

Standardizing defluoridation of community waters using bone char

Godfrey K. Mbabaye, Felix Mtaló, Rwaichi J. A. Minja and Isack Legonda

ABSTRACT

Different bone char (BC) preparation methods affect the physical chemical properties and therefore the capacity to remove fluoride. Fluoride removal capacities of BC prepared at three controlled temperatures, with particle grain sizes of 250–500 μm , 500–1,000 μm and 1,000–1,800 μm were determined in column experiments with an initial fluoride concentration of 8.55 mg/L. The sorbent was calcined at 400°C, 500°C and 600°C. BC calcined at 400°C had better fluoride removal performance compared to those calcined at 500°C and 600°C due to decreased pore volume and surface area as the calcination temperature was increased. There was a reduced mass transfer effect to the adsorption sites in pores as the BC particle size was increased. The equilibrium adsorptions Langmuir and Freundlich isotherms were tested. For the Langmuir equilibrium adsorption isotherm, maximum monolayer coverage (Q) was determined to be 3.512 mg/g, and the value of the separation factor (r) obtained was 0.1394 indicating favorable adsorption as it lies between 0 and 1. The Freundlich governing equilibrium adsorption isotherm model gave a value of $1/n$ equal to 0.445. This indicates a favorable adsorption process, since the bond energies increase with surface density of the adsorbent.

Key words | adsorption isotherms, bone char, defluoridation, Freundlich isotherms, Langmuir isotherm, pore surface area

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INTRODUCTION

Application of the adsorption process using locally available adsorbents is one of the cheap methods of defluoridation of fluoridated water in Tanzania. However, the adsorbents have to be prepared properly. Calcination of animal bones produces an effective material for removing fluoride. Depending on the charring process, the bones may result in a product with low defluoridation capacity (Dahi 2000). The production process of bone char (BC) therefore is one of the most important steps for development of proper structural and physico-chemical properties of BC. Calcination temperature and duration influence the pore structure formation and the surface chemistry of BC (Bonilla-Petriciolet *et al.* 2013).

Good quality BC is an effective material for removing fluoride (Jacobsen 2007). Effective bone calcination ensures

removal of organic matter that otherwise adds taste and color to water (Dahi 2000).

This paper reports on BC preparation method using cow bones obtained from abattoirs around Arusha in Tanzania. The objective of the research was to study the effects of temperature on the quality of BC developed and its adsorption performance.

MATERIALS AND METHODS

BC preparation

Cattle bones were collected from slaughter houses in the Arusha area. The collected bones were calcined under

controlled temperature and rate of heating in a thermo scientific (Thermolyne) programmable furnace in a laboratory condition. Bones were calcined at temperatures of 400°C, 500°C and 600°C in a furnace. According to Posner (1987), temperatures higher than 600°C damage the apatite structure, while temperatures below 400°C result in charred bones, giving a bad taste and odor when used for treating water owing to incomplete volatilization of organic matter. The temperature of the electric furnace was raised from room temperature at a controlled rate of 8°C/min and maintained at a constant temperature for three hours. Smoke release was used as an indicator of removal of volatile matter. For example, at 400°C, smoke started to be emitted at 250°C and at 350°C dense smoke was emitted. Smoking stopped after one hour at 400°C, which means charring was continued for a sufficient time at 400°C to allow all volatile matter to be exhausted and an extra time to allow for necessary structural change without leading to sintering. After charring, grinding and sieving, the BC was washed with deionised water in order to remove the dust before packing into the column filter. In so doing smell, color and bad taste were controlled. The calcined bones were allowed to cool to room temperature before further treatment.

Calcined bones were milled using a laboratory milling machine, Retschmuhle, GmbH, type: SK1 Number 72307. Thereafter, ground BC was sieved and graded using electromagnetic vibration sieves set with diameters ranging between 100 µm and 1,800 µm in order to get different BC particle sizes according to Laboratory Test Sieve ISO 565, part no. 667924, Endecotts Ltd, London, UK. These sizes comply with wire mesh series standard ISO 3310-1:2000 and BS 410-1:2000.

