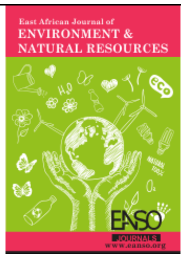
East African Journal of Environment and Natural Resources, Volume 8, Issue 1, 2025
Article DOI: https://doi.org/10.37284/eajenr.8.1.2921



East African Journal of Environment and Natural Resources
eajenr.eanso.org
Volume 8, Issue 1, 2025
Print ISSN: 2707-4234 | Online ISSN: 2707-4242
Title DOI: https://doi.org/10.37284/2707-4242

EAST AFRICAN NATURE & SCIENCE ORGANIZATION

Original Article

Effectiveness and Efficiency of Eggshells and Tamarindus indica (Tamarind) Seeds as Absorbents in Removal of Fluoride Ions from Water at Different PH Levels

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Article DOI: https://doi.org/10.37284/eajenr.8.1.2921

Date Published:

28 April 2025

Keywords:

Eggshells, Tamarindus indica (Tamarind) seeds, Absorbents, Fluoride ions, PH level.

ABSTRACT

Excessive fluoride ingestion predominantly comes from drinking water sources. Information on the extent of health problems caused by fluoride is scanty. Prevalence and severity of dental and skeletal fluorosis in northern Tanzania have been reported to be higher than would be expected from ingestion of fluoride through drinking water alone. To fill in this gap, a study was conducted aiming to develop a natural adsorbent suitable for the defluoridation of drinking water by using the Jar test method. The water sample in the investigation was synthesized by mixing sodium fluoride (NaF), with deionized water and the determination of fluoride ions and data analysis was performed by using a fluoride meter and Excel computer program respectively. The adsorption study was conducted by batch experiment method in three phases, in the first phase involved the investigation of the influence of adsorbents' particle size (1.18mm, 0.6mm, 0.45mm, 0.3mm, and 0.15mm) in different pH levels (2.8, 5.6, 7, and 9.6) and it has been observed that the particle size with higher uptake capacity of sorbents was 0.6mm mesh at optimum pH of 7. The second and third phases involved checking the influence of the adsorbents' dosage, it has been observed that the higher the dosage, the great the uptake capacity of sorbents on the adsorbents would be and the effluence of physisorption in the removal of fluoride ions from water, it was revealed that the long the contact time the higher uptake of sorbents on adsorbent would be until the equilibrium attained at optimum time of 225 minutes which removed 87.2% by eggshells and 270 minutes which removed 88.6% by tamarind seeds consecutively. The adsorption isotherms show that the correlation factor, R2 at optimum conditions was greater than 0.7 an indication that the adsorption process conformed to the Freundlich isotherms, it was further revealed that the removal efficiency was higher at neutral conditions (pH of 7.3). Moreover, the removal efficiency was observed to increase with an increase in the amount of adsorbents' dosage. It was therefore recommended that the eggshells and tamarind seeds can be used for

Article DOI: https://doi.org/10.37284/eajenr.8.1.2921

defluoridation of drinking water as an alternative and cost-effective technology. Also, it was suggested that further studies should be carried out on removal efficiency under mixed adsorbents' dosage to find out the most efficient and convenient adsorbents for fluoride removal.

APA CITATION

Kingongo, H. M., Kihila, J., Mwageni, N., & Mkumbo, S. (2025). Effectiveness and Efficiency of Eggshells and Tamarindus indica (Tamarind) Seeds as Absorbents in Removal of Fluoride Ions from Water at Different PH Levels. *East African Journal of Environment and Natural Resources*, *8*(1), 300-320. https://doi.org/10.37284/eajenr.8.1.2921.

CHICAGO CITATION

Kingongo, Hamisi M., Jacob Kihila, Nicholaus Mwageni and Stalin Mkumbo. 2025. "Effectiveness and Efficiency of Eggshells and Tamarindus indica (Tamarind) Seeds as Absorbents in Removal of Fluoride Ions from Water at Different PH Levels". *East African Journal of Environment and Natural Resources* 8 (1), 300-320. https://doi.org/10.37284/eajenr.8.1.2921

HARVARD CITATION

Kingongo, H. M., Kihila, J., Mwageni, N., & Mkumbo, S. (2025) "Effectiveness and Efficiency of Eggshells and Tamarindus indica (Tamarind) Seeds as Absorbents in Removal of Fluoride Ions from Water at Different PH Levels", *East African Journal of Environment and Natural Resources*, 8 (1), pp. 300-320. doi: 10.37284/eajenr.8.1.2921.

IEEE CITATION

H. M., Kingongo, J., Kihila, N., Mwageni & S., Mkumbo "Effectiveness and Efficiency of Eggshells and Tamarindus indica (Tamarind) Seeds as Absorbents in Removal of Fluoride Ions from Water at Different PH Levels", *EAJENR*, vol. 8, no. 1, pp. 300-320, Apr. 2025. doi: 10.37284/eajenr.8.1.2921

MLA CITATION

Kingongo, Hamisi M., Jacob Kihila, Nicholaus Mwageni & Stalin Mkumbo. "Effectiveness and Efficiency of Eggshells and Tamarindus indica (Tamarind) Seeds as Absorbents in Removal of Fluoride Ions from Water at Different PH Levels". *East African Journal of Environment and Natural Resources*, Vol. 8, no. 1, Apr 2025, pp. 300-320, doi:10.37284/eajenr.8.1.2921

INTRODUCTION

Fluoride is a naturally occurring element found in the environment, making up approximately 0.06% to 0.09% of the Earth's crust. It is present in water, food, and air. While fluoride is beneficial for maintaining healthy bones and teeth, excessive consumption can lead to skeletal fluorosis, which manifests as dental mottling, joint stiffness, and severe bone deformities. According to the World Health Organisation (WHO), the optimal fluoride concentration in drinking water ranges from 1.1 mg/L to 1.8 mg/L, with an upper safety limit of 1.5 mg/L (WHO, 2006). However, fluoride levels in several Tanzanian regions range from 2 mg/L to 10 mg/L, exceeding the recommended concentration.

Fluoride removal from polluted water is critical for environmental conservation and public health. Several techniques, including reverse osmosis, coagulation, precipitation, electro-dialytic membrane technologies, and adsorption, are employed to reduce fluoride levels to the recommended standards. These methods are categorised into four primary groups: adsorption, ion exchange, precipitation, and other techniques like electrochemical defluoridation and reverse osmosis. Among these, adsorption is the most widely used due to its cost-effectiveness, simplicity, and environmental friendliness.

The development of an adsorption unit begins with selecting an appropriate adsorbent. Over 100 fluoride adsorbents have been documented, including cow bone char, aluminium hydroxides, calcite magnesia composites, and iron-impregnated granular ceramics (Bhavnagar et al., 2011). Hydroxyapatite has also been studied as a fluoride sorbent under various conditions, considering factors such as contact time, sorbent dose, particle size, pH, temperature, and the presence of competing anions (Fan et al., 2000).

In African countries, particularly Tanzania, dental and skeletal fluorosis are prevalent health concerns, especially in communities along the Great Rift Valley. Approximately 90% of the population in these areas is affected by dental fluorosis to varying

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degrees. The Tanzania Food and Drug Authority (TFDA) has identified dental fluorosis as the fifth most common nutritional disorder. Consequently, the Ministry of Health and Social Welfare is actively seeking effective defluoridation methods to reduce fluoride-related health risks (Vuhahula et al., 2008).

Cost-effective and locally available adsorbents like tamarind seed powder and eggshells have shown promise in fluoride removal. These materials, obtained through carbonisation, not only help reduce fluoride levels in drinking water but also minimise environmental waste. Given the toxic nature of fluoride at high concentrations, its removal from drinking water remains an essential public health priority (Kaseva, 2006).

