



***Moringa oleifera*: a sustainable method to treat fluoride-contaminated water**

Shyla Chowdareddy, Chethan Rajesh, Rakshitha Rajashekara and Pallavi Nagaraju   *

Department of Environmental Science, School of Life Science, JSS Academy of Higher Education and Research, Mysuru 570015, India

*Corresponding author. E-mail: anupallavi@jssuni.edu.in

 PN, 0000-0001-5594-1447

ABSTRACT

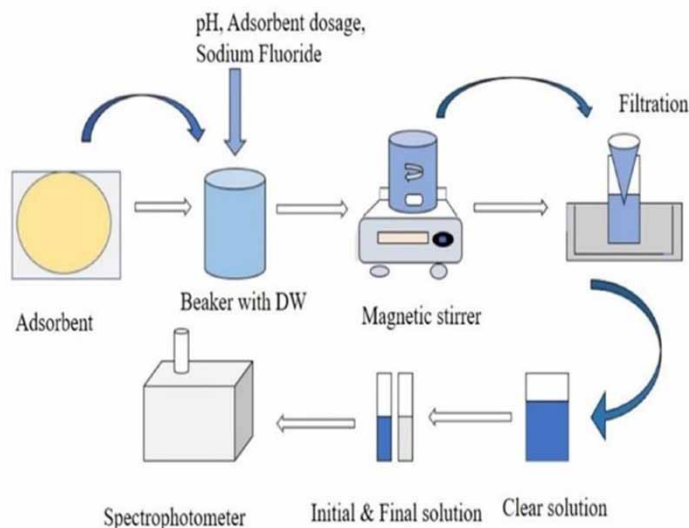
Excessive fluoride (F) is introduced into the water either by natural phenomenon or by industrial effluents. However, F in adequate amounts is necessary for sustaining good dental health. Contrarily, F concentrations in drinking water surpassing 1.5 mg/L may, nevertheless, be harmful to human health. F pollution in drinking water is a serious and challenging problem that affects people all over the world. Adsorption appears to be an effective, environmentally beneficial, and cost-effective approach to eliminating excessive F from drinking water in the present day. In this study, *Moringa oleifera* seed powder was employed as a natural adsorbent for defluoridation. The maximum removal efficiency was achieved for water with pH of 7.0, 1 mg/L of initial F concentration, contact time of 120 min, and adsorbent dose of 200 mg/L. SEM and FTIR were used to determine the structure, morphology, and other chemical composition features of the prepared *M. oleifera* seed powder. This work is anticipated to be readily implemented in real-time F elimination.

Key words: adsorption, *Moringa oleifera*, natural adsorbent, public health, water treatment

HIGHLIGHTS

- The *Moringa oleifera* seeds were collected locally to determine the adsorption capacity.
- Compared to other natural bio sorbents, *M. oleifera* seeds are edible, and there is not much health impacts.
- The *M. oleifera* seeds are dried in hot air oven at suitable temperatures.
- All parameters were analyzed before and after treatment and changes produced by the adsorbent was evaluated for each of parameters.

GRAPHICAL ABSTRACT

Moringa pods with seeds**Moringa seed powder****1. INTRODUCTION**

Higher fluoride (F) dosages in underground water have been noted in many countries, both developed and developing countries, including the USA, Africa, and Asia. It has been discovered that 17 different Indian states, particularly, Chhattisgarh, Kerala, Delhi, Haryana, Jharkhand, etc. are negatively impacted by excessive fluoride concentration (Baboo *et al.* 2022). Fluoride is an electronegative toxic element and its toxicity can cause adverse health effects for both human beings and animals (Bhaumik *et al.* 2017). World Health Organization (WHO) has already set a permissible limit of 1.5 mg/L. Fluoride ions are consumed in large quantities through drinking. Excessive fluoride in the body leads to a problem called fluorosis. This fluorosis affects the teeth, the skeleton, and the non-skeletal system (George & Tembhurkar 2019a). At concentrations above the permissible level, fluoride is a very hazardous ion and may be a factor in several health issues, including skeletal fluorosis, dental fluorosis, neurological problems, osteoporosis, parathyroid damage, Alzheimer's disease, male infertility, thyroid, kidney issues, and liver injury (Duvva *et al.* 2022).

