ARTICLE IN PRESS

Waste Management xxx (2015) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Sustainable approach for recycling waste lamb and chicken bones for fluoride removal from water followed by reusing fluoride-bearing waste in concrete

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ARTICLE INFO

Article history:
Received 10 April 2015
Revised 11 June 2015
Accepted 30 June 2015
Available online xxxx

Keywords:
Lamb bone
Chicken bone
Fluoride removal
Water treatment
Concrete

ABSTRACT

Sustainable management of waste materials is an attractive approach for modern societies. In this study, recycling of raw waste lamb and chicken bones for defluoridation of water has been estimated. The effects of several experimental parameters including contact time, pH, bone dose, fluoride initial concentration, bone grains size, agitation rate, and the effect of co-existing anions in actual samples of wastewater were studied for fluoride removal from aqueous solutions. Results indicated excellent fluoride removal efficiency up to 99.4% and 99.8% using lamb and chicken bones, respectively at fluoride initial concentration of 10 mg F/L and 120 min contact time. Maximum fluoride uptake was obtained at neutral pH range 6–7. Fluoride removal kinetic was well described by the pseudo-second order kinetic model. Both, Langmuir and Freundlich isotherm models could fit the experimental data well with correlation coefficient values >0.99 suggesting favorable conditions of the process. Furthermore, for complete sustainable management of waste bones, the resulted fluoride-bearing sludge was reused in concrete mixes to partially replace sand. Tests of the mechanical properties of fluoride sludge-modified concrete mixes indicated a potential environmentally friendly approach to dispose fluoride sludge in concrete and simultaneously enhance concrete properties.

1. Introduction

High fluoride levels in drinking water has become one of the most critical health hazards of this century as it induces intense impact on human health including skeletal and dental fluorosis (Sujana et al., 2009). Fluoride containing wastewater is generated in various industries such as, semiconductor manufacturing industries, coal power plants, glass and ceramic production units, uranium refinement units, electroplating, rubber, fertilizer manufacturing units contribute a lot toward fluoride pollution (Paudyal et al., 2011; Swain et al., 2012). The optimum fluoride level in drinking water safe for consumption, set by WHO, is considered to be between 0.5 and 1.0 mg/L (Swain et al., 2012; Biswas et al., 2009; Karthikeyan et al., 2011). Due to the public health significance of high fluorides consumption in drinking water, defluoridation is important (Emamjomeh et al., 2011).

During recent years, several methods including sorption, chemical treatment, ion exchange, membrane separation, electrolytic defluoridation and electro-dialysis have been developed to remove fluoride ions from water (Mohapatra et al., 2009; Emamjomeh and Sivakumar, 2009; Bia et al., 2012). Among these methods, sorption is still one of the most extensively used methods for the removal of fluoride ions from aqueous solution due to its low cost and viability (Swain et al., 2012; Biswas et al., 2009; Jamode et al., 2004; Chakrapani et al., 2010). In recent years considerable attention has been devoted to develop new cost effective natural materials for the removal of fluoride from water such as wheat straw, sawdust, Bagasse carbon of sugarcane, bone char, treated citrus limonum (*lemon*) leaf, as reported by Yadav et al. (2013), Rojas-Mayorga et al. (2013) and Tomar et al. (2014). Several studies on using modified waste bones for fluoride removal have been previously reported. However, none of them dealt with using raw unmodified waste bones for this purpose. On the other hand, management of sludge generated by treatment of groundwater contaminated with geogenic contaminants including fluoride, arsenic, and iron is a major issue in developing nations. Frequently, the sludge is stored in a pit or disposed on ground leading to possible contamination of surface water and groundwater sources. The construction industry has shown great gains in the recycling of generated waste, including solid and semi-solid wastes. Recycling of waste materials to partially replace the fine

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http://dx.doi.org/10.1016/j.wasman.2015.06.039
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aggregate in concrete industry not only saves landfill space but also reduces the demand for extraction of natural raw material for construction activity (Ismail and Al-Hashmi, 2009). Re-use of geogenic sludge materials in the manufacturing of bricks was being explored as pathways for minimizing their impact on the geo-environment, similar to studies made with industrial/municipal sludge (Rao et al., 2009). Baeza-Brotons et al. (2014) studied the viability of using sewage sludge ash (SSA) as a raw material in the composition of concrete. Zhan and Poon (2015) explored the feasibility of reutilizing textile effluent sludge (TES) for producing concrete blocks.

This study was undertaken to examine for the first time the validity of recycling raw unmodified lamb and chicken bones as an eco-friendly solid waste of no economic value for the removal of excess fluoride from aqueous solutions. Additionally, for a complete sustainable waste management, a novel reutilization of the generated fluoride bearing-sludge to partially replace fine aggregate in concrete mixes was investigated.