Impact of Excessive Fluoride in Drinking Water

Fluoride has both positive and negative effects on human health. It plays a crucial role in dental health, as its presence in drinking water reduces the occurrence of dental caries. However, an increase in fluoride concentration is directly linked to a higher prevalence of dental fluorosis. In regions where fluoride levels in drinking water and food are excessive, skeletal fluorosis and an increased risk of bone fractures are significant concerns (WHO, 2006). While fluoride may be essential for animals and humans, its necessity for human health has not been definitively proven, and no specific nutritional requirement has been established. Acute fluoride poisoning occurs when a person ingests at least 1 mg of fluoride per kg of body weight (Janssen et al., 1988).

Epidemiological studies confirm that prolonged fluoride ingestion mainly affects skeletal tissues (Fitzgerald et al., 2000). At low concentrations, fluoride protects against dental caries, particularly in children. It aids tooth development and strengthens enamel, with optimal benefits seen at fluoride concentrations of approximately 0.5–2 mg/l in drinking water. However, exposure to fluoride levels of 0.9–1.2 mg/l can cause mild dental fluorosis, with a prevalence of 12–33% (Dean, 1942). In cooler climates, dental fluorosis is uncommon below concentrations of 1.5–2 mg/l, whereas in warmer regions, where people consume more water, fluorosis can occur at lower levels (WHO, 1984; Cao et al., 1992). Severe skeletal fluorosis arises at fluoride concentrations between 3–6 mg/l, with crippling skeletal fluorosis typically developing when concentrations exceed 10 mg/l (WHO, 1984). The U.S. EPA (1985) has set 4 mg/l as the threshold for preventing severe skeletal fluorosis.

Fluoride Measurement and Occurrence

Fluoride levels in water are measured using ion-selective electrodes, which detect free and complex-bound fluoride. The method is effective for concentrations above 20 μg/l (Slooff et al., 1988). Fluoride occurs naturally due to volcanic activity, geothermal waters, and specific mineral deposits. High fluoride groundwater is commonly found in parts of Iraq, Iran, Syria, Turkey, North Africa, and the East African Rift Valley. In Kenya, Tanzania, and South Africa, fluoride concentrations in groundwater can reach hazardous levels, sometimes exceeding 40 mg/l (Dahi, 2006; Masamba et al., 2008).

Methods of Defluoridation

The removal of fluoride from drinking water has been extensively researched worldwide using various materials. However, the primary mechanisms remain adsorption, ion exchange, precipitation, coagulation, membrane processes, distillation, and electrolysis (George et al., 2010). Defluoridation is a widely tested and conventional method to ensure safe drinking water for communities affected by fluorosis. This process involves lowering the fluoride concentration in drinking water to an optimal level (Hiremath, 2006). Numerous materials and techniques have been explored for water defluoridation, which can be broadly classified into four main categories: adsorption, ion-exchange, precipitation, and other

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techniques such as electrochemical defluoridation and reverse osmosis (Vasudevan et al., 2008).

Adsorption-Based Defluoridation Techniques

Adsorption is a widely used method that involves fluoride ions adhering to the surface of an active agent. Several adsorbents have been tested, including activated alumina, activated carbon, and bone char.

Activated Alumina

The use of activated alumina for domestic defluoridation was promoted by UNICEF in rural India (Hiremath, 2006). The process utilizes aluminum hydroxide, which can be obtained from rejected industrial batches (Vasudevan et al., 2008). A successful community defluoridation plant using activated alumina was first implemented in Bartlet, Texas, USA, in 1952 (Yegnaraman, 2002). However, adsorption is effective only within a specific pH range, necessitating pre- and post-treatment adjustments. Frequent regeneration is required, making the method costly. Additionally, the regeneration process produces concentrated fluoride waste, posing disposal challenges. Over multiple usage-regeneration cycles, the efficiency of activated alumina declines (Yegnaraman, 2002).

Bone Char

Bone char removes fluoride through ion exchange and adsorption, where fluoride interacts with carbonate in apatite-based bone char (Larsen, 2002). The efficiency of fluoride removal is influenced by temperature, pH, and contact time. Maximum fluoride adsorption occurs at 25°C, 35°C, and 45°C, with adsorption values of 21.1, 22.4, and 25.7 μmol per gram of bone char, respectively. The optimal adsorption time is nine hours, and the ideal pH range is 7.00 to 7.50. Calcium in raw water precipitates fluoride, enhancing defluoridation. This method is cost-effective, with an efficiency of 62-66% (Bryan et al., 1993). However, bone char can harbour bacteria, requiring proper maintenance. Additionally, the charring process is crucial for efficacy, and its use may be met with cultural and religious resistance (Pearce et al., 2002).

Alternative Adsorption Media

Brick Pieces Column: Brick pieces function similarly to activated alumina, as aluminum oxide in the bricks becomes activated during kiln burning. This method requires filter media replacement every three months for water with a fluoride concentration of 2.50 mg/L (Mariappan et al., 2011).

Mud Pots: Heat-treated mud pots exhibit minor fluoride adsorption, reducing levels from 1.8 ppm to 1.4 ppm over four days. However, this method significantly increases pH beyond acceptable limits, making it impractical for defluoridation (Pearce et al., 2002).

Natural Adsorbents: Various plant-based materials, including drumstick tree seeds, vetiver grass roots, and tamarind seeds, have been tested for defluoridation. While some studies show promising results, large quantities are often required for effective treatment (Subramanian, 2006; Murugan & Subramanian, 2006).

Ion-Exchange Defluoridation

Synthetic anion and cation exchange resins, such as Polyanion (NCL), Tul-sion A-27, and Amberlite IRA-400, have been employed for fluoride removal (Bahena et al., 2006). Fluoride removal efficiency depends on the fluoride-to-total-anion ratio in water. The presence of sulfates and bicarbonates can reduce resin effectiveness (Popot et al., 1993). While these resins can be regenerated using sodium chloride solutions, the process produces large volumes of waste, making it expensive and complex (Wiatrowsky et al., 2002).

Defluoridation by Precipitation Technique

Ion-exchange and adsorption methods for defluoridation have two significant limitations: they require a continuous flow system, which is

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challenging in areas without a piped water supply, and detecting the depletion of the active agent is not straightforward. To address these issues, precipitation techniques have been developed (Caries, 1992). These methods involve adding chemical coagulants and coagulant aids, leading to the precipitation of fluoride as an insoluble compound, such as fluorapatite (Fitzgerald et al., 2000). The removal process includes separating the solid fluoride compounds from the liquid. Commonly used chemicals for defluoridation through precipitation include aluminium salts (such as alum), lime, poly aluminium chloride, poly aluminium hydroxy sulphate, and brushite (Fitzgerald et al., 2000).

Nalgonda Technique

The National Environmental Engineering Research Institute (NEERI), Nagpur, developed an economical and straightforward defluoridation method known as the Nalgonda Technique (Nawlakhe et al., 1974). This method involves adding aluminium salts, lime, and bleaching powder, followed by rapid mixing, flocculation, sedimentation, filtration, and disinfection. Aluminium salts, used as aluminium sulphate (alum) or aluminium chloride, are responsible for fluoride removal. The required dosage increases with the fluoride and alkalinity levels of the raw water. The choice between aluminium sulphate and aluminium chloride depends on the sulphate and chloride content of the raw water to maintain permissible limits (Fejerskov et al., 1996). Lime enhances the formation of dense flocs for effective settling, with an empirically determined dose of one-twentieth of the aluminium salt dose. Bleaching powder is used at a concentration of 3 mg/l for disinfection.

