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Defluoridation of Water Supplies Using Coconut Shells Activated Carbon: Batch Studies

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Abstract: *Drinking water with elevated fluoride levels results in serious irreparable health problem that has attained an alarming dimension all over the world, Tanzania being one of the affected countries; techniques have been under study for years. Batch experiments were carried out to determine the effect of various adsorbent factors such as adsorbent dose, initial pH, particle size and contact time on adsorption process. Adsorption efficiency was observed to increase with decrease in particle size, the highest efficiency recorded was 68.2 and 65.9% for field and synthetic water respectively when particle size less than 150 μ of coconut shells activated carbon were used. Adsorption was observed to be favoured by pH in acidic range and the maximum efficiency of 58.4% was recorded at pH of 2.0 and particle sizes between 4.18-2.36mm. The pH of the effluent was lowered to acidic range which necessitates further treatment of the effluent or coupling with other materials for pH elevation before use. Adsorption increases with adsorbent dose hence at a smaller influent concentration; the required standard of 1.5mg-F/l can be met. Equilibrium isotherms have been analysed using Langmuir and Freundlich isotherm models, and both of the models fit to explain the adsorption behaviour of fluoride ions onto Coconut shell activated carbons, during this study it was established that when properly activated; coconut shells can be appropriate for use household filters that could be cost effective in rural areas of Tanzania due its local availability and its colour adsorption property.*

Keywords: Defluoridation, Fluorosis, Coconut shell activated carbon, Adsorption.

1. Introduction

The availability of abundant contaminants deters the safety of water as life giving medium, posing a decisive challenge as a natural environmental life support resource [2]. The world health organization claims about 1.1 billion People in low and medium-class income countries to lack access to safe potable water, personal and domestic use [29]. One of the well-known natural contaminant for ground water resources all over the globe is Fluorine, available in the form of fluorides in a fairly number of minerals and in many rocks [5]. In accordance to Water Quality Association, about 0.08% of the Earth's crust is made up of fluoride compounds.

Fluoride is widespread and can be found in different sources ranging from water to food stuffs, vegetables contains low fluoride levels ranging from 0.1 to 0.4 mg/kg. Food stuffs with elevated fluoride a level includes but not limited to barley and rice with about 2 mg/kg and fish with 2-5 mg/kg [28]. IPCS argues that canned fish may contain up to 370 mg-F/kg [10], Dry tea leaves may contain fluoride concentration of up to 400mg/kg and the exposure of ranges 0.04 to 2.7 mg/person/day [17]. Ingestion of fluoride contaminated water has been reported to be the biggest contributor of fluoride exposure to humans, accounting 75-90% of daily intake of the fluoride ion [14].

According to Mkongo and Mjengera; the government of Tanzania realised the fluoride problem in the country since 1950 [16]. High fluoride in water threatens the Northern and western parts of country [21]. Mount Meru, one among these areas is reported to contain fluoride concentrations as high as 68 mg/L [8]. Drinking of fluoride contaminated water poses a health risk to people in fluorotic areas, these effects ranges from mottling of teeth, softening of bones and ossification of tendons and ligaments [2]. It is shown that, over 70 million

people are affected by fluorosis worldwide [7]

The World Health Organization (WHO) recommends a fluoride content of 1.5 mg-F/l for drinking water [28] while Tanzania bureau of standards (TBS) recommends the lower limit fluoride to be 1.5mg-F/l and 4.0mg-F/l as the tolerable fluoride level for potable water [22], however the observation from the world health organization claim the level in range of 1.5 to 4.0 mg-F/l to cause dental fluorosis [27] which necessitate the amendment in Tanzanian standards for drinking water.

Chaturvedi reported defluoridation of water to be the only measure to prevent fluorosis [4], however according to Jamode and colleagues; the conventional defluoridation materials are expensive, non-sustainable or environmental unfriendly [11]. It is therefore important to investigate defluoridation by sustainable, cheap and environmental friendly materials such as activated carbons from coconut shells.

