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Comparative Study on Removal of Fluoride from Groundwater by Natural and Modified Bagasse Carbon of Sugarcane

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Authors' contributions

This work was carried out in collaboration among all authors. Author Sudhanshu Kanaujia designed the study, mobilized resources, engaged in laboratory activities and wrote the first draft of manuscript. Authors Bharat Singh and Sanjay Kumar Singh managed the analyses of the study. All authors read and approved the final manuscript.

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ABSTRACT

Aim: To design and develop a novel, cost effective strategy for fluoride removal, applicable to rural areas of developing countries.

Study Design: Laboratory analysis for different parameters.

Place and Duration of Study: Department of Chemistry, University of Allahabad and Department of Chemistry, Institute of Engineering and Technology. June 2014 to January 2015 is the time duration for this analysis.

Methodology: Batch adsorption study was conducted to determine the optimum conditions for defluoridation. The effects of pH, adsorbent dose, initial fluoride concentration, and contact time on

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adsorption were studied. Natural Bagasse Carbon (NBC) and Modified Bagasse Carbon (MBC) were used as low-cost agricultural biomass based adsorbents.

Results: The NBC and MBC removed 51.8 and 58.4% respectively from an aqueous solution of 5 mg/L fluoride at pH 6.0, with the contact period of 75 min and a dose of 4 gm/L. All adsorption experimental data followed the Freundlich isotherm. Adsorption of fluoride on all studied adsorbents from aqueous solution followed pseudo-second order reaction, and the mechanism of fluoride removal on adsorbents was found to be complex.

Conclusion: Significant amount of fluoride can be reduced in the neutral pH range by these adsorbents, which makes them suitable for utilize in drinking water treatment, especially in rural areas where minimum facilities are available.

Keywords: Fluoride removal; sugarcane bagasse; adsorption; low cost agricultural biomass.

1. INTRODUCTION

The chemical nature of water is one of the most imperative criteria that determine its usefulness for a precise need. Apart from fluoride, arsenic and nitrate are few of major water pollutants which cause large scale health issues, but in cutting-edge the most serious pollutant is fluoride [1].

According to the World Health Organization, the maximum acceptable concentration of fluoride ions in drinking water lies below 1.5 mg/L. Depending on the concentrations and the duration of fluoride intake, it could have positive effect on dental caries [2]. On the contrary, long term consumption of water containing excessive amounts of fluoride can lead to fluorosis of the teeth and bones [3]. The excessive intake of fluoride may cause dental [4] and skeletal disorders [5].

In India, this problem is common in states such as Andhra Pradesh, Tamilnadu, Karnataka, Kerala, Rajasthan, Gujarat, Uttar Pradesh, Punjab, Orissa and Jammu and Kashmir [6]. Raebareli is one of the district of Uttar Pradesh state in India. The land area of this district is covered by two rivers namely Ganga and Sai. The ground water of this district is not safe because it is contaminated with fluoride, arsenic and coliform bacteria. The fluoride level in some of the villages has concentration 4.2 mg/L and more which is harmful for the villagers of this district [7]. Arsenic (As) is also found in different parts of Rae Bareli district with concentration more than WHO permissible limit (0.01 mg/L). Even few blocks have arsenic contamination more than 0.05 mg/L. Homemade bio-sand filter is one of the option for the removal of fluoride, arsenic and coliform bacteria and this is also economical to construct, operate and maintain [8].

Adsorption is one of the significant technique in which fluoride is adsorbed onto a membrane, or a fixed bed packed with resin or other mineral particles. Many natural and low cost materials such as red mud [9,10], zirconium impregnated coconut shell carbon [11], cashew nut shell carbon [12], ground nut shell carbon [13] and clays [14] have been used as adsorbents for fluoride removal from drinking water (Table 1).

Pyrolysis is one form of energy recovery processes, which has the potential to generate char, oil and gas product [22]. Because of the thermal treatment, which removes the moisture and the volatile matters of the biomass, the remaining solid char shows different properties than the parent biomass materials. The remarkable differences are mainly in porosity, surface area, pore structures (micropores, mesopores and macropores) and physicochemical properties such as composition, elemental analysis and ash content [23]. These changes in the properties usually lead to high reactivity, and hence, an alternative usage of char as an adsorbent material becomes possible. Thus, the char becomes an attractive by-product, with applications including production of activated carbons (ACs), which is useful as a sorbent for wastewater treatment [24]. ACs are carbons of highly micro porous form with both high internal surface area and porosity, and commercially the most common adsorbents used for the removal of organic compounds from air and water streams. They also often serve as catalysts and catalyst supports.