The Nalgonda Technique is widely used due to its affordability and ease of application (Bulusu et al., 1979). It is adaptable for large communities, small-scale community defluoridation, rural water supply, and domestic use.

Mechanisms for Different Applications

For large communities, the process involves multiple stages, including rapid mixing, chemical reaction, flocculation, filtration, disinfection, and sludge concentration for water and chemical recovery (Tewari& Jalili, 1986). For small communities (200-2000 people), a fill-and-draw defluoridation plant is recommended, utilizing a cylindrical tank with a stirring mechanism. Chemicals are added, stirred for 10 minutes, and allowed to settle for 1-2 hours before filtering and distributing the treated water (Tiwari & Prakash, 1986). Rural defluoridation plants use reaction-cum-sedimentation tanks made from materials like HDPE, Ferro-cement, or RCC. These tanks include a motorized stirring mechanism, an outlet for treated water, and a sludge disposal system (Muthu, 2003). At the domestic level, a simple 20-50L container with a tap can be used, requiring minimal investment and utilizing readily available chemicals (Tewari & Jalili, 1986).

Advantages and Disadvantages of Nalgonda Technique

The advantages of the Nalgonda technique include its simplicity, affordability, and lack of need for media regeneration. It effectively removes fluoride and other contaminants while minimizing water wastage (Tewari, & Jalili, 1986). However, drawbacks include high sludge generation, large alum requirements, and the need for careful pH control to prevent high residual aluminium levels (Herschel & Mariappan, 2002).

Alternative Defluoridation Techniques

Other defluoridation methods include reverse osmosis, electrodialysis, and electrolysis. Reverse osmosis uses hydraulic pressure to separate fluoride through a semi-permeable membrane, but it requires significant pre-treatment and is prone to fouling (Hall & Crow, 1993). Electrodialysis involves using an electric current to move ions through membranes but is energy-intensive and expensive (Kumar et al.,

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2009). Electrolysis generates aluminium hydroxide in situ for fluoride adsorption, requiring minimal chemical additions and producing less sludge, but electrode replacement is necessary (Padmapriya et al., 2003).

Review of Tamarind Seed as an Adsorbent

Fluoride removal from drinking water is conventionally achieved through methods such as ion-exchange, reverse osmosis, and adsorption. Among these, ion-exchange and reverse osmosis are costlier alternatives, making adsorption a more practical and widely used technique. Several plant-based materials have been found to effectively adsorb fluoride, including tamarind gel (Maruthamuthu & Venkatanarayana, 1987b), Duckweed *Spirodela polyrrhiza* (Shirke & Chandra, 1991), *Hydrilla verticillata* (Sinha et al., 2000), and Aloe vera (Murugan & Subramanian, 2000). However, research on biosorptive defluoridation remains limited, prompting the present study on tamarind seed.

Tamarind seed, a common household waste product after pulp extraction, consists mainly of polysaccharides (Meltzer, 1976) and is often used as cattle feed. Due to its widespread availability, this study investigates its potential as a cost-effective fluoride adsorbent. The study utilizes both laboratory-prepared aqueous solutions and groundwater samples to develop a simple and affordable defluoridation method, particularly suited for rural and urban communities.

Groundwater samples were collected from a bore well in Pallikottai village (a fluoride-prone area in Tirunelveli District) and analysed following standard protocols (APHA, 1992). The defluoridation process was evaluated using micronized tamarind seed (75-micron particle size), which exhibited favourable surface characteristics for fluoride adsorption. Experimental parameters such as agitation time, initial fluoride concentration, pH, temperature, particle size, and sorbent dosage were systematically assessed. Results indicated that equilibrium was reached within 60 minutes, with fluoride removal rates of 68.1%, 50.1%, and 41.3% for initial concentrations of 50, 60, and 70 mg/L, respectively. The adsorption process followed a monolayer coverage pattern.

Fluoride removal was most effective within the pH range of 6.0–8.0, peaking at pH 7.0. The positively charged surface at this pH facilitated greater interaction between fluoride ions and the tamarind seed surface. Temperature was found to negatively impact adsorption, with increased temperatures weakening adsorptive forces (Ajmal, 1998). Furthermore, reducing particle size from 300 to 75 microns enhanced adsorption, as smaller particles provide more surface area and sorption sites. The optimal sorbent dosage for 40 mg/L fluoride concentration was determined to be 2.2 g.

Review of Eggshells as an Adsorbent

Eggshells, composed of cellulosic structures and amino acid groups, have been studied as biosorbents (G. Kaylan et al., 2009). Their outer surface is coated with mucin protein, which facilitates fluoride adsorption. This study investigates eggshell powder combined with leaf alumina as a low-cost adsorbent.

Experimental parameters such as adsorbent dosage, initial fluoride concentration, pH, and temperature were analysed. Results demonstrated that increasing the adsorbent dosage from 0.02 to 0.1 g/100 mL improved fluoride removal, though further increases had minimal impact. Fluoride adsorption was most effective at pH 7.0, achieving over 80% removal efficiency.

Adsorption isotherms were analysed using Freundlich and Langmuir models. The Freundlich isotherm indicated heterogeneous adsorption, while the Langmuir isotherm suggested monolayer adsorption with equilibrium distribution between solid and liquid phases (Langmuir, 2006). The Langmuir separation factor (RL) indicated that fluoride adsorption by eggshells was favourable ($0 < RL < 1$).

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Problem Statement and Justification

The excessive presence of fluoride in drinking water poses serious health risks, including dental fluorosis and skeletal disorders such as crippling, tooth mottling, joint stiffness, and bone abnormalities. While various defluoridation methods exist, many are prohibitively expensive, making them impractical for widespread use (Mahvi et al., 2006). Consequently, there is a pressing need for efficient, cost-effective, and sustainable defluoridation techniques before water consumption by communities. Currently, Tanzania employs bone char as a defluoridation method. However, this technique has several drawbacks: it harbours bacteria, posing hygiene concerns; it lacks an indicator for exhaustion without regular fluoride analysis; and its use raises cultural and religious objections (Larsen & Pearce, 2002). Due to these limitations, alternative adsorption methods that are safe, environmentally friendly, cost-effective, and widely acceptable are required to replace bone char.

Adsorption efficiency depends on specific properties of the adsorbent material, including surface area, particle size, organic matter content, and bulk density. While numerous natural adsorbents have been explored for fluoride removal, tamarind seeds and eggshells have not been extensively studied, despite their potential to serve as effective and affordable alternatives. Scientifically, it is well known that adsorption depends on the physicochemical properties of materials, yet research on the inorganic mineral content of tamarind seeds and eggshells and their efficiency in fluoride removal remains limited. This study aimed to fill this gap by evaluating the potential of tamarind seeds and eggshells as natural adsorbents for defluoridation by assessing their mineral composition and adsorption capacity as well, it sought to determine whether these materials could serve as efficient, safe, and culturally acceptable alternatives to bone char, ultimately providing a viable solution for fluoride removal in drinking water.

MATERIAL AND METHODS

The natural adsorbents, tamarind seeds and eggshells, were sourced from different locations. Tamarind seeds were collected from the Kariakoo market, while eggshells were obtained from Ardhi University cafeterias. To prepare fluoride-containing water for the study, a solution was synthesized in the laboratory using distilled water and a standardized concentration of fluoride from sodium fluoride (molar mass: 41.98871 g/mol).

Various laboratory instruments were utilized to conduct the experiment. A pH meter was used to measure the pH of the solutions, while an oven was employed for drying the adsorbent materials. A mortar and pestle were used for grinding the adsorbents into different particle sizes. Other essential equipment included a funnel, beakers, a stirring machine, a sieve shaker, and sieving plates of different mesh sizes: 1.18 mm, 600 µm, 450 µm, 300 µm, and 150 µm. The collected data were analysed using descriptive statistical methods, including histograms and mean values. Microsoft Excel was used to manage and process the data efficiently.