Coconut is among cash crops which are largely cultivated in coastal regions in Tanzania [19] and southern coastal regions of the country [15] respectively. Coconut cultivated land in Tanzania is about 240,000 hectares [19].

The current use of these wastes as alternative source of charcoal rises due to its high calorific blessing [26] and high carbon content [22]. Its high carbon content triggers its application for water purification; coconut shell charcoal can be activated and became potential for water purification [25].

The purpose of this work is therefore to explore the potential of coconut shells as a least cost method for fluoride removal from drinking water. This study therefore focuses more on activating the coconut shells activated carbon (CSC) and

their application in batch studies at various conditions for defluorination of synthetic water and also samples of water collected from the field. It is necessary to compare such two samples since they differ in composition.

2. Experimental Methodology

2.1 Materials

Plastic apparatus and analytical reagent grade chemicals were used. The use of plastic materials was aimed to minimize the possibility of Fluoride getting absorbed. The apparatus were washed off with deionized water before and after use, solutions of different concentrations were prepared by dissolving an appropriate amount of sodium fluoride (NaF) in deionized water to make Fluoride contaminated water [23]. 0.05M Sulphuric acid and 0.1M sodium hydroxide were used for adjusting the pH values to suit either acidic and/or alkaline conditions.

2.2. Activation of coconut shells carbon

Coconut shells were obtained from local traders in Arusha region, these were prepared and carbonization was done at Ngurdoto Defluorination Research Station at the temperature of 400°C in a furnace with controlled air supply. The obtained charcoal was activated with 25% NaOH, 25% CaCl₂, 2M H₂SO₄ and 2M HNO₃ respectively for 12 hours. The charcoal was then washed thoroughly with deionized water to remove any residue of activating agents, oven dried and grinded to get granular activated carbon that was finally sieved at 4.75mm, 2.36mm, 1.18mm, 600µ, 300µ and 150µ sieve sizes for the purpose of meeting different particle sizes.

2.3 Testing of activated carbons

Non-flow batch sorption studies were conducted to study the effect of controlling parameters like contact time, adsorbent dosage and solution pH. After giving a required contact time in a shaker at a speed of 100-150rpm and 25°C, the contents of the flasks were filtered using whatmann's filter paper number 41. The above procedure was repeated for different pH, contact time, adsorbent doses, particle sizes and different initial fluoride ion concentrations. The use of granular activated carbon was aimed to eliminate the need to separate the carbon from the effluent [3]. Each experiment was done in triplicate under identical conditions to minimize experimental errors.

Fluoride removal efficiency was determined in accordance to the following equation [1].

$$E = \frac{C_o - C_f}{C_o} \times 100\% \quad (0.1)$$

Where: C_o is initial fluoride concentration in water sample (mg-F/l), C_f is residual fluoride concentration at equilibrium time (mg-F/l) and E is the fluoride removal efficiency.

3. Results and Discussion

3.1 Effect of activation agent on Fluoride adsorption

With initial concentration of 5mg/l and neutral pH, the effect of activating agents in fluoride adsorption produced promising results. For Sulphuric acid activated carbon Fluoride removal was 58.4%, 14.2% for NaOH activated carbons, 23.8% for Nitric acid activated carbons, 18.7% for calcium chloride activated carbons and 3.33% for commercial purchased activated carbon (figure 1). This suggests that the locally activated coconut shell carbons produces a far better results than the commercial ones hence reducing the costs associated with sourcing and purchase of the activated carbons for F⁻ removal. However the pH of the effluent was lowered to 2.8 which are in contrary with the potable water specification range of 6.5- 9.2 as argued by the Tanzania Bureau of Standards (TBS) [22] while that treated using NaOH produced the effluent of 10.6 which exceed the TBS guideline by 1.4 in alkaline range.

