Bio-Filtration as a Solution for the Detrimental Health Effect of Excess Fluoride in Drinking Water

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Introduction

One of the main importance of water filtration is to prevent water-related illnesses and diseases. To this day, various explored methods were used in the remediation of water of different types of contaminants such as flocculation [1,2], coagulation [3], solvent-extraction [4], co-precipitation [5], precipitation [6], ion-exchange [7], photo catalysis [8], adsorption-desorption [9], reverse-osmosis [10], nano-membrane filtration [11]. The adsorption technique is considered one of the attractive and commercial options to eliminate the most pollutant substances whether macro- or micro- organic/inorganic ions from water, due to its simplicity of steps and high activity. Moreover, the purification of water by using adsorption technique has been applied to several matter as absorbent such as agricultural residues and industrial residues, and biomaterials wastes, which are modified and applied in bio-sorption of contaminants from water [7,9-12]. Fluoride is an ionic form of fluorine and can be found in food and numerous sources of drinking water. It can be also purchased as a dietary supplement [12]. Many kinds of toothpastes contain fluoride because these ions serve as an armor against tooth decay [13]. Approximately 80% of the fluoride taken orally is absorbed. Humans retain around 50% of the fluoride they consume, and most of this amount is deposited in teeth and bones (Graphical Abstracts 1 & 2). The remaining 50% is excreted in the urine.
However, young children can retain an especially high percentage of the consumed fluoride, because their bones and teeth absorb more fluoride than those of adults [14]. The principle and mechanism of pollutants removal by adsorption technology from different types of water based on forms layer of condensate pollutants (called adsorbate) which is migrated from aqueous solution to the surface of solid (called adsorbent) as either in the form of liquid-solid interface [11,15]. Excess fluoride is harmful to human health. Groundwater wells worldwide have been reported to contain water with fluoride concentrations exceeding the acceptable level of 1.5 mg/L. This fact was attributed to local ground stones exhibiting high percentages of fluoride in their composition [14]. Excessive fluoride consumption can lead to several health issues, such as skeletal fluorosis, a disorder marked by bone and joint pain as well as joint tenderness. The overconsumption of fluoride during the formative years of tooth enamel can also lead to dental fluorosis, which leads to tooth discoloration, and/or tooth pitting [15]. Many researchers have attempted to develop water filtration strategies to lower the levels of fluoride or metals that contaminate water [16]. Biomaterials have shown great promise in water filtration because of their environmentally favorable properties, high filtering efficiency, and low cost [17].

Charcoal, which may be manufactured from a variety of biomaterials, is one of the most commonly used bio-filters. Water contaminated with heavy metals can be treated using different processes, which include ion exchange, precipitation, reverse osmosis, catalysis, coagulation, and adsorption [18,19]. Heavy metal adsorption is dependent on the nature of the adsorbents, which can be composed of natural or man-made mate-rials including clay [10], sludge [11], industrial waste, activated carbon, and plants [12]. Bone char is an adsorbent made up of 90% calcium phosphate and 10% carbon. It can be created using one of the two methods: treatment with chemicals and physical treatment leading to carbonization of bones [20,21]. Cow bone char is com-monly used to purify water contaminated with heavy metals, as shown in (Table 1). The aim of the current study was to produce a low-cost, effective, and environmentally friendly bio-filter from cow bone through physical activation (carbonization), and subsequently use it to remove fluoride from the polluted water [22]. Furthermore, it was also proposed to enhance this bio-filter by adding nanocomposites. Nanocomposites have very high surface-to-volume ratios, which makes them ideal components for the adsorption process [23,24].

| Material | Observations | Benefits | Ref |
|----------|--------------|----------|-----|
| Fish bone | Testing the effect of charring temperature on fluoride removal capacity | The ability to effectively re-move fluoride without water discoloration at 500°C | [23] |
| Cow bone | Its use at some temperatures can cause the water to become colored | The ability to remove both fluoride and arsenic from water | [23] |
| Cow bone | The bones were dried for 72 hours at 110°C and calcined at 400°C | The ability to filter out tannery effluents from water | [24] |
| Cow bone | The bones were calcined at 700°C and 800°C and washed in HCl solution | Presenting a contrast between theoretical and practical results using Langmuir and Freundlich models to predict fluoride ad-sorption | [25] |