Materials Preparation

Tamarind Seed Preparation

Tamarind seeds were sourced from Kariakoo Market, thoroughly washed with water, and sun-dried for three days. To further reduce moisture content, they were taken to the Soil Department at the Ministry of Water Authority in Ubungo, where they were oven-dried at 100°C for six hours. This heat treatment was essential in removing moisture, preventing biological activity, and eliminating any additional taste in the adsorbent as indicated in Plate 1.

Plate 1: (Heat Treatment of Tamarind Seeds at 100°C to Remove Moisture Content)

Once dried, the tamarind seeds were cooled to room temperature, ground using a mortar and pestle, and then sieved into different particle sizes ranging from 150 micrometres to 1.18 mm. The sieving process was conducted using an automatic sieve shaker for five minutes. The prepared adsorbents were then transferred to Ardhi University's Laboratory for use in fluoride ion removal experiments, Plate 2: Ground tamarind seeds and sieving process at the Soil Department, Ministry of Water Authority.

Plate 2: (Ground Tamarind Seeds and Sieving Process at the Soil Department, Ministry of Water Authority)

Source: Field Work, 2016

Eggshell Preparation

Eggshells were collected from Ardhi University's cafeterias and left to dry under sunlight for one day. They were then transported to the Soil Department's laboratory, where they underwent further drying in an oven at 100°C for five minutes. After drying, the eggshells were cooled at ambient temperature for ten minutes before being ground into fine particles. The ground eggshells were then sieved into different particle sizes, ranging from 150 micrometres to 1.18 mm, using a sieve shaker for five minutes. This ensured uniform particle size distribution, which is crucial for adsorption efficiency. The separated particles were retained on sieving plates after shaking and prepared for defluoridation experiments, Plate 3: Preparation of eggshell adsorbents for defluoridation at different particle sizes.

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Plate 3: (Preparation of Eggshell Adsorbents for Defluoridation at Different Particle Sizes)

Source: Field Work, 2016

Batch Experiment Studies

Batch experiments were conducted in three phases to systematically evaluate the effectiveness of tamarind seeds and eggshells as adsorbents for fluoride removal from water. Each phase focused on different adsorption parameters, including particle size, pH, dosage, and contact time, to determine the optimal conditions for maximum fluoride removal.

First Batch Effect of Particle Size and pH

The first phase examined how the adsorbents' particle size and pH influenced fluoride ion removal. Experiments were conducted at room temperature (25°C) using 100 mL beakers, where 25 mL of fluoride solution with an initial fluoride ion concentration of 12.6 mg/L (prepared from sodium fluoride) was placed in each beaker. Various particle sizes of tamarind seeds and eggshells (1.18 mm, 600 µm, 450 µm, 300 µm, and 150 µm) were tested, with 5 grams of each adsorbent added to the solution. The pH of the solution was adjusted from acidic (pH 2) to alkaline (pH 10) using NaOH and HNO_3. The solution was stirred at 500 rpm for 45 minutes using a magnetic stirrer to ensure proper mixing. After the experiment, the treated water was filtered using filter paper, and the fluoride ion concentration was analysed using a fluoride meter. The particle size that exhibited the highest fluoride removal efficiency was selected for further study in the second phase.

Second Batch Effect of Adsorbent Dosage and pH

In this batch, the most effective particle size (600 µm) identified from the first batch was used to investigate the influence of adsorbent dosage and pH on fluoride removal. Different masses of the adsorbent (0.5, 1, 1.5, 2, 2.5, 3, and 3.5 grams) were added to the fluoride solution to determine the optimal dosage required for efficient adsorption. The pH of the solution was adjusted using NaOH and HNO_3 to acidic (pH 5.2), neutral (pH 7.3), and alkaline (pH 11.8) conditions. Similar to the first batch, the solution was stirred at 500 rpm for 45 minutes to ensure an even distribution of adsorbent particles. After stirring, the treated water was filtered, and the fluoride ion concentration was measured using a fluoride meter. This batch assisted in determining the optimum mass and pH for effective fluoride removal.

Third Batch Effect of Contact Time

It aimed to analyze the impact of contact time on fluoride ion removal. Based on the results from the second batch, the optimum mass (2.5 grams) and pH (7.3) were selected for further evaluation. A fluoride solution with an initial concentration of 10.7 mg/L was placed in a 500 mL beaker, and the mixture was stirred at 500 rpm using a magnetic stirrer. To assess how contact time influences fluoride removal, 25 mL of treated water were sampled at different time intervals: 45 min, 90 min, 135 min, 180 min, 225 min, and 270 min. After each interval, the sample was filtered and analysed for fluoride concentration using a fluoride meter.

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Significance of the Batch Studies

These batch studies were crucial in identifying the most effective conditions for fluoride removal using tamarind seeds and eggshells. Phase one determined the ideal particle size for maximum adsorption, phase two established the optimal adsorbent dosage and pH, and phase three evaluated the required contact time for efficient fluoride removal. By systematically examining these parameters, the study aimed to develop a cost-effective and environmentally friendly defluoridation method that could serve as an alternative to existing techniques

Data Analysis

Fluoride Measurement

A fluoride meter was used to determine the concentration of fluoride ions in water. The probe of the fluoride meter was immersed into the sample containing fluoride ions, and the concentration was displayed on the meter. This method ensured accurate measurement of fluoride levels before and after treatment.

Calculation of Removal Efficiency

The fluoride concentration was expressed in mg/L (ppm), and the percentage removal efficiency was calculated using the following formula:

The removal efficiency (%) = $\frac{A-B}{A} \times 100\%$ $\frac{A-B}{A} \times 100\%$; Where, A = Initial concentration, (mg/L); B = Final concentration, (mg/L)

Where:

- A = Initial fluoride concentration (mg/L)
- B = Final fluoride concentration (mg/L)

Organic Matter Analysis

The organic matter content of tamarind seeds and eggshells was determined using a Vecstar furnace. First, the samples were dried in an oven at 105°C for 24 hours. A small portion of each dried sample was placed in a crucible and weighed (M_1). The crucible was then placed in a furnace set at 400°C for 24 hours. After cooling, the sample was weighed again to obtain M_2. The organic matter content (M_3) was calculated as the difference between the initial and final mass using the formula:

$$\%M3 = \frac{M1 - M2}{M1} \times 100\%$$

Organic Matter Content Results

As shown in **Table 1**, tamarind seeds had an organic matter content of 44.4%, while **Table 2** shows that eggshells had an organic matter content of 36%.

Table 1: Organic Matter Content of Tamarind Seeds

M_1 (Mass at 105°C)	M_2 (Mass at 400°C)	Organic Matter Content (%)
10 g	5.56 g	44.4%

Table 2: Organic Matter Content of Eggshells

M_1 (Mass at 105°C)	M_2 (Mass at 400°C)	Organic Matter Content (%)
10 g	6.4 g	36%

These results suggested that the increased organic matter concentration of tamarind seeds compared to eggshells may affect their adsorption capabilities.

Bulk Density Determination

The bulk density of tamarind seeds and eggshells was measured by wetting the samples and recording the mass of a beaker containing wetted tamarind seeds (M_1) and another with wetted eggshells (M_1).