Experimental set up

A defluoridating BC column unit was constructed using a transparent Plexiglas pipe with a diameter of 38 mm. The column was closed at the lower end with an inlet regulating valve and at the upper end with an outlet valve. Naturally fluoridated water with an initial concentration of 8.55 mg/L maintained an upward flow through the column at a rate of 2.2 cm³/s. The water flow rate was maintained through the Plexiglas pipe by a constant head reservoir. The Plexiglas pipe had a height of 370 mm and was packed with 250 g of BC. The diameters of BC particle sizes used were within

250–500 µm, 500–1,000 µm and 1,000–1,800 µm, each calcined at temperatures of 400°C, 500°C and 600°C. The BC with particles of diameter less than 250 µm clogged the flow in the experiment column and therefore it was not used in this study. The flow rate was adjusted by inlet valve and maintained at 2.2 cm³/s (Figure 1). The effluent was collected in a sampling tube. Samples were collected at a time interval of 2 minutes for the first 10 minutes, then at intervals of 5 minutes up to 30 minutes, 10 minutes up to 60 minutes, 30 minutes up to 180 minutes and finally 1 hour up to 6 hours. This was to take care of fast fluoride adsorption in the initial stages.

Fluoride concentration measurement

Standard analytical methods (APHA 1989; HACH 1997) were used to quantify the presence of fluoride ion concentration. A 5 ml sample solution was mixed with 5 ml of 1,2-cyclohexanedinitrilo-tetraacetic acid-total ionic strength adjustment buffer, and the fluoride concentrations were measured using an ion selective electrode technique as described by Thole et al. (2012).

BC morphology and pore structure

The scanning electron microphotograph (SEM) of the calcined BC was studied using TESCO SEM model

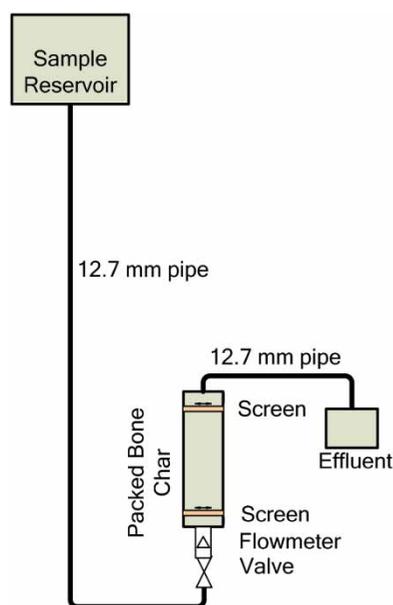


Figure 1 | A schematic diagram of BC column experimental set up.

HCITMSAAW11, 5 V, 500 mA, SC: 102535 to evaluate surface morphology. The surface area and pore volume of the BC were determined by the Brunauer–Emmet–Teller method using a computer-controlled nitrogen gas adsorption analyzer at 273.0 K in the Chemistry Department laboratory at the University of Dar es Salaam.

BC X-ray fluorescence analysis

The X-ray fluorescence (XRF) Bruker AXS S8 Tiger model with serial number 206548 was used to determine the chemical composition of the BC media charred at different temperatures. Results are presented in Table 1. Taking BC charred at 400°C as a base, the oxide composition was found to change for some of the minor components (SiO₂, SO₃, Al₂O₃, Fe₂O₃ and CuO) in BC charred at temperatures of 500°C and 600°C. Therefore, this is an indication of structural change in the developed BC as the charring temperature was changed.

Defluoridation capacity of BC

The defluoridation capacity of the prepared BC was determined using the following mass balance equation

(Thole *et al.* 2012):

$$D_c = \frac{C_i - C_f}{m} * V \quad (1)$$

where D_c is the defluoridation capacity (mg/g), C_i is the initial fluoride concentration (mg/L), C_f is the residual fluoride concentration (mg/L), V is the volume of treated water (L), and m is the mass of BC media used (g).