As for purpose of eliminating too many fluoride ions from drinking water and industrial effluent, traditional treatment technologies such as precipitation, electrolysis, membranes, adsorption, ion exchange, electrodialysis, coagulation/precipitation, dialysis, reverse osmosis, nanofiltration, ultrafiltration, and others have been developed and tested (Jamwal & Slathia 2022). However, these procedures have several drawbacks, like a failed removal, an arduous treatment method, high operating costs, high energy costs, chemical usage, and the formation of hazardous sludge or other waste products, which must be disposed of again (Peng *et al.* 2017). The effectiveness of fluoride removal is always dependent on the qualities of raw water, including the original fluorine level, pH, heat, preparation time, and adsorbent dosage, other factors that affect adsorption include capacity, specificity for fluoride, renewability, price, granule and pores size, and adaptability (Mondal & Roy 2018). Therefore, the goal is to discover a practical, affordable, environmental friendly method that an average person could use to remove fluoride from drinking water.

Among all the plant materials examined over the years, a powder of *Moringa* seed, in comparison to alumina, a conventional chemical coagulant (Laney *et al.* 2020), exhibited an efficiency for water treatments. *Moringa oleifera* is known as a 'multipurpose tree' because of all these benefits. Properties of adsorption, demagnetizing, and interparticle bridging were suggested as the coagulation mechanisms of the *M. oleifera* coagulant protein. The high molecular weight polyelectrolytes are more prone to flocculation due to interparticle bridging. Recently, Khader and team reported *M. oleifera* seed beads mixed with sodium alginate for the adsorption of ions of copper, nickel, manganese, lead, and fluoride from polluted

water (Khader *et al.* 2022). Importantly, a recent review on the application of *M. oleifera* by Gomes and others highlights its potential application for the removal of emerging contaminants in water (Gomes *et al.* 2022). Several portions of *M. oleifera*, including the seeds, leaves, and bark, are promising biosorbents, according to analytical methods like scanning electronic microscopy (SEM), as they exhibit optimal surface area and porosity in their morphological structure for adsorption of pollutants (Jiraungkoorskul & Jiraungkoorskul 2016). Specifically, these seeds are lignocellulosic materials that show themselves as an ecological, cheap, and rugged alternative for water treatment. Therefore, all plant residue is considered lignocellulosic material and can be used as a biosorbent.

Hence, the current work sought to determine whether *M. oleifera* seeds could effectively adsorb fluoride from aqueous solutions under a variety of operating circumstances such as pH, adsorbent dosage, contact time, initial fluoride concentration, and temperature. The prepared adsorbents were further examined by SEM and FTIR to gain knowledge of the metal ion loading procedure and the fluoride adsorption mechanism. The ultimate goal of this effort is to create large-scale water treatment procedures that provide the general public with safe drinking water and make use of natural and eco-friendly components that are widely available.

2. MATERIALS AND METHODS

2.1. Chemicals and reagents

In the current work, analytical grades of chemicals, such as SPADNS solution, zirconyl acid reagent, sodium arsenite solution, sodium fluoride, stock, and standard solution were used. *M. oleifera* is used as a natural adsorbent.

The fluoride standard sample was made by diluting the standard fluoride solution in the range of 1–11 mg/L with the necessary volumes of distilled water. After that, 5 mL of the zirconyl acid solution and SPADNS solution were taken into each standard, and thoroughly combined. The spectrophotometer was calibrated using the standard solution and setting the value of absorbance to zero. As a blank solution, the reference solution was employed. As per the standard method procedure, a spectrophotometer with a wavelength of 540 nm was employed (Nazri *et al.* 2020).

2.2. Adsorbent preparation

For this experiment, *M. oleifera* pods were collected from local trees in villages J. V. Colony (Kolar). The seeds were then collected and dried for up to 8 h in a hot air oven at 60 °C. The seed coat was removed after collecting the seeds. The seeds of *M. oleifera* were then grounded into a fine powder, as shown in Figure 1. Then sieved to obtain particles in micro length which is then used as an adsorbent. To prevent spoilage, the *M. oleifera* powder was finally gathered and kept in an airtight container. Figure 2 depicts the schematic representation of the defluoridation of drinking water using a natural adsorbent.

2.3. Characterization of adsorbent

SEM and FTIR were utilized in this experiment to analyze the morphological structure of *M. oleifera*. To study the morphological structure, the sieved *M. oleifera* powder was taken.