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The initial volume (V_1) was also noted. After 24 hours, the mass (M_2) and volume (V_2) were recorded again. Bulk density was then calculated using these values. In this study, bulk density was essential to be determined for assessing the adsorbents' packing ability, porosity, and overall efficiency in fluoride removal, as higher bulk-density materials may enhance adsorption performance and water filtration capabilities.

$$D = \frac{M2 - M1}{V2 - V1}$$

RESULTS AND DISCUSSION

The results demonstrated that increasing the amount of adsorbents, varying pH levels, adjusting particle sizes, and extending contact time enhanced the fluoride ion removal efficiency using eggshells. Under optimal conditions, with a particle size of 0.6 mm, and 5 g of adsorbent at a neutral pH of 7, a maximum fluoride ion removal efficiency of 46.2% was achieved within 45 minutes. Similarly, tamarind seeds under the same conditions exhibited a slightly higher removal efficiency of 48.4%. The adsorption isotherm study indicated that at an optimum pH of 11.8, the correlation coefficient (R^2) was greater than 0.9, confirming that the adsorption followed the Freundlich isotherm model, suggesting a multi-layered adsorption process dominated by physisorption.

Efficiency of Eggshells and Tamarind Seeds in Fluoride Ion Removal from Drinking Water

The efficiency of eggshells and tamarind seeds as adsorbents was examined under different parameters, including particle size, contact time (physisorption), adsorbent dosage, and pH. The results are discussed in the following sections.

Effect of Adsorbent Particle Size on Fluoride Ion Removal

Particle sizes were obtained through a sieving process, ranging from 0.15 mm to 1.18 mm. These particle sizes were used in synthesized water samples containing an initial fluoride concentration of 12.6 mg/L. Each particle size was tested with a 5 g adsorbent dosage. Figures 4 and 5 illustrate the influence of particle size at different pH levels.

Figure 4: (Influence of Eggshell Particle Size on Fluoride Ion Removal)

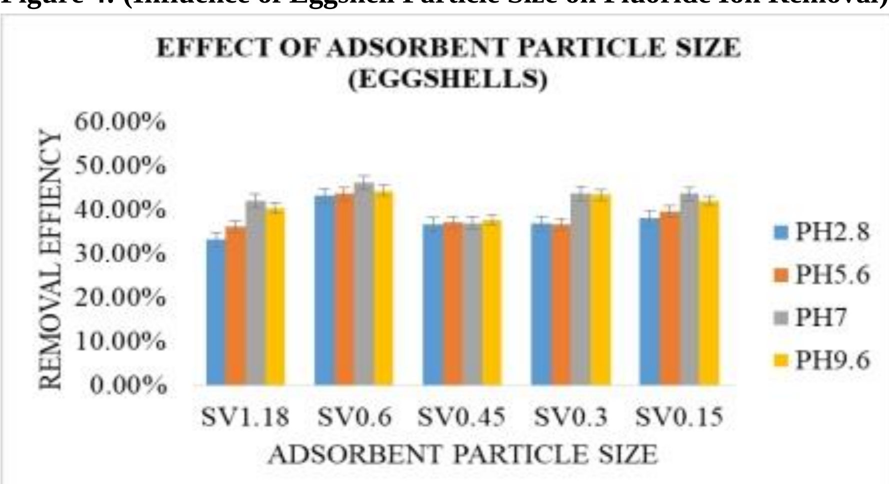

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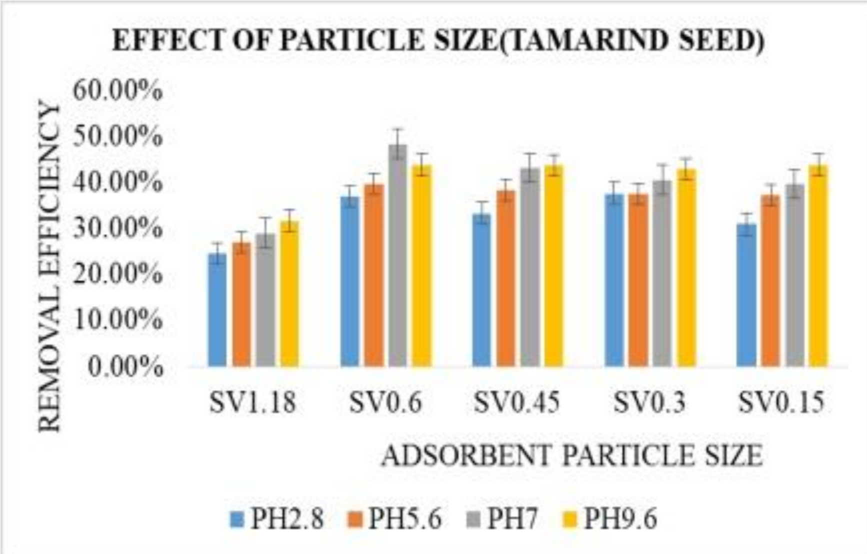
Figure 5: (Influence of Tamarind Seed Particle Size on Fluoride Ion Removal at Different pH Levels)

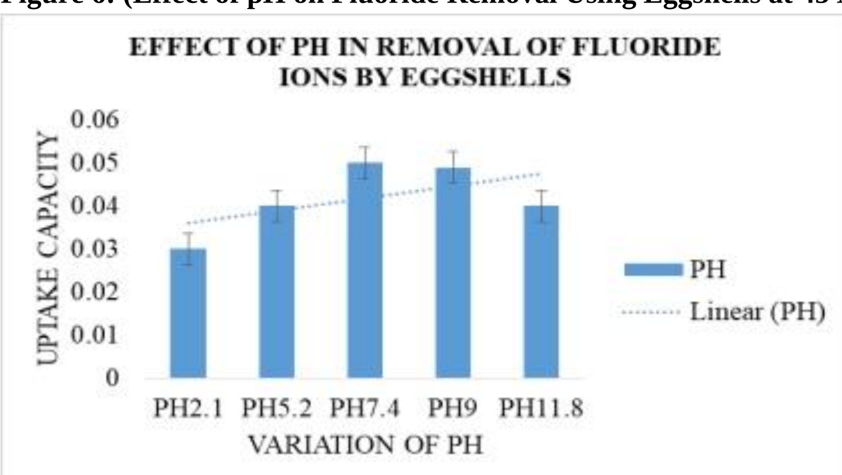


As shown in Figures 4 and 5, smaller particle sizes (0.6 mm) demonstrated higher fluoride removal efficiency compared to larger particles (1.18 mm). According to Kaseva (2006), smaller particles possess a higher defluoridation capacity due to increased surface area, which enhances the adsorption process. Similarly, Mjengera (1988) reported that finer particles exhibit better uptake efficiency due to their larger surface area-to-volume ratio. Additionally, Hauge et al. (1994) highlighted that an increase in surface area improves the efficiency of fluoride adsorption due to enhanced flow rate through the adsorbent material.

Effect of pH on Fluoride Ion Removal Using Eggshells and Tamarind Seeds

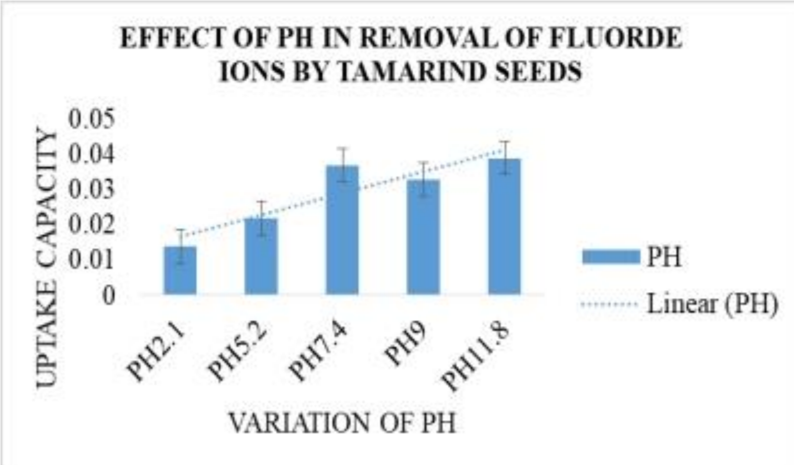
The impact of pH on fluoride removal was analysed in synthesized water with an initial fluoride concentration of 5.36 mg/L within 45 minutes of contact time. The findings are presented in Figures 6 and 7.