The governing equilibrium adsorption isotherm

The governing equilibrium adsorption isotherm for the BC prepared under controlled conditions was studied. The relationship between the equilibrium amount of fluoride on the adsorbent and solute concentration in water is well described by an isotherm. The purpose of an adsorption isotherm is to relate the adsorbate concentration in the bulk solution to the amount adsorbed at the solid/water interface (Kishore *et al.* 2011). To model the process, a batch experiment was carried out. The equilibrium data were fitted to both Langmuir and Freundlich isotherm equations. The Langmuir adsorption isotherm is used to describe the formation of monolayer adsorbate on the outer surface of the adsorbent quantitatively at the equilibrium concentration

Table 1 | Result of XRF analysis showing major component of bones charred at different temperatures

Compound	Bones charred at 400°C Compound content (%)	Bones charred at 500°C		Bones charred at 600°C	
		Compound content (%)	% decrease or increase	Compound content (%)	% decrease or increase
CaO	55.21	55.27	0.1	55.2	0.0
P ₂ O ₅	39.23	40.88	4.2	41.43	5.6
Na ₂ O	1.5	1.44	−4.0	1.41	−6.0
MgO	1.22	1.18	−3.3	1.21	−0.8
SiO ₂	1.04	0.37	−64.4	0.13	−87.5
SO ₃	0.83	0.31	−62.7	0.12	−85.5
Al ₂ O ₃	0.28	0	−100.0		−100.0
Cl	0.18	0.17	−5.6	0.16	−11.1
Fe ₂ O ₃	0.15	0.07	−53.3	0.08	−46.7
SrO	0.13	0.13	0.0	0.13	0.0
K ₂ O	0.1	0.11	10.0	0.11	10.0
CuO	0.09	0.03	−66.7		−100.0
ZnO	0.02	0.02	0.0	0.03	50.0

whereby no further adsorption takes place, and hence representing the equilibrium distribution of the ions between the solid and liquid phase (Said & Machunda 2014).

Batch experiments for adsorption studies were carried out in the laboratory by adding a known amount of BC to four flasks containing 500 mL of fluoride water with an initial fluoride concentration of 21 mg/L. The mixture was stirred at the rate of 100 rpm using Phipps & Birds, PB-700TM Jartester Machine (Catalog No. 7790 – 752B, Serial No. 198070026 269900) until the equilibrium of residual fluoride concentration was attained. Table 2 gives the residual fluoride concentration in each flask at equilibrium.

The amount of solute adsorbed at equilibrium was determined using Equations (2) and (3) as tabulated in Table 3.

$$\text{Amount adsorbed at equilibrium, } (X) = \frac{(C_i - C_e) * V}{1,000} \quad (2)$$

$$q_e = \frac{X}{m} \quad (3)$$

where C_i is the initial fluoride concentration (mg/L), C_e is the fluoride concentration at the equilibrium (mg/L), m is

the mass of BC used (g), V is the volume of fluoridated water used (mL) and q_e is the equilibrium amount of fluoride adsorbed on BC (mg/g).

The Langmuir isotherm (Mutchimadilok et al. 2014) is given by

$$q_e = \frac{QbC_e}{1 + bC_e} \quad (4)$$

where q_e is the equilibrium amount of fluoride adsorbed on BC (mg/g), C_e is the equilibrium fluoride concentration (mg/L), Q is the Langmuir isotherm constant related to capacity (mg/g), and b is the Langmuir isotherm constant related to energy (L/mg).