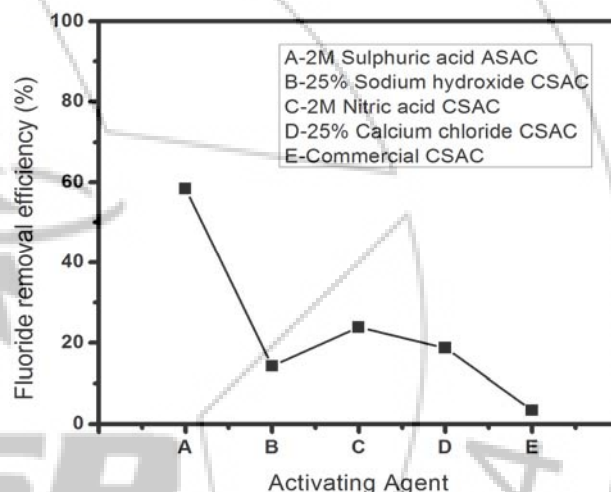


Figure 1: F⁻ adsorption as a function of activating agent

3.2 Effects of pH on F⁻ adsorption by H₂SO₄ CSAC

Fluoride removal efficiency was observed to vary with initial pH of the raw water, however the highest efficiency was 51.2% with initial raw water concentration of 15mg/l and particle size between 2.36-1.18mm which was met a pH of 2, and this means that adsorption using coconut shells activated carbon works best at acidic pH.

3.3 Effects of adsorbent dose on F⁻ adsorption by H₂SO₄ CSAC

Further experiments to determine the effect of other controlling parameters were conducted; using initial fluoride concentration of 14mg/l and 20.13mg/l for synthetic and field water respectively shows decrease in F⁻ concentration with increase in adsorbent dose for both samples. Observation shows decrease in F⁻ concentration for synthetic water up to 160g/l, beyond which the increase was not linear. A sharp drop in defluorination efficiency for field water after 120g/l was observed as depicted in figure 2. This behaviour probably indicated the saturation that might be a result of competing ions available in field water.

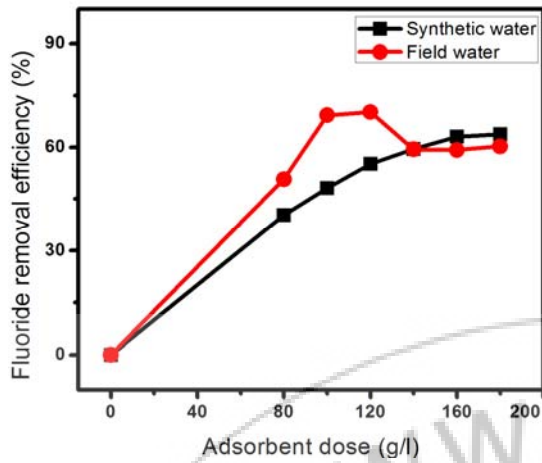


Figure 2: Fluoride removal efficiency as a function of adsorbent dose

3.4 Influence of particle size on F⁻ adsorption by H₂SO₄ CSAC

The influence of particle size in fluoride adsorption by 50g/l of 2.0M H₂SO₄ coconut shells activated carbon was investigated using 8.22mg- F/l and 13.2mg- F/l of synthetic and F contaminated water collected from one of the surface water sources in Arusha region. The fluoride removal efficiency was observed to increase with decrease in particle size (figure 3) where the highest removal efficiency was 68.2% as obtained from field water using particle size of less than 150μ. The highest removal efficiency was recorded at 65.9% with particle size of less than 150μ for synthetic water, this increase may be a result of increase in surface area which creates more adsorption sites on the small particles. Non-linear efficiency was observed when field water was used, this may be influenced by competing ions present in field water. However; the use of very fine particles lowers the quality of water by increasing turbidity which necessitate further treatment. The pH of the treated water was observed to be acidic which necessitate neutralization/pH adjustment before using it for drinking purposes or other sensitive applications. It is envisaged that coupling the coconut shell activated carbon with other materials would regulate the resultant effluent pH to neutral [18].