Figure 6: (Effect of pH on Fluoride Removal Using Eggshells at 45 Minutes)



Article DOI: https://doi.org/10.37284/eajenr.8.1.2921

Figure 7: (Influence of pH on Fluoride Removal Using Tamarind Seeds at 45 Minutes)



According to Manwar et al. (2015), the fluoride removal efficiency of adsorbents such as eggshells increases as pH rises from 3 to 7, indicating that protonation of the adsorbent surface in acidic conditions enhances fluoride ion attraction. The study found that at pH 7, fluoride removal efficiency exceeded 80%, aligning with findings by Manwar et al. (2015), which demonstrated that optimal adsorption occurs in neutral conditions due to increased electrostatic interactions between the positively charged adsorbent surface and negatively charged fluoride ions.

Effect of Adsorbent Mass on Fluoride Ion Removal

The effect of adsorbent dosage was studied using synthesized water samples with an initial fluoride concentration of 10.6 mg/L. The results are shown in Figures 8 and 9.

Figure 8: (Effect of Adsorbent Dosage on Fluoride Removal Using Tamarind Seeds at 45 Minutes)

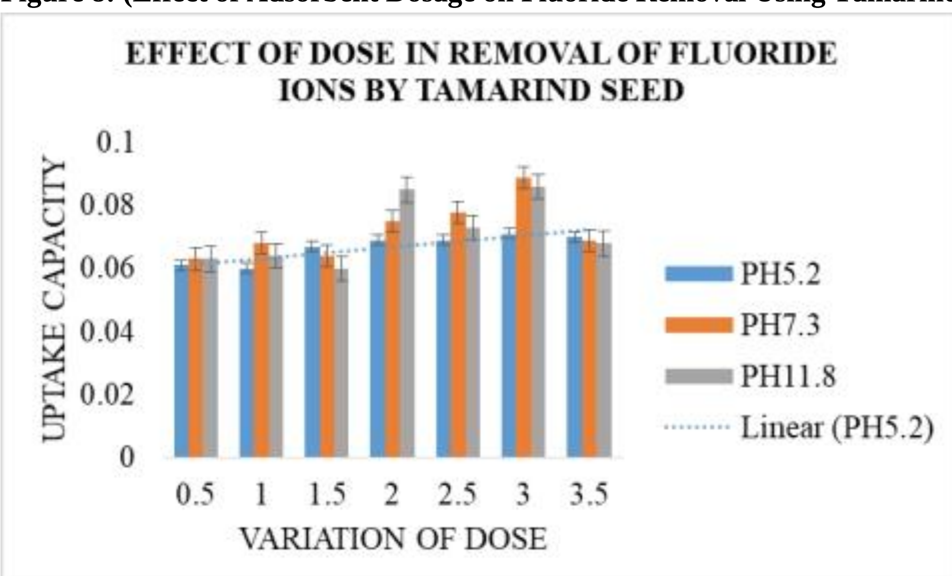


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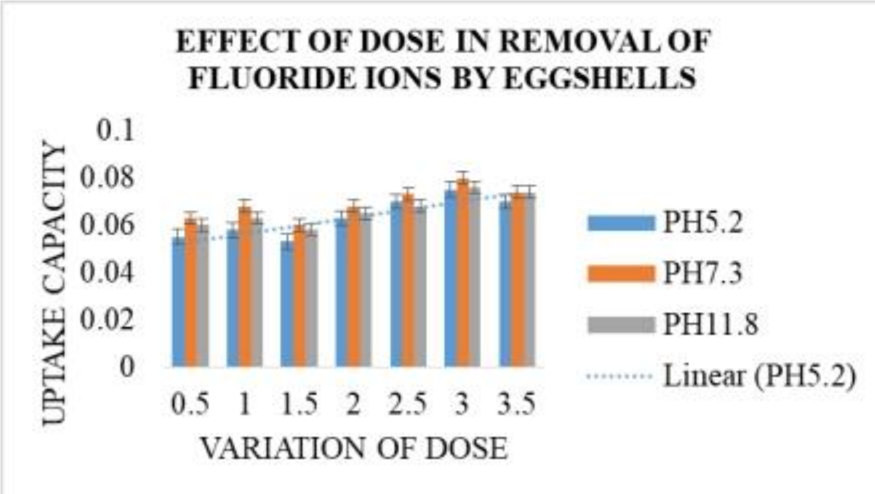
Figure 9: (Effect of Adsorbent Dosage on Fluoride Removal Using Eggshells at 45 Minutes)



Figures 8 and 9 depict the influence of varying adsorbent dosages (0.5 g to 5 g) at different pH levels (5.2, 7.3, and 11.8) within 45 minutes. The highest removal percentages recorded were 33.1% for eggshells and 30.5% for tamarind seeds. The study confirmed that fluoride removal efficiency increased with a higher adsorbent dosage, consistent with Kaseva (2006), who found that fluoride removal depends on the amount of adsorbent material used.

Effect of Contact Time on Fluoride Ion Removal

The impact of contact time was analysed over a period of 45 to 405 minutes using synthesized water with an initial fluoride concentration of 9.6 mg/L. The results are presented in Figures 10 and 11.

Figure 10: (Effect of Contact Time on Fluoride Removal Using Eggshells)