To determine the Langmuir adsorption parameters, the linear form transformed equation was used:

$$\frac{C_e}{q_e} = \frac{C_e}{Q} + \frac{1}{Qb} \quad (5)$$

or

$$\frac{1}{q_e} = \frac{1}{Q} + \frac{1}{Qb} * \frac{1}{C_e} \quad (6)$$

Table 2 | Residual fluoride values at equilibrium

Flask no.	Mass of BC (g)	Volume in flask (mL)	Initial fluoride concentration (mg/L)	Residual fluoride at equilibrium, C_e (mg/L)
1	1	500	21	15
2	5	500	21	3.5
3	7	500	21	2.2
4	10	500	21	1.3
5	0	500	21	21

Table 3 | Computation of equilibrium amount of fluoride adsorbed on BC

Flask no.	Mass of adsorbate (g)	q_e (mg/g)
1	1	3
2	5	1.75
3	7	1.34
4	10	0.99

Using the values in Tables 2 and 3, the Langmuir equation parameters in Table 4 were calculated.

The Freundlich isotherm model is assumed to be associated with heterogeneous sites (Sarkar et al. 2006) and is used for fitting results of sorption experiment. The equation is as follows (Jayapriya et al. 2011):

$$q_e = K_d C_e^{1/n} \quad (7)$$

Table 4 | Computed Langmuir equation parameters

Flask no.	$1/C_e$	$1/q_e$
1	0.06667	0.33333
2	0.28571	0.57143
3	0.45455	0.74468
4	0.76923	1.01523

where q_e is the amount of fluoride ions adsorbed per unit weight of adsorbent (mg/g), K_d and $1/n$ are the Freundlich constants and C_e is the equilibrium fluoride concentration (mg/L).

Kishore *et al.* (2011) described this equation as follows: if $1/n < 1$, the bond energies increase with the surface density of the adsorbent, and if $1/n > 1$ the bond energy decreases with the surface density of the adsorbent, and if $1/n = 1$ all surface sites are equivalent related to sorption capacity and sorption intensity.

The values of C_e and q_e from Tables 2 and 3 were used to compute Freundlich isotherm parameters as tabulated in Table 5.

$$\log(q_e) = \log(K_d) + \frac{1}{n} * \log(C_e) \quad (8)$$

Table 5 | Computed Freundlich adsorption isotherm parameters

Flask no.	C_e (mg/L)	Log C_e	q_e (mg/g)	Log q_e
1	15	1.17609	3	0.47712
2	3.5	0.54407	1.75	0.24304
3	2.2	0.34242	1.34	0.12803
4	1.3	0.11394	0.99	-0.00656

RESULTS AND DISCUSSION

Effect of particle size and temperature on fluoride removal

The effect of particle size on fluoride removal for BC charred at 400°C and 500°C are shown in Figure 2. Reduced mass transfer effects to the adsorption sites in pores and on the particle surface is obvious as the particle size increases. Fluoride adsorption with particle sizes above 1,000 μm indicates the process to be mass transfer limited. The results are similar for all BC calcination temperatures of 400°C, 500°C and 600°C. Studies by Mavura *et al.* (2004) and Thole *et al.* (2012) also gave similar trends. In this study the effect of particle size was also studied by batch method with an initial fluoride concentration of 10.2 mg/L. The findings showed that the smallest particle sizes, that is 0.1–0.15 mm, had better adsorption ability than the rest. But particle sizes below 0.25 mm were found to clog the column, therefore it was found not suitable for use in a column applications. It was hence decided to work on particle size (d_p) in the range of $0.25 \text{ mm} < d_p < 1.8 \text{ mm}$. For which, the smallest particle size suitable for column defluoridation adsorption was recommended to be 0.25–0.5 mm.