**Experiments**

Cow bones were collected and processed by rinsing them several times with hot water to remove residual muscles and other joint tissues. Then the bones were allowed to dry in the open air. The dried bones were calcined in a furnace for 1 h at 400°C. The resultant charcoal adsorbent was crushed using a gate mortar and stabilized for further testing. It can be stated that the filters were produced with little to no expenses. A stock solution of CaF$_2$ with a 100-ppm concentration of fluoride was prepared to test its adsorption in the cow bone sample. First, 1 g of charcoal was added to the 100 mL solution in a separate beaker, which was then placed in a shaker operated at room temperature and a 251 min$^{-1}$ frequency for 5 h. Next, on the same day, the solution containing cow bone charcoal was filtered from impurities and charcoal leftovers using filter paper to prepare a sample for ionic chromatography (IC) [25]. IC was utilized for determining the concentrations of calcium and
fluoride ions in the prepared \( \text{CaF}_2 \) solution. Ion chromatographs can measure concentrations of important anions such as fluoride, chloride, nitrate, nitrite, and sulfate, as well as major cations such as lithium, sodium, ammonium, potassium, calcium, and magnesium in the parts-per-million (ppm) range. Ion chromatography can also be used to determine the concentrations of organic acid [26].

The surface structure of cow bone charcoal was studied by using a scanning electron microscope (SEM) (Model: JSM-7100F) at 10000 × magnification [27]. In conjunction with SEM, the energy dispersive X-ray (EDX) analysis technique was used to perform the elemental analysis and chemical characterization of the material. EDX utilizes an electron beam that strikes the surface of a conducting sample (placed under SEM) to determine its elemental content [28].

Results and Discussion

The SEM micrographs of the cow bone biochar explained the sample morphology by analyzing the microstructure of the bone powder, as shown in (Figure 1a). The corresponding EDX graph is presented in (Figure 1b). A close look at (Figure 1a) reveals cracks and irregular surfaces, as expected after grinding, where Vickers hardness can be determined (it is planned to be measured in the upcoming study). Furthermore, SEM micrographs showed that the studied samples had several heterogeneous porous layers, which represent a key feature relevant for fluoride adsorption. The pores appeared to be localized symmetrically in certain regions in the grooves on the surface of the bone. The sample composition revealed by EDX results was as follows: \( \text{O}_2 \), 39.6 wt%; \( \text{Ca} \), 33.9 wt%; \( \text{C} \), 12.8 wt%; \( \text{P} \), 12.8 wt%; \( \text{Na} \) and \( \text{Mg} \), 0.9 wt%.

Figure 1: (a) Scanning electron microscope (SEM) micrograph of the cow bone charcoal shows several groups of symmetrically distributed pores. (b) The corresponding energy dispersive X-ray (EDX) graph of the bone constituents.
The surface morphology of the cow bone is particularly distinctive in terms of the hard appearance and the distribution of pores, which are of critical importance in filtering large species from water (Figure 2). Hence, the surface geometry and the source of adsorbent provided different types of active sites onto surface of adsorbent [29-35] (Table 2).

**Figure 2:** (a) SEM micrograph of cow bone charcoal shows symmetrically distributed pores on the surface (red arrows) with diameters in the range of several hundred nanometers, as shown in (b).

| Sample size (μm) | Concentration of fluoride (ppm) | Concentration of fluoride after the experiment (ppm) | Adsorbate % |
|------------------|-------------------------------|--------------------------------------------------|-------------|
| ≤125             | 100                           | 6.4                                              | 93.6        |

**Table 2:** The effect of bio-filtration with cow bone charcoal on fluoride removal.

**Conclusion**

The examined cow bone charcoal filter achieved maximal fluoride adsorption of 93.6%, which can be attributed to its porous nature. The prepared bio-filter has a very high adsorption capacity and is energy-efficient as it works at room temperature and does not require energy consumption. It means that using cow bone charcoal is a cost-effective filtration technique that should be further investigated to optimize the performance of nanocomposites and to set up measures for its widespread manufacturing.

**Supplementary Materials**

N/A.
Author’s Contribution

Alasmari, E, M, carried out experiments and engaged in various article-related tasks. Alsharif, S, H. performed the experimental measure-ments. Khayyat, M, M. designed the experiments and prepared the final version of the manuscript.

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Data Availability Statement

The obtained results have been presented in the cur-rent article, and any further details can be provided upon request made to the cor-reponding author.

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Conflicts of Interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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