A Fourier transform infrared (FTIR) *M. oleifera* seeds contain functional groups that were identified using a spectrometer. The powdered samples were collected. With 25 scans and a resolution of 16 cm⁻¹, the spectral spectrum ranged from 4,000 to 500 cm. The aim of employing this methodology was to analyze the major group function available in the material, particularly the accessibility of the principal groups implicated in adsorption activities (Gogoi & Dutta 2016). A SEM was also utilized to study the morphological properties of *M. oleifera* seeds to evaluate the material profile for absorbing fluoride ions. After collecting the samples, an electronic acceleration voltage of 5.00 kV was applied (George & Tembhurkar 2019b).

3. RESULTS AND DISCUSSION

3.1. Characterization of *M. oleifera*

Figures 3 and 4 display the findings of the examination of the physicochemical characteristics of the *M. oleifera* plant. The morphological structure of *M. oleifera* seeds was examined using SEM. Using an advanced device called an FTIR spectrophotometer, the functional groups of the created biosorbent were examined.



Figure 1 | (a) *Moringa oleifera* fruit, (b) *M. oleifera* seeds being dried in a hot air oven, (c) *M. oleifera* kernels, and (d) *M. oleifera* seed powder.

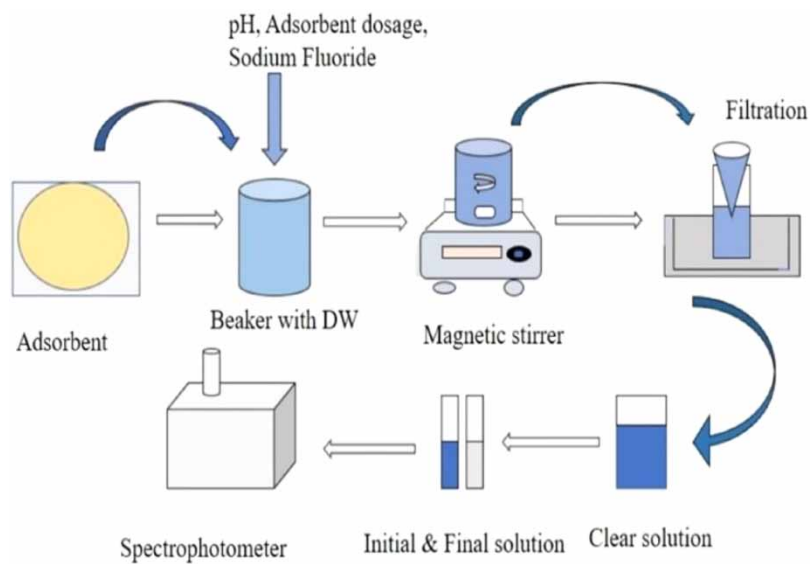


Figure 2 | Schematic representation of defluoridation of natural adsorbents.

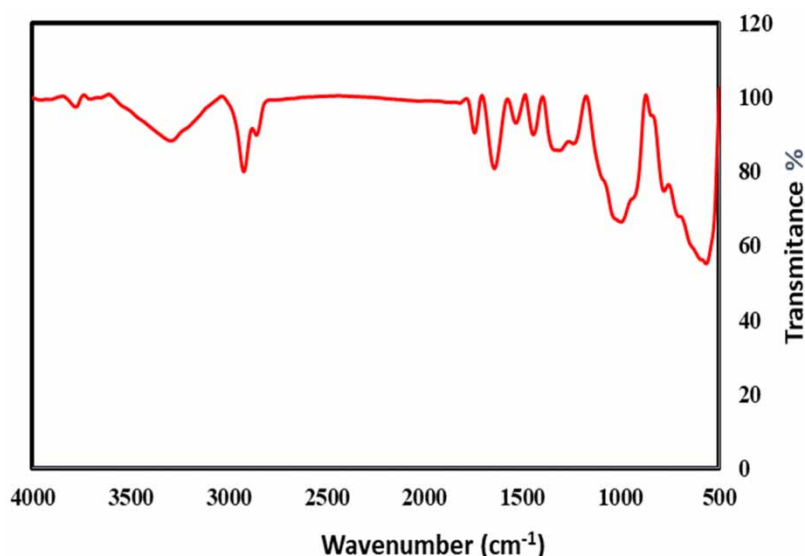


Figure 3 | FTIR spectrum of *Moringa oleifera* seed powder. The peaks represent the highest signal obtained.

3.2. FTIR analysis

The FTIR method was employed to research the functional groups discovered in *M. oleifera* seeds. This approach enables the identification of several essential functional groups capable of adsorbing metal ions. The below graph shows the FTIR spectra of *M. oleifera* seed powder.