The effect of calcination temperature on the BC adsorption capacity was observed by looking at the residual fluoride concentration as a function of time. BC calcined

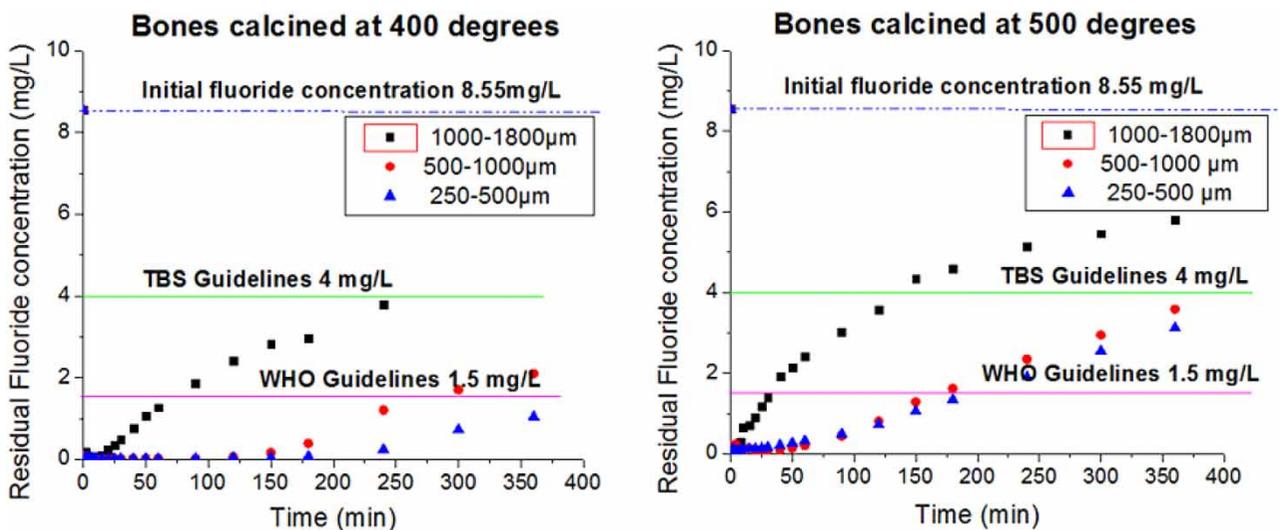


Figure 2 | Effect of particle size and calcination temperature on fluoride removal.

at 400°C could be used for a longer time than one calcined at 500°C before reaching a maximum residual fluoride concentration of either 1.5 mg/L or 4 mg/L as per the World Health Organization standard and Tanzania Bureau of Standards, respectively.

The decrease in adsorption removal performance as the calcination temperature was increased from 400°C to 500°C and 600°C can be explained by the results of Figure 3 which show decreased pore volume and surface area of the BC as the calcination temperature was increased. Increase in calcination temperature led to sintering, hence, a decrease in pore volume and surface area. The resultant effect was a decrease in mobility of solution in the column and a decrease adsorption capacity of the adsorbent. The sintering effect is observed in the results of the SEM in Figure 4, which shows that, at low temperature (400°C) the surface of the BC is rough compared to that at higher temperature (600°C), which is smoother.

BC removal capacity

The effect of BC calcination temperature on fluoride removal rate capacities is shown in Figure 5. This study was done using 250 g of the media with particle grain size of 250–500 µm. It can be seen that the BC calcined at 400°C had the best removal rate capacity compared with those calcined at 500°C and 600°C. Thus, the calcination temperature has an effect on the developed structural morphology and temperatures above 400°C result in loss of fluoride adsorption performance of the BC because of the sintering effect.

The effect of particle size on the fluoride removal capacity for BC calcined at different temperatures were determined using Equation (1), at fluoride removal breakthrough point of 1.5 mg/L as shown in Figure 6. Equation (1) indicates the amount of fluoride in milligrams removed by one gram of BC used with the initial fluoride