Any cheap material, with a high carbon content and low inorganic, can be used as a raw material for the production of AC [25]; agricultural by-products have proved to be promising raw materials for the production of ACs because of their availability at a low price. They can be used for the production of AC with a high adsorption capacity, considerable mechanical strength, and

low ash content [26]. There have been many attempts to obtain low-cost AC or adsorbent from agricultural wastes such as wheat and corn straw [27], bagasse [28,29], sugar cane bagasse [30,31], almond shells [26], bamboo waste [32], rice husks [33] and rice straw [34] (Table 3).

However, challenges such as access to real low-cost adsorbents, simpler processes for development of efficient adsorbent at local levels, sufficient availability at or near to fluoride affected areas are still prevailing. These challenges have to be solved for developing efficient fluoride removal technology for rural areas in the developing, and undeveloped countries.

With this in perspective, current work was taken to explore the feasibility of fluoride adsorption from aqueous solutions and groundwater with Natural Bagasse Carbon (NBC) and Modified Bagasse Carbon (MBC) adsorbents. These low cost adsorbents are readily available in almost all parts of the rural areas in India.

2. MATERIALS AND METHODS

2.1 Natural Bagasse Carbon (NBC) Preparation

To prepare NBC as an adsorbent, raw sugarcane bagasse was collected from sugarcane shops and cut into small pieces after several washings with double distilled water. For activation of carbon the dry pieces of collected material were thermally activated in a muffle furnace at 500°C for 20-25 minutes followed by several washings with double distilled water and drying at 100°C for a period of 24 hours. Further, 300 gm of activated sugarcane carbon was treated with 3 ml of 4N HCl for 10 minutes and filtered. The filtrate solution is again treated with 3 ml of 4N NaOH and filtered through a filter paper and washed several times with distilled water until pH 7 was attained [35,37]. Finally the dried material was grinded and sieved to the desired particle sizes. Finally, the product was stored in vacuum desiccators until required.

Table 1. Comparison of fluoride removal technologies [15-21]

Technology	Advantages	Disadvantages
Coagulation/precipitation: calcium hydroxide; aluminum hydroxide	High efficiency; commercially available chemical	Expensive, efficiency depends of pH and presence of co-ions in water, adjustment and readjustment of pH is required, elevated residual aluminum concentration, formation of sludge with high amount of toxic aluminum fluoride complex and high amount of retained water (sludge dewatering is required prior disposal)
Membrane filtration: reverse osmosis; nanofiltration	High efficiency; remove other contaminates	High capital high running and maintenance costs toxic waste water produced
Electrochemical treatments: dialysis; electro-dialysis; electro-coagulation	High efficiency; high selectivity	High cost during installation and maintenance
Ion-exchange: Strong basic anion-exchange resin with quaternary ammonium functional groups	High efficiency	Expensive, vulnerable to interfering ions (sulfate, phosphate, chloride, bicarbonate, etc.), replacement of media after multiple regenerations, used media present toxic solid waste, regeneration creates toxic liquid waste, efficiency highly pH-dependent
Adsorptive materials: activated alumina; activated carbons; other natural and synthetic adsorbents	Greater accessibility, low cost, simple operation, availability of wide range of adsorbents	High efficiency often demand adjustment and readjustment of pH, some common water ions can interfere fluoride adsorption

2.2 Modified Bagasse Carbon (MBC) Preparation

Sugarcane bagasse was collected from a local sugarcane juice shop. It was washed with distilled water and dried in the sunlight. One portion of dried sugarcane bagasse was mixed with one portion of concentrated sulphuric acid and heated in an air oven for 24 h at 150°C. Dilute sulphuric acid is normally used as impregnating agent as it effectively hydrolyzes hemicellulose into monomeric sugars (xylose, arabinose, galactose, glucose, and mannose) and oligomers [36]. The heated material was washed with distilled water, and then soaked in 1% sodium bicarbonate solution overnight to remove residual acid. The material was dried in an oven at 105°C for 24 h, and then sieved in the size range of 20–50 mesh [37]. The material was subsequently placed in an airtight container, for further use.

2.3 Adsorption Experiments

The stock solution of 100 mg/L fluoride was prepared by dissolving 221 mg of anhydrous NaF (powdered form without water) in one liter of distilled water. Test solution of 5 mg/L fluoride concentration was prepared from stock solution. This concentration was selected, as it is the normal fluoride concentration in groundwater. All the experiments were carried out in 250 ml conical flasks with 100 ml test solution at room temperature (25°C). These flasks, along with test solution and adsorbent, were shaken in a horizontal shaker, to study the various parameters. At the end of desired contact time, the conical flasks were removed from the shaker, and allowed to stand for 5 min for the adsorbent to settle. Subsequently, samples were filtered using Whatman No. 42 filter paper and filtrate was analyzed for residual fluoride concentration. The fluoride ion concentration was measured with a specific ion selective electrode by use of total ionic strength adjustment buffer II (TISAB II) solution to maintain pH and to eliminate the interference effect of complexing ions. Batch study was conducted to determine the optimum conditions. The effects of pH, adsorbent dose, initial fluoride concentration, and contact time on adsorption were studied. The effect of pH on fluoride was studied by adjusting the pH of test solution using 0.1 N HCl or 0.1 N NaOH on fixed quantity of adsorbent. The physicochemical property of groundwater samples was determined prior to the batch study in accordance with the standard methods [38].

3. RESULTS AND DISCUSSION

3.1 The Effect of pH

The effect of pH on removal of fluoride was studied in the pH range of 4.0–9.0 and results are illustrated in Fig. 1 pH played a major role in fluoride adsorption on the biosorbent. In this study, the highest biosorption values (58% and 52%) were observed at acidic pH, where overall surface charges on the biosorbent may be positive. This enabled the binding of negatively charged fluoride ion. At lower pH values, the surface of the adsorbent turned out to be positively charged and this facilitated sorption of fluoride ions, probably by the anionic exchange sorption. At acidic pH due to the protonation, effect of surface functional groups such as amino, carboxyl, thiol, etc., imparts positive charge on the surface [37].

Relative sorption inhibition observed at basic pH range, might be attributed to the increase of hydroxyl ion leading to formation of aqua-complexes; thereby, retarding the sorption. According to results of this study, the pH of 7 and 6 were selected as the optimum pH when using NBC and MBC as adsorbents, respectively. The reason for choosing higher pH when using ABC and MBC was the limited difference between the fluoride removal efficiencies at this pH in comparison to lower the pH. Another reason is its cost saving advantage and a possibility to explore the minimum requirement in order to utilize the biosorbent in the rural areas, where pH adjustment may not be a preferable solution.

3.2 Effect of Contact Time

Fig. 2 illustrates the percentage removal of fluoride by two considered adsorbents at different contact times. As contact time was increased, the percentage removal also increased. However, it gradually approached an almost constant value, denoting attainment of equilibrium. In the current case, the equilibrium times were obtained at 90 and 75 min for NBC and MBC respectively.

3.3 The Effect of Adsorbent Dose

The effect of adsorbent dosage on adsorption of fluoride at pH 6 and contact time of 75 min for the considered adsorbents were studied. The results are presented as fluoride removal efficiency versus function of adsorbent dosage in Fig. 3. It has been observed that up to certain

level of higher doses of adsorbents resulted higher removal of fluoride. This might be because of higher availability of surface area and pore volume at higher doses. More the fluoride removal efficiencies increased from 41.8 to 60.2% and 56.2 to 78.4% for 2.0–10.0 gm/L dosage of NBC and MBC respectively. However, it can be seen from Fig. 3 that after dosage of 8 gm/L for NBC and MBC there was no significant change in percentage removal of fluoride. It was due to the overlapping of active sites at higher dosage, thus reducing of the net surface area.

3.4 Effect of Initial Fluoride Concentration

The effect of initial fluoride concentration was investigated by adding fixed amounts of adsorbents onto different fluoride concentration solutions (2.5, 5, 7.5, 10, 12.5, and 15 mg/L). The effect of initial fluoride concentration on the fluoride removal efficiency is shown by Fig. 4.