The spectra all have an identical profile: a wide band's center is $3,953.36\text{ cm}^{-1}$ which is ascribed to O–ZH extending. This operational unit is mostly found in the proteins and fatty acid structures found in *M. oleifera* seeds. Because of the high protein composition of seeds, the N–ZH stretching of amide groups also contributes to this area. Peaks at 559.25 and 987.37 cm^{-1} are attributed to symmetric and asymmetric stretching, respectively (Manna *et al.* 2015). Three powerful bands are ascribed to the C–O bond expanding in the area between $1,315$ and $1,454\text{ cm}^{-1}$. The carbonyl group can be found in fatty acid and protein structures. This example shows the spectra indicate double groups connected with fatty acids at $1,531$ and $1,654\text{ cm}^{-1}$ and a band related to the protein's amide group at $1,754\text{ cm}^{-1}$. This band's existence validates the structure of proteins in *Moringa* seeds (Amin *et al.* 2015).

3.3. Scanning electron microscopy

SEM analysis is also another important method for analyzing an adsorbent's surface morphology. Figure 4 shows the clear structures of *M. oleifera* powder and the material's morphology depicts a fragmented and mostly porosity core (Mihayo *et al.* 2021). This morphological composition enhances fluoride ion absorption in water. It is because of the intercellular spaces and, most significantly, the proteins element of the seed. Based on these properties, it is possible to conclude that this substance has a suitable structural character for holding fluoride ions in the water sample (Li *et al.* 2016).

3.4. Adsorbent dosage

For sorbent dosages greater than 100 mg, the removal efficiency was reported. The maximum rate of removal was 80.6%. (Mupa *et al.* 2016). Experiments were conducted with concentrations ranging from 50, 100, 150, 200, 250, 300, 350, and 400 mg/L to determine the optimum dosage of adsorbent required to bring dosage levels below the permissible limit. We maintained the pH constant at 7, the initial fluoride concentration was 1 mg/L, and the time was 120 min to calculate the adsorbent dosage. Figure 5 illustrates a plot of fluoride removal effectiveness vs. adsorbent dose (Dhanasekaran *et al.* 2016).

3.5. pH

As pretests, the effect of pH on fluoride adsorption in the range of 2–11 (2, 4, 6, 7, 9, 11) was examined. To determine the effect of pH, the pretest experiments utilized an adsorbent dose of 200 mg/L, a contact time of 2 h, and an initial fluoride concentration of 1 mg/L. According to the findings, a pH of 7 is the optimal pH for *M. oleifera* seed powder to remove