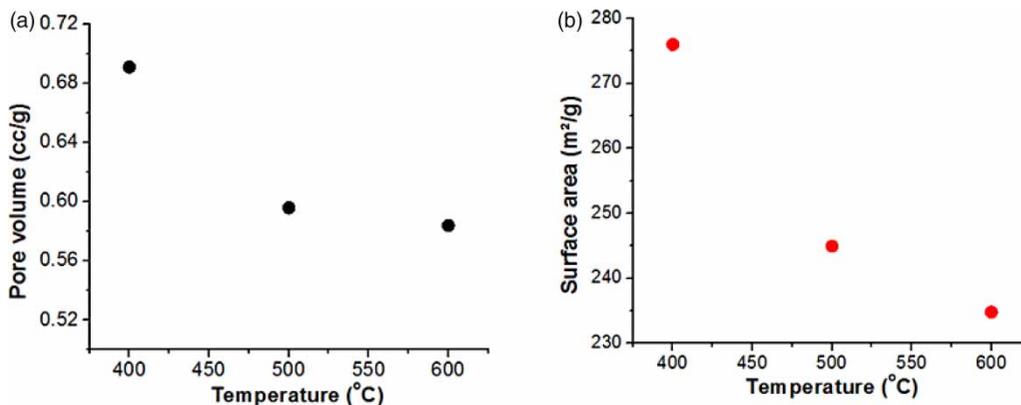


Figure 3 | Effect of calcination temperature on BC pore volume and surface area.

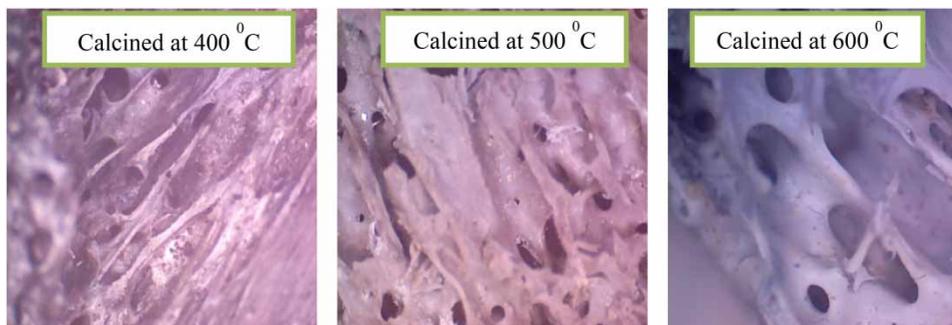


Figure 4 | SEM analysis of BC calcined at different temperatures.

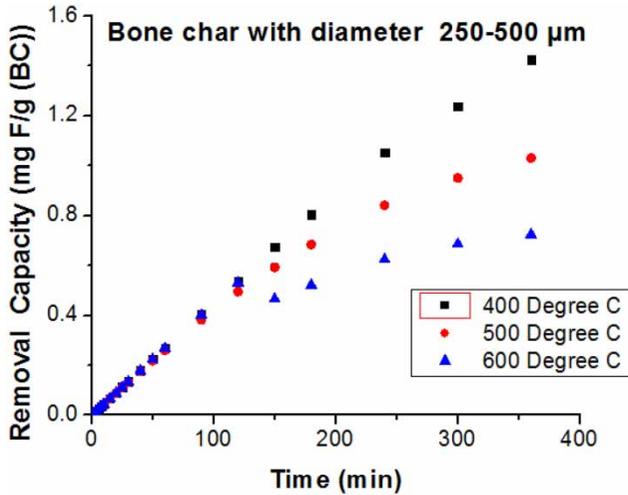


Figure 5 | Effect of temperature on fluoride removal capacities with time.

concentration being 8.55 mg/L. The results show that, BC with a particle size of 250–500 µm calcined at 400°C had the best fluoride removal capacity. Similar findings have been reported by Mjengera (1988) and Hauge *et al.* (1994), who reported that fluoride removal by BC is a surface reaction process and that as the surface area is increased, the effectiveness of the BC in removing fluoride also increases. This is clearly shown by the higher removal capacity with the BC particle range of 250–500 µm compared with the other, larger particles. Figure 6 also shows a decrease in fluoride removal capacity as the calcination temperature was