2. Materials and methods

2.1. Preparation of raw bone particles

Waste bones were collected from local slaughter houses, butchers, and restaurants. The bones were manually cleaned from meat pieces, repeatedly washed, and subsequently boiled in distilled water for 4 h in order to remove fats. The cleaning process was repeated 4 times. Thereafter, the clean bones were dried at 100 °C for 8 h. The dried bones were crushed, milled, and sieved into 4 different particle size ranges which were (>0.075), (0.075–0.30), (0.3–1.18) and (1.18–2.34) mm for the powder, small, medium, and large size-particles, respectively.

2.2. Characterization of bone material

Samples of raw bone waste materials were analyzed by X-ray diffraction (XRD), using X-ray Diffractometer with rotation speed of (1000°/min). The changes in the diffracted X-ray intensities are plotted against the rotation angles of the sample; analysis of the peak positions enables qualitative analysis, lattice constant determination and/or stress determination of the sample. The specific surface area was determined according to the Brunauer–Emmett–Teller (BET) theory, using N_2 as the adsorbate at 77 K (−196 °C). Measurements were conducted by surface analyzer type Q surf 9600. Before each run, the samples were exposed to a flow of 70% helium and 30% nitrogen for 1 h at 200 °C, pore volume measured in the same apparatus using multi-point capability which allows complete analysis of sorption isotherm and total pore volume of the sample. Detection of the surface functional groups for the bone waste material before and after the uptake of fluoride was carried out using Fourier Transforms Infrared (FT-IR) technique using an EQUINOX FT-IR 55 spectrometer. The bone waste samples were grounded with 200 mg of KBr in a mortar and pressed into a 10 mm diameter disks. The scanning range was 4000–400 cm^{-1}. X-ray Fluorescence analysis was carried out to determine the mineral compositions of the raw bone samples. The system consists of lithium silicon detector; measuring samples as a compressible oven dried- bone powder weighting 2–4 g.

2.3. Analysis of fluoride concentration

Fluoride concentrations in aqueous samples were measured using combined ion selective electrode (Ion lab pH/Ion/Cond 750, WTW). Double checking of fluoride concentrations was performed using ion chromatography. Triple checking was carried out using another type of separate Ion Selective Electrode (Model Orion).

2.4. Fluoride sorption experiments

Fluoride sorption batch experiments were carried out at room temperature by agitating 100 mL of the prepared fluoride solutions at various initial concentrations contained in 250 mL-plastic Erlenmeyer flasks. After proper pre-determined time intervals, samples of supernatant were filtered and the filtrates were analyzed to determine the residual fluoride concentration in the supernatant. The pH was adjusted to different values over a range of 3–11 by the addition of either 1 M NaOH or 1 M HCl.

In order to determine the optimum conditions for maximum uptake of fluoride by the raw bone waste materials, the effect of several key parameters including bone dose (1, 5, 10, 15, 20, and 25 g/L), particles size of (>0.075), (0.075–0.30), (0.3–1.18) and (1.18–2.34) mm, fluoride initial concentration (5, 10, 25, and 35 mg F/L), contact time ranged from 15 to 240 min, agitation rate (100, 150, and 200 rpm), and pH range of 3–11 were carefully considered in this study.

2.5. Preparation of modified concrete mix with fluoride bearing-sludge

In order to check the overall efficiency, feasibility, and sustainability of the suggested treatment approach, the resulted fluoride-loaded sludge needs to be further processed. Regeneration of the exhausted waste material for excessive treatment cycles seems to be a promising process, but on one side, additional cost could shift the process from being feasible. On the other hand, results of the fluoride removal experiments indicated that chemical precipitation was one of the major governing mechanisms for F^- removal, causing the formation of fluoride-bearing precipitate which needs to be treated in an environmentally friendly approach. Accordingly, a decision was made to examine the validity of utilizing the fluoride-loaded waste materials in concrete mixes to partially replace sand by 2.5%, 5.0%, 7.5%, and 10% by weight. The effect of sand replacement by fluoride-loaded sludge on the mechanical properties including the slump, compressive strength, flexural strength and dry density of concrete mixes after 7 and 28 day was investigated. Ordinary Portland cement was used in the preparation of concrete specimens. Natural crushed stone aggregate of maximum size (20 mm) was used as coarse aggregate while, the fine aggregate used in this study was natural sand of 4.75 mm maximum size and of desert origin.

3. Results and discussion

3.1. Characterization of bone waste samples

The XRF analysis of raw bone waste materials before fluoride uptake indicated that the main elemental components of the raw bone samples are calcium and phosphorus, as well as traces of other elements, mainly sodium, magnesium, aluminum, iron and zinc. However, results of (XRF) analysis after fluoride removal revealed that the concentration of the main components; calcium and phosphorus were reduced indicating the involvement of these elements in the fluoride uptake process.

Surface area values determined by Brunauer–Emmett–Teller (BET) method revealed that the surface area values decreased with increasing the particle size. As given in Tables 1 and 2, the values of surface area and pore volumes, respectively could be a notable indication for the difference in fluoride removal capacity between the lamb and chicken bones.