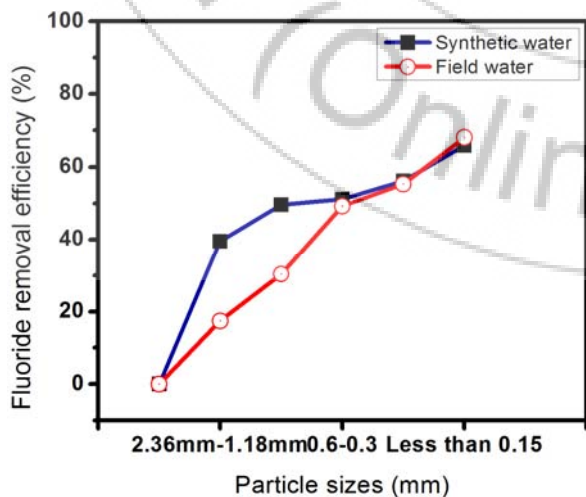


Figure 3: Fluoride adsorption as a function of particle size

3.5 effects of contact time on fluoride removal by CSAC

Using the initial fluoride concentration of 8.2 mg/l and 20.13mg/l for synthetic and field water respectively, shaking speed of 150rpm and particle size between 1.18mm and 600μ adsorption was observed to increase with increase in contact time at room temperature as depicted in figure 4. However; adsorption was nonlinear beyond 15 hours indicating the attainment of equilibrium. Field water followed almost the same trend except the sharpness of the graph which may be attributed by competing ions.

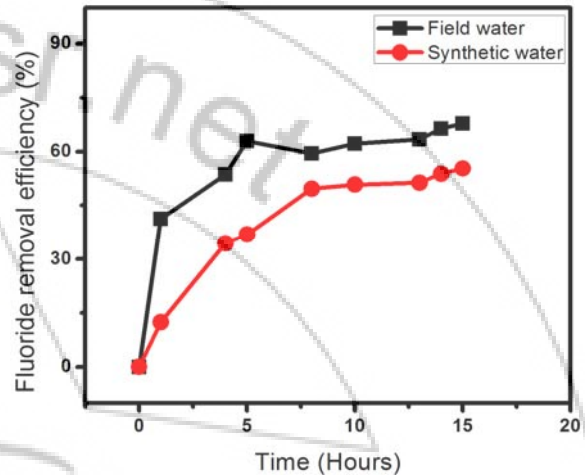


Figure 4: removal efficiency as a function of contact time

3.6 Equilibrium adsorption isothermal study

Equilibrium adsorption isothermal study to determine the adsorption capacity of coconut shell activated carbon on removing fluoride ions form water was done by using Langmuir and Freundlich adsorption theories. Langmuir adsorption isotherm is used to describe the formation of a monolayer adsorbate on the outer surface of the adsorbent quantitatively, this is done at equilibrium concentration thereby no further adsorption takes place and hence representing the equilibrium distribution of metal ions between the solid and liquid phases [24]

$$R_L = \frac{1}{1 + (1 + C_o K_L)} \quad (0.2)$$

To determine Langmuir adsorption parameters, the linear form transformed equation was used as shown below [6];

$$\frac{1}{q_e} = \frac{1}{Q_o} + \frac{1}{Q_o K_L C_e} \quad (0.3)$$

Where;

C_e is the equilibrium concentration of F⁻ (mg/L⁻¹), q_e is the amount of F⁻ adsorbed per gram of the coconut shell activated carbons at equilibrium (mg/g), Q₀ = maximum monolayer coverage capacity (mg/g) and K_L = Langmuir isotherm constant (L/mg).

The plot of 1/q_e against 1/C_e was made [13] and a straight line with slope 1/Q₀K_L was obtained which shows that the adsorption follows the Langmuir isotherm as shown in Fig. 5. The Langmuir constants K_L and Q₀ were calculated from the slope and intercept which are shown in table 2.

The essential features of the Langmuir isotherm may be expressed in terms of equilibrium parameter R_L , which is a dimensionless constant referred to as separation factor or equilibrium parameter [9].

$$R_L = \frac{1}{1 + C_o K_L} \quad (0.4)$$

Table 1: R_L values at different initial fluoride concentrations

Initial [F ⁻] (Co) (mg/L)	3.16	5.28	6.05	8.2	20.13
R_L values	0.025	0.015	0.013	0.01	0.004

The average R_L obtained was 0.0134 which indicate that the adsorption process is favourable ($0 < R_L < 1$) for the adsorbent in the removal of fluoride ions [9] and R^2 value of 0.9672 proves that the sorption data fitted to Langmuir Isotherm model.