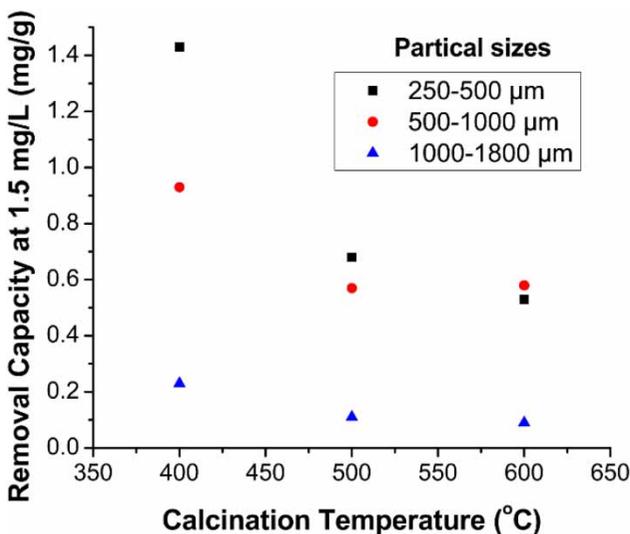


Figure 6 | Fluoride removal capacities of BC at 1.5 mg/L breakthrough point.

increased to 500°C and 600°C as a result of a decrease in pore volume and surface area.

BC equilibrium adsorption isotherm: determination of the Langmuir isotherm equation parameters

The Langmuir adsorption isotherm describes quantitatively the formation of a monolayer adsorbate on the outer surface of the adsorbent. The transformed linear Langmuir isotherm, Equation (6), was plotted using values in Table 4. A plot of $1/q_e$ against $1/C_e$ (Equation (6)) should yield a straight line if the data fit the model. The results gave a linear relationship as shown in Figure 7(a), with R^2 value of 0.994. The maximum monolayer coverage capacity, Q , and Langmuir isotherm constant related to energy, b , in Equation (6) were obtained from the intercept and slope, respectively. From this research work, the values of the constants Q and b calculated from the intercept and slope were 3.512 mg/g and 0.294 L/mg, respectively.

The essential features of the Langmuir isotherm are described by a dimensionless quantity, r , referred to as separation factor (Prasad *et al.* 2000), which indicates the isotherm's shape and the nature of the adsorption process as unfavorable, favorable, linear or irreversible. The dimensionless quantity is expressed as

$$r = \frac{1}{1 + b \cdot C_0} \quad (9)$$

where C_0 is the initial fluoride concentration.

The feasibility criteria of the adsorption process were judged from the r -values as shown in Table 6 (Sarkar *et al.* 2006; Hameed *et al.* 2008).

The value of r obtained was 0.139, indicating that the adsorption process was favorable ($0 < r < 1$) for the adsorbent in the removal of fluoride ions, and the R^2 value of 0.994 proves that the adsorption data fitted well to the Langmuir isotherm model.

Freundlich isotherm

The Freundlich isotherm describes the adsorption characteristics for the heterogeneous surface. It is an empirical equation that encompasses the heterogeneity of sites and

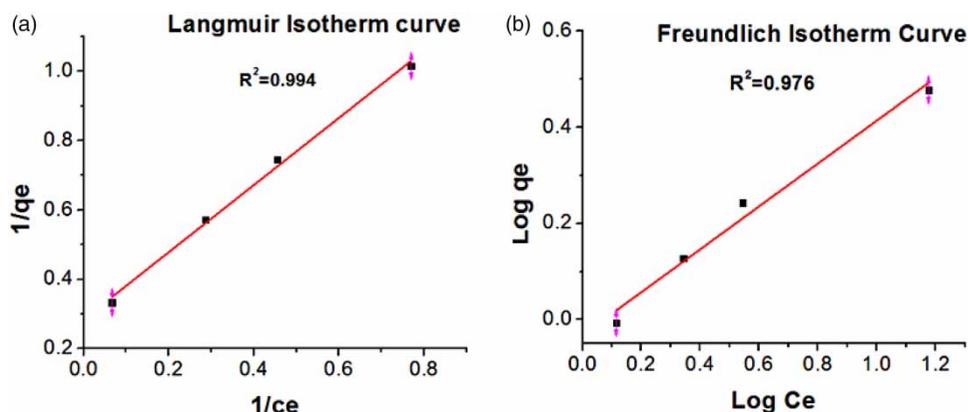


Figure 7 | Langmuir and Freundlich isotherm curves for BC fluoride adsorption.