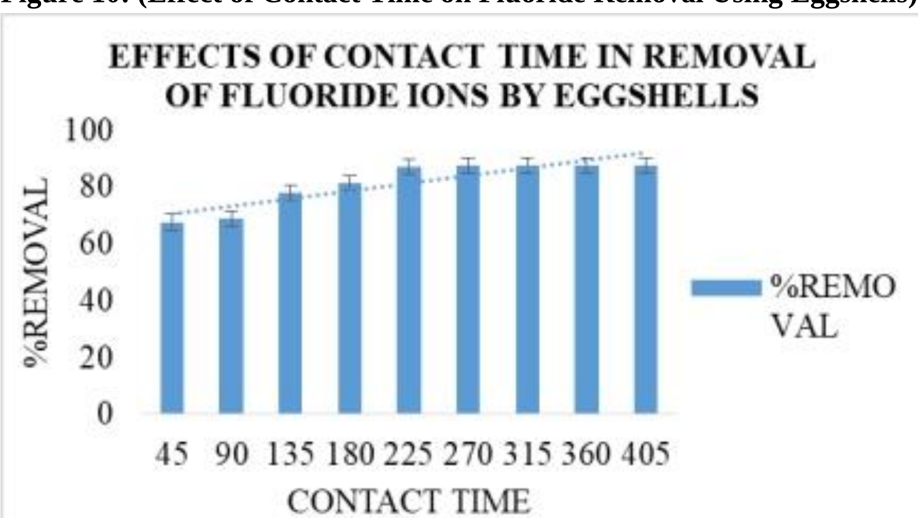


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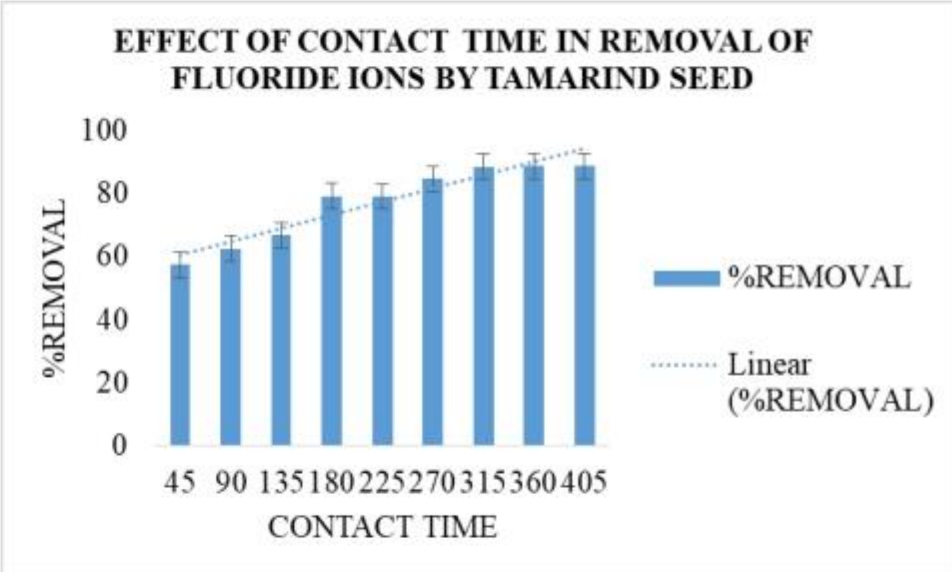
Figure 11: (Effect of Contact Time on Fluoride Removal Using Tamarind Seeds)



Figures 10 and 11 illustrate that the highest fluoride removal efficiencies, 87.2% for eggshells and 88.8% for tamarind seeds, occurred within 180 to 405 minutes. The initial rapid increase in fluoride removal slowed over time as adsorption sites became saturated. According to Henrik et al. (1995), fluoride ions initially bind quickly to active adsorption sites, but once equilibrium is reached, the adsorption rate decreases.

Freundlich Isotherm

The experiments were carried out to assess the adsorption isotherm of fluoride ions using eggshells and tamarind seeds as adsorbents. A 5g mass of each adsorbent was added to 25 mL of synthetic water with a pH of 11.8, and the initial fluoride concentration was 9.6 mg/L, as presented in Table 3.

Adsorption Isortherms

Table 3: Freundlich Models Constants and Correlation Coefficients for Removal of Fluoride Ions from Water by Eggshells and Tamarind Seed Adsorbents.

Freundlich models constants and correlation coefficients at a pH of 11.8	Eggshells adsorbents	Tamarind seed adsorbents
K	0.04424	0.05902
1/n	-0.2577	-3.141
R^2	0.9796	0.9523

The final fluoride concentrations after adsorption, denoted as Ce or Ct, are also shown in these tables. The Freundlich model was applied to analyze the adsorption isotherm, based on the assumption of heterogeneous surface energies. This model considers the varying effects of different adsorbent dosages of eggshells and tamarind seeds, leading to distinct fluoride adsorption behaviours. The plot of Log(X/m) versus Log Ce yielded a straight line, confirming that the adsorption process follows the Freundlich equation.

$$\text{Log}\,\frac{X}{m} = \text{Log}k + \frac{1}{n}\text{Log Ce}$$

In the Freundlich equation, X/m represents the amount of fluoride ions adsorbed per unit mass of adsorbent, while Ce is the equilibrium concentration of fluoride ions in the solution.

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The parameter k is derived from the intercept, and the slope corresponds to 1/n. The Freundlich adsorption isotherms for fluoride removal by eggshells and tamarind seeds are depicted in Figures 12 and 13, respectively.

Figure 12: (The Freundlich Adsorption Isotherm for the Removal of Fluoride Ions from Water by Eggshells)

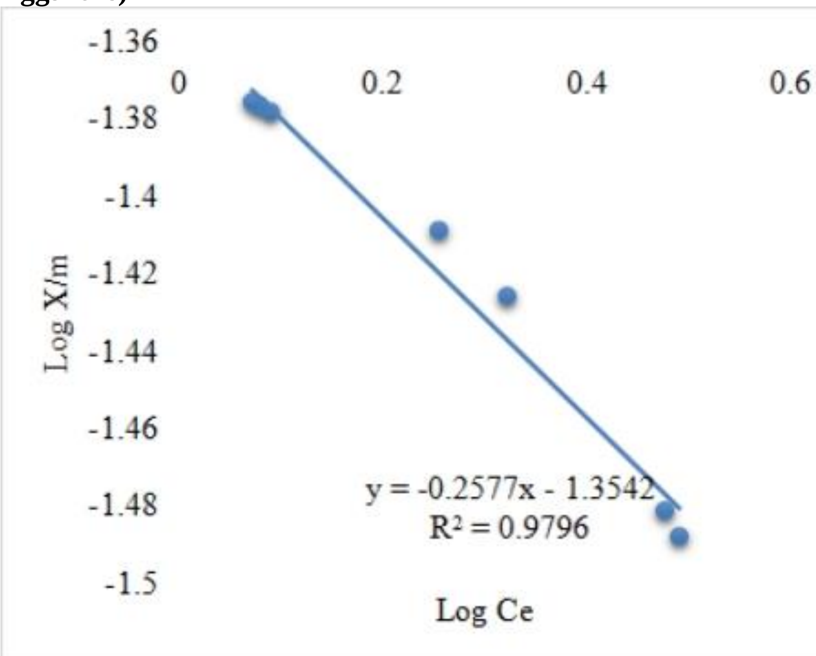
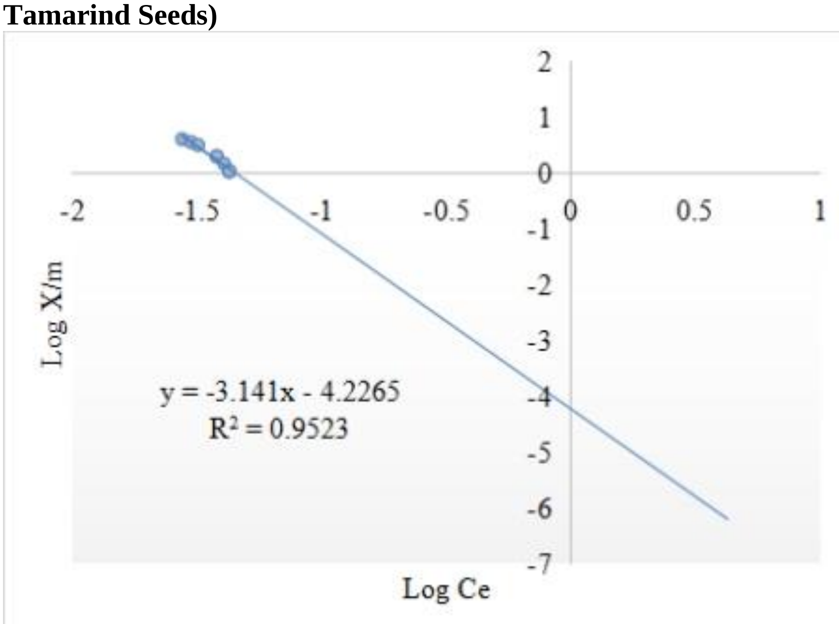


Figure 13: (The Freundlich Adsorption Isotherm for the Removal of Fluoride Ions from Water by Tamarind Seeds)



These figures demonstrate the adsorption behaviour and capacity of both adsorbents in removing fluoride ions from water, confirming that the process adheres to the Freundlich isotherm model.

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Langmuir Isotherm

Experiments were carried out to determine the adsorption isotherm of fluoride ions by using eggshells and tamarind seeds at five grams in 25 ml of synthesized water with a pH of 11.8. The initial concentration of fluoride ions was 9.6 mgF/L, and the final concentrations (Ce) of fluoride ions after being adsorbed by eggshells and tamarind seed adsorbents are shown in Table 4.