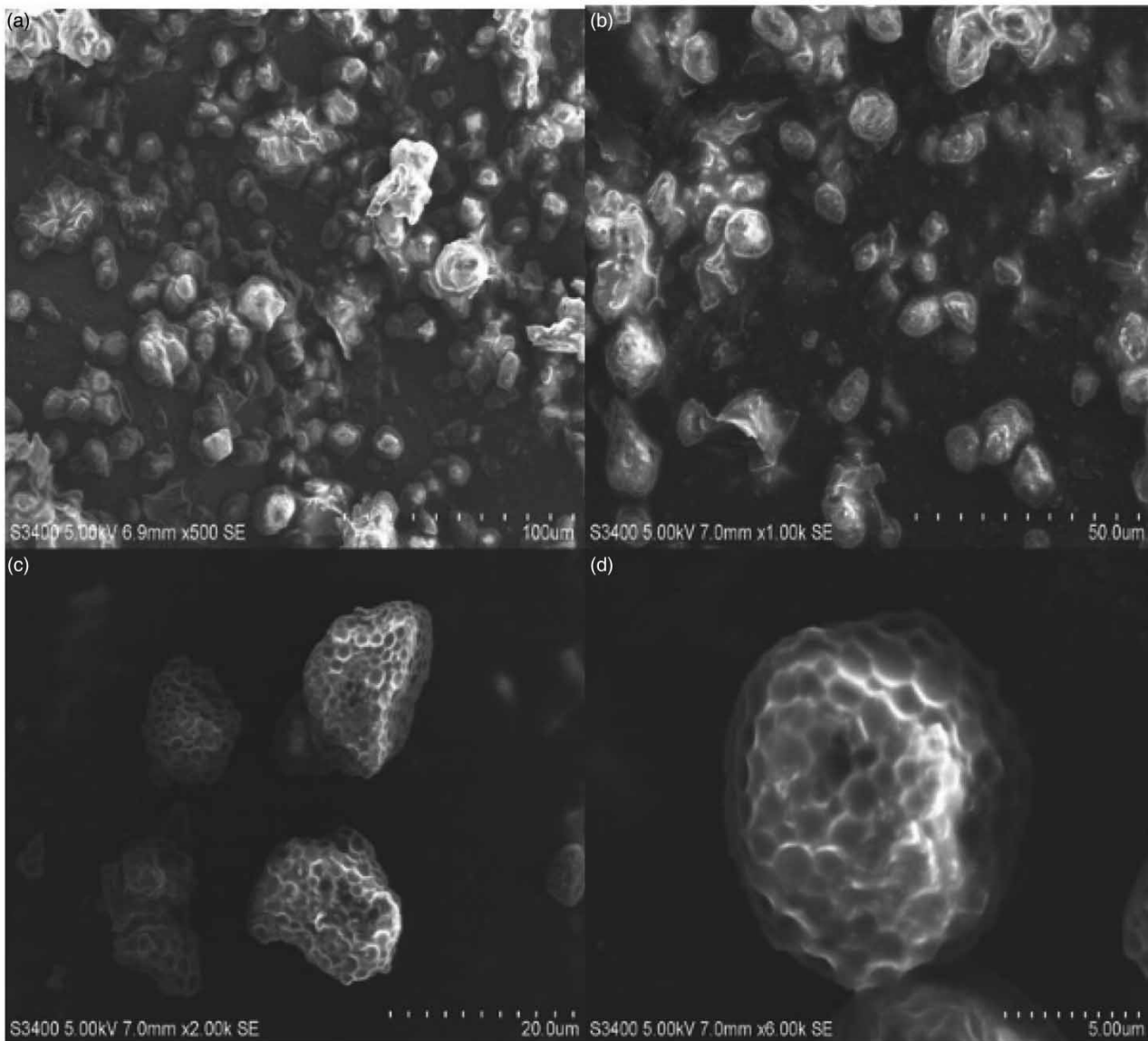


Figure 4 | (a–d) Scanning electron microscope images.

fluoride. 88.7% of fluoride was eliminated under pH 7. In each experiment, the concentration of sodium fluoride ions in the stock was measured. Stock sodium fluoride concentrations were measured after the solutions were stirred for 2 h by using a magnetic stirrer (Getachew *et al.* 2015). The elimination of fluoride by various adsorbents showed the same results. As a result, the pH value of 7 has been chosen as the best pH for all studies (Nazri *et al.* 2020). Figure 6 illustrates a plot of fluoride removal effectiveness vs. pH.

3.6. Initial fluoride concentration

Experiments were conducted to determine the effect of initial fluoride concentrations that affected *M. oleifera*'s ability to remove fluoride. Initial fluoride concentrations of 0.2, 0.4, 0.6, 0.8, 1, 1.2, 1.4, 1.6, 1.8, 2, and 2.5 mg F/L, agitation rate of 300 rpm, contact time of 120 min, pH of 7.0, and adsorbent dosage of 200 mg/L have been used in the experiments. The highest percentage of removal was 86.3% in 1 mg/L (Ismail & AbdelKareem 2015). Figure 7 illustrates a plot of fluoride removal effectiveness vs. adsorbent dose.

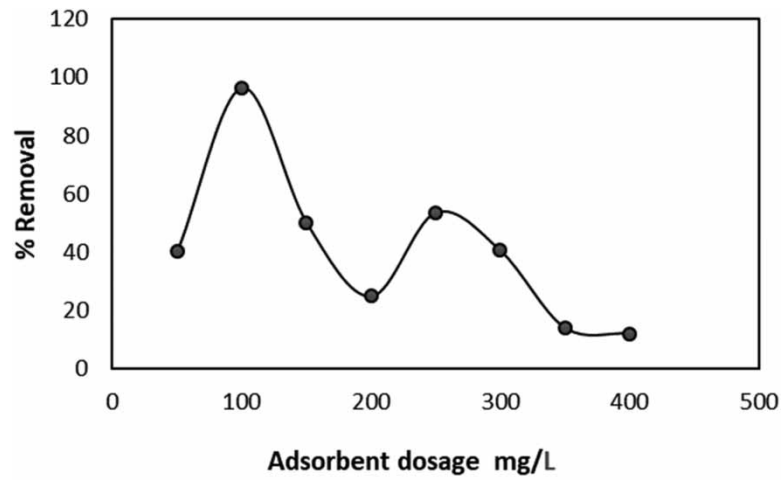


Figure 5 | Percentage of fluoride removal against adsorbent dosage.