Table 6 | The feasibility criteria of r -values

Value	Description
$r > 1$	Unfavorable
$r = 0$	Irreversible
$0 < r < 1$	Favorable
$r = 1$	Linear

the exponential distribution of sites and their energies (Oladaja et al. 2008). Fitting of the computed Freundlich isotherm parameters in Table 5 using the linear form of the Freundlich isotherm, Equation (8), gave a correlation as shown in Figure 7(b). The constant, K_d , is an approximate indicator of adsorption capacity, while $1/n$ is a function of the strength of adsorption in the adsorption process (Dada et al. 2012). The plot of $\log q_e$ against $\log C_e$ yielded a linear relationship as expected, with slope $1/n$ and the intercept $\log K_d$ of values 0.446 and -0.032 , respectively.

Kishore et al. (2011) reported that, if $1/n < 1$, bond energies between sorbent and sorbate increase with the surface density of the adsorbent; if $1/n > 1$, bond energies decrease with the surface density of the adsorbent, and if $1/n = 1$ all surface sites are equivalent. Dada et al. (2012) said that if the value of $1/n$ is below 1 it indicates a normal and

favorable adsorption process. In this work, the plot of $\log (C_e)$ versus $\log(q_e)$ was linear with $R^2 = 0.976$, as shown in Figure 7(b), with $1/n = 0.445$ confirming the applicability of the Freundlich isotherm model for the removal of fluoride ions by the BC calcined at 400°C .

The isotherm parameters evaluated from BC calcined at a temperature of 400°C for Langmuir and Freundlich equations are summarized in Table 7.

CONCLUSION

The SEM of the calcined BC showed an increased sintering effect as the calcination temperature was increased from 400°C to 500°C and 600°C . This decreases the adsorption capacity of the adsorbent and hence reduces its adsorption performance as calcination temperature was increased.

Investigation of equilibrium adsorption carried out indicated that for the Langmuir adsorption model, the separation factor or equilibrium parameter (r) value calculated was greater than 0 and less than 1, indicating that the Langmuir adsorption isotherm is favorable, while for the Freundlich model the value of $1/n$ indicates the strength of adsorption is less than 1, indicating a normal and favorable

Table 7 | Isotherm parameters

Calcination temperature (°C)	Langmuir			Freundlich	
	Q (mg/g)	b (L/mg)	r	1/n	K_d (L/g)
400	3.512	0.294	0.139	0.446	0.929

adsorption process. From this result, it can be concluded that the process of adsorption fitted the Langmuir model ($R^2 = 0.994$) better than the Freundlich model ($R^2 = 0.976$).

In this work, therefore, favorable preparation conditions for calcination of BC and particle sizing for fluoride adsorption were: calcination temperature of 400°C and a particle sizes range of 250–500 µm for the removal of fluoride from naturally fluoridated water. BC calcined at temperatures of 500°C and 600°C appeared to have less pore volume and reduced surface area. Particle sizes below 250 µm led to column clogging, while particles above 1,000 µm led to limited mass transfer.

ACKNOWLEDGEMENTS

The author acknowledges the financial support received from the Royal Society of London through Leverhulme Royal Society Africa Award Scheme. The sponsors did not have any involvement in the research design, execution and data analysis nor in the preparation and submission of this paper.

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First received 17 June 2016; accepted in revised form 26 October 2016. Available online 30 January 2017