The Langmuir isotherm equation is stated as shown below:

$$\frac{x}{m} = \frac{abCe}{1 + bCe}$$

Where X/m indicates the amount of adsorbate adsorbed per unit weight of adsorbent, Ce represents the equilibrium concentration of adsorbate in the solution after the adsorption process, a is the number of moles of solute per unit weight of adsorbents to form a monolayer on the surface, and b is the empirical constant. The empirical constants in this equation can be determined by plotting 1/qe versus 1/Ce or 1/(X/m) versus 1/Ce, and these are known as the Langmuir constants.

$$Ce = \frac{1}{ab} + \frac{1}{aCe}$$

The values of Langmuir constants were determined after linearizing the equation through linear regression analysis. From the intercept, the parameter 1/a is obtained, while the slope is equal to 1/ab.

Figure 14 shows the Langmuir adsorption isotherm for the removal of fluoride ions from water by eggshells and Figure 15 shows the Langmuir adsorption isotherm for the removal of fluoride ions from water by tamarind seeds respectively.

Figure 14: (The Langmuir Adsorption Isotherm for the Removal of Fluoride Ions from Water by Eggshells)

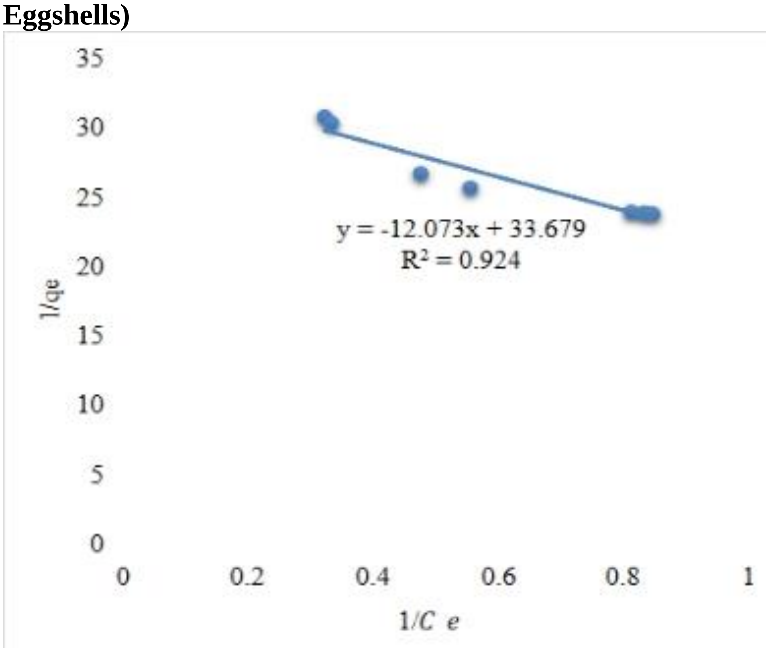

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Figure 15: (The Langmuir Adsorption Isotherm for the Removal of Fluoride Ions from Water by Tamarind Seeds)

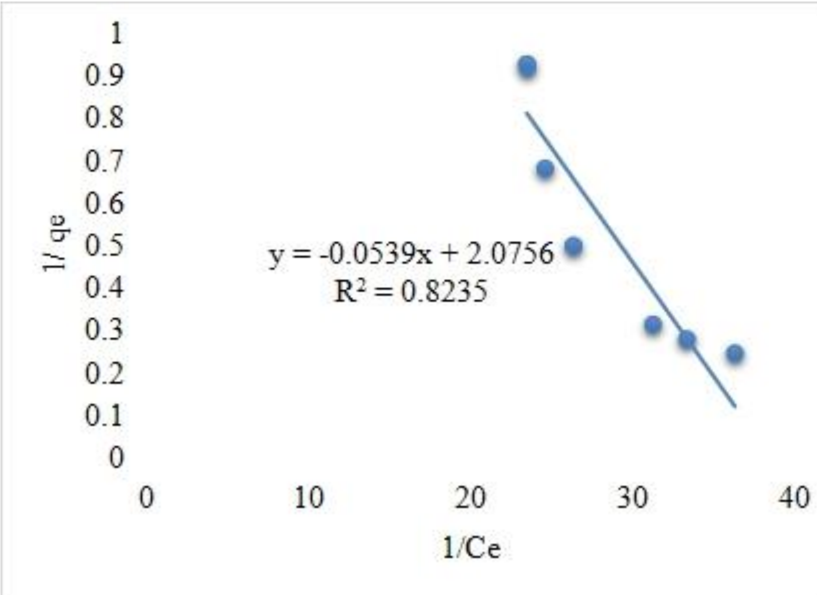

Table 4: Summarizes the Langmuir Model Constants and Correlation Coefficients for the Removal of Fluoride Ions by Eggshells and Tamarind Seeds:

Langmuir models constants and correlation coefficients	Eggshells adsorbents	Tamarind seed adsorbents
A	0.02969	0.4818
B	-2.7898	-1.8507
R^2	0.924	0.8235

Table 4 indicates that the value of b for the tamarind seeds is higher at -1.8507 compared to the eggshell adsorbent, which has a value of -2.7898. This implies that tamarind seed adsorbents possess a higher uptake capacity of fluoride ions from water compared to eggshell adsorbents, as observed in the Langmuir adsorption isotherm experiments. Also, according to Chakravarti (1974), the uptake capacity of adsorbent materials depends on the values of the Langmuir constant, b. If the value of b is high, it reflects the adsorption capacity of the media used in the adsorption process

CONCLUSION AND RECOMMENDATIONS

Conclusion

The study on the removal of fluoride ions from water using eggshells and tamarind seeds as adsorbents concluded that both materials are effective for defluoridation at around pH 7.3. For maximum fluoride ion removal of 33.1% within 45 minutes, 3 grams of eggshells in 25 ml of water containing an initial fluoride concentration of 10.6 mgF/L was required, resulting in a remaining fluoride concentration of 7.09 mgF/L. Similarly, 3 grams of tamarind seeds in 25 ml of water achieved a 30.5% fluoride removal in 45 minutes, leaving a fluoride concentration of 7.37 mgF/L. The optimal physisorption times were 225 minutes for eggshells and 270 minutes for tamarind seeds, removing up to 87.7% and 84.7% of fluoride ions, respectively, with residual fluoride concentrations of 1.23 mgF/L and 1.47 mgF/L. The experimental data matched well with both the Freundlich and Langmuir adsorption models, as indicated by correlation coefficients (R^2) greater than 0.7. The Freundlich and Langmuir isotherms showed a strong fit to the

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experimental data, with R^2 values of 0.9796 and 0.924 for eggshells, and 0.9523 and 0.8235 for tamarind seeds, respectively

Recommendations

The recommendations based on the findings of this study suggest that additional research should focus on water with higher initial fluoride concentrations to determine the optimal contact time and adsorbent loading of tamarind seeds and eggshells, aiming to reduce fluoride levels to the WHO recommended range of 1 mgF/L to 1.5 mgF/L.

Future studies should also explore the effectiveness of combining tamarind seeds and eggshells in various ratios. Given the abundance of eggshells and tamarind seeds, both in rural and urban areas, this method could potentially be implemented at the household level. Additionally, further research is needed to evaluate the performance of these adsorbents in water containing heavy metals such as copper, lead, mercury, and chromium.

Acknowledgement

This study was made possible through the support and contributions of various institutions, families, and individuals. First and foremost, the authors acknowledge the blessings and guidance of the Almighty God, whose support was present in every step of this journey. We extend our heartfelt gratitude to the government of Tanzania for providing financial assistance, which played a crucial role in the successful completion of this study.

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