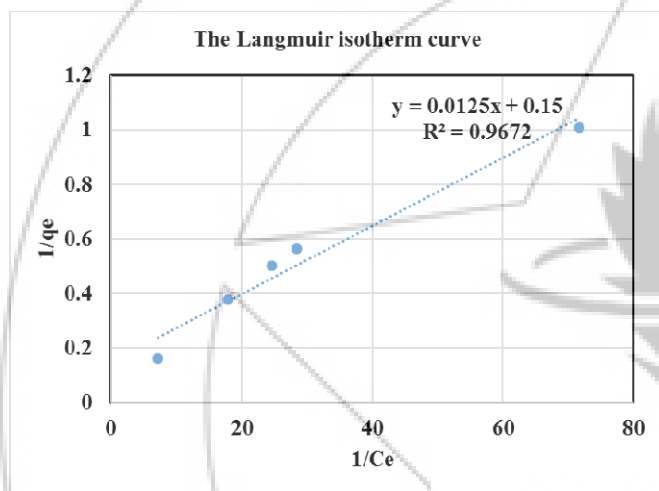


Figure 5: The Langmuir isotherm curve

Table 2: The Langmuir and Freundlich isotherm parameters

Langmuir isotherm				Freundlich isotherm		
R_L	R^2	Q_o	K_L	K_f	n	R^2
0.0134	0.9672	6.67	12	0.016	0.807	0.9826

The Freundlich isotherm model is expressed by the following linear equation [12]

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad (0.5)$$

Where q_e is the amount of F⁻ adsorbed per unit weight of adsorbent at equilibrium (mg/g), C_e is the equilibrium concentration of F⁻ in solution after adsorption (mg/L), K_f is the empirical Freundlich constant or capacity factor (mg/g) and $1/n$ is the Freundlich exponent. Values for R^2 for Freundlich (Figure 6) indicate the applicability of this isotherm in this study.

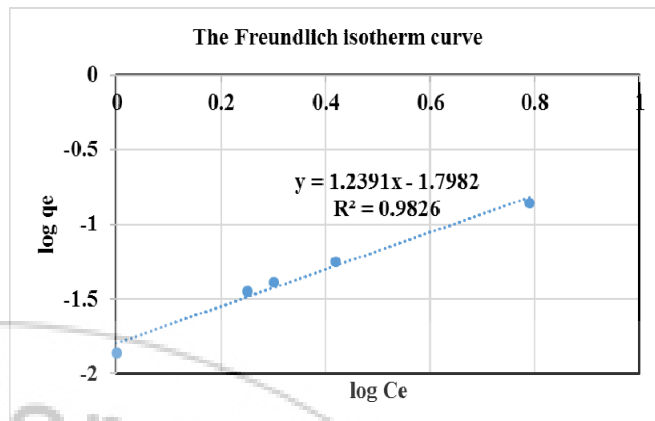


Figure 6: The Freundlich isotherm curve

4. Conclusion

This work proposes that when coconut shells activated carbon is chemically activated, provides better F⁻ adsorption hence increases its potential as a naturally available and a low cost agro waste. Coupling of coconut shell activated carbon with other techniques could improve the defluoridation efficiency and colour removal thus reducing treatment costs, the use of properly activated coconut shells carbon of particle size increases its surface area hence increasing its fluoride adsorption. The colour adsorption property appeals a dual value of coconut shells activated carbon for water treatment, the efficiency of coconut shells activated carbon in F⁻ uptake was not to a large extent at the mercy of competing ions. It is envisaged during this study that a properly activated coconut shells are suitable adsorbents for defluoridation that could be effective in rural areas of Tanzania due to its local availability and the possibility of processing the material locally.

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