock solutions. Series of standard solutions were prepared by pipetting suitable volumes of Cr solution (5 mg/L) using a Thermo Scientific FJ40512 Finnpiptette. Fresh solutions were prepared prior to each experiment. Moreover, simulated tap water containing major ions (Ca^{2+} 58.64 mg/L, Mg^{2+} 29.26 mg/L, Na^+ 92.67 mg/L, K^+ 20 mg/L, HCO_3^- 470 mg/L and SO_4^{2-} 60 mg/L) spiked with 5 mg/L of Cr was prepared and tested.

2.2. Experimental setup

Prior to starting the experiment, smooth plastic containers of 250 mL capacity were selected. These containers are required to avoid inclusion of contaminants in ice crystals due to the roughness of surfaces involved, which may cause serious contamination during ice crystal formation [22]. Deionized water and simulated tap water spiked with Cr(VI) were tested. In all experiments, the concentration of Cr was 5 mg/L except in the experiments focused on studying the impact of concentration variations. In the latter experiments, varying concentrations of Cr(VI) were studied, from 1 to 300 mg/L. Moreover, the removal of Cr(VI) was tested in the presence of variable chloride ion

concentrations (5, 10, 250 and 3550 mg/L). To cast off the concentrated chromium, a plastic tube of ca. 20 mm diameter was inserted with its upside down in each plastic beaker (Fig. 1). After each studied time interval, i.e. after different partial freezing steps, three beakers were taken from the freezer and ice crystals were separated from the remaining water by rejecting the water. The whole procedure was repeated after different time intervals until only a small volume of concentrated solution remained. To desorb Cr weakly adsorbed to the surface of the ice, the surface was washed three times by rinsing with 20–25 mL deionized water. Subsequently, the ice crystals were melted and analyzed for Cr content using a colorimetric method. Thus, 250 mg chelating agent, 1,5-Diphenylcarbazide (BDH, Poole, England), was dissolved in 50 mL acetone (HIMEDIA, India) and stored in an amber bottle. The pH of sample solutions was adjusted to 2.0 ± 0.5 using 0.5 M sulfuric acid. Then 2 mL of the diphenylcarbazide solution was added to each sample (1 mL) and the mixture was allowed to stand for 10 min. to obtain full color development. Absorbance was measured at 540 nm using a JANWAY 6051 colorimeter. Background correction was performed by analyzing blanks. Calcium, magnesium, potassium and sodium concentrations were measured in the melted ice using an inductively coupled plasma optical emission spectrometer (ICP-OES, Varian VISTA-MPX). Conductivity and solution pH were measured using a microprocessor conductivity meter (WTW LF 537) and a digital pH meter ORION star A211, respectively.

The performance of the freeze separation process was expressed using the following parameter:

$$\% \text{ freeze separation} = \frac{(C_0 - C_s)}{C_0} \times 100 \quad (1)$$

where, C_0 and C_s are the Cr concentrations in the initial feed solution and the remaining ice phase, respectively.

3. Results and discussion

3.1. Deionized water spiked with Cr

Fig. 2 presents the removal of Cr from the deionized water spiked with Cr as function of freezing time and fraction of water transformed into ice. It can be concluded that initially the removal percentage increases up to nearly 97% as the freezing time and volume of ice increases. Afterwards, the entrapment of chromium (VI) in the ice phase increased and the removal percentage started decreasing. Such a trend has been observed also for the removal of organic pollutants [23]. Baker (1967) stated that, if the residual liquid volume is becoming too small, the removal efficiency decreases again, because it is

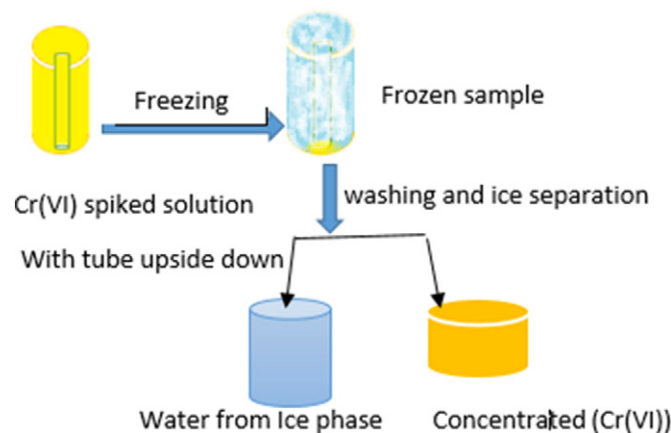


Fig. 1. Overview of the experimental setup with ca. 20 mm diameter plastic tubes being inserted upside down in plastic beakers containing Cr contaminated water.

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