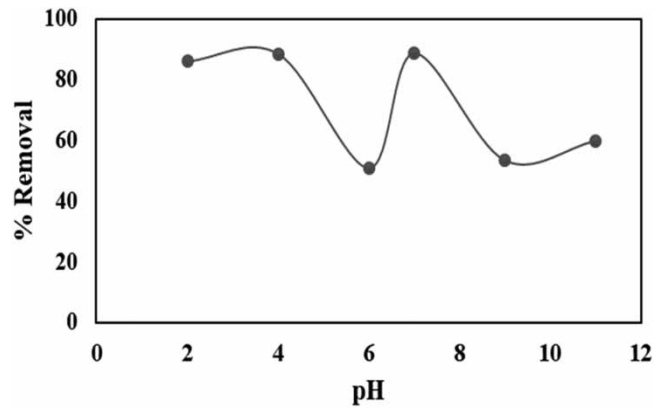


Figure 6 | Percentage of fluoride removal against pH.

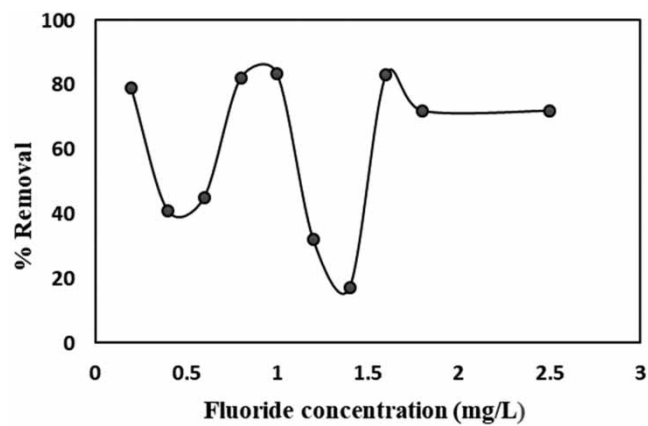


Figure 7 | Percentage of fluoride removal against fluoride concentration.

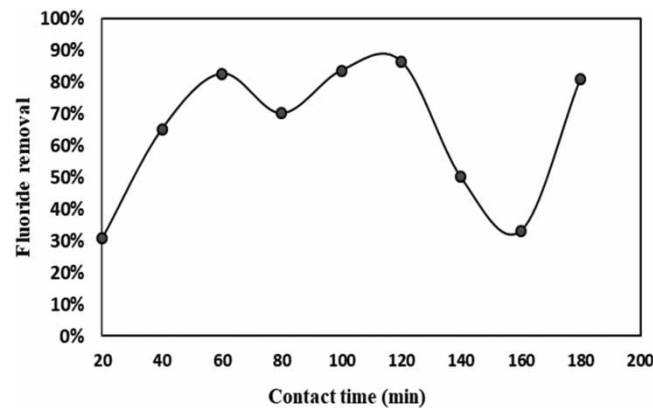


Figure 8 | Percentage of fluoride removal as a function of contact time.

3.7. Contact time

When it comes to optimizing adsorption procedures, the impact of contact time is important (Mupa *et al.* 2016). It was observed that when the contact time increased, fluoride removal increased as well. At 1 h, almost saturation was obtained, and equilibrium was reached at 2 h, with a percentage elimination of 86.6%. There was no significant fluoride removal after 2 h, thus it was considered that this was the best time for defluorination research. The mass transfer rate between the adsorbent and the aqueous medium dropped as the contact time increased, and the adsorption rate eventually decreased (Mehta *et al.* 2016). Figure 8 illustrates a graph representing fluoride elimination in the water against contact time.

4. CONCLUSION

In summary, *M. oleifera* seeds offered a unique advantage in fluoride elimination from aqueous solution in terms of its easy, inexpensive, and carbon-neutral impact. SEM and FTIR validated the internal features of the adsorbent. The primary operating parameters like pH, initial fluoride concentration, adsorbent dose, and contact time were optimized. The maximum removal efficiency was obtained at pH of 7, 1 mg/L of initial fluoride concentration with 120 min of contact time, and an adsorbent dose of 200 mg/L. Hence, this work is the potential for real-time application.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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First received 14 July 2022; accepted in revised form 22 January 2023. Available online 2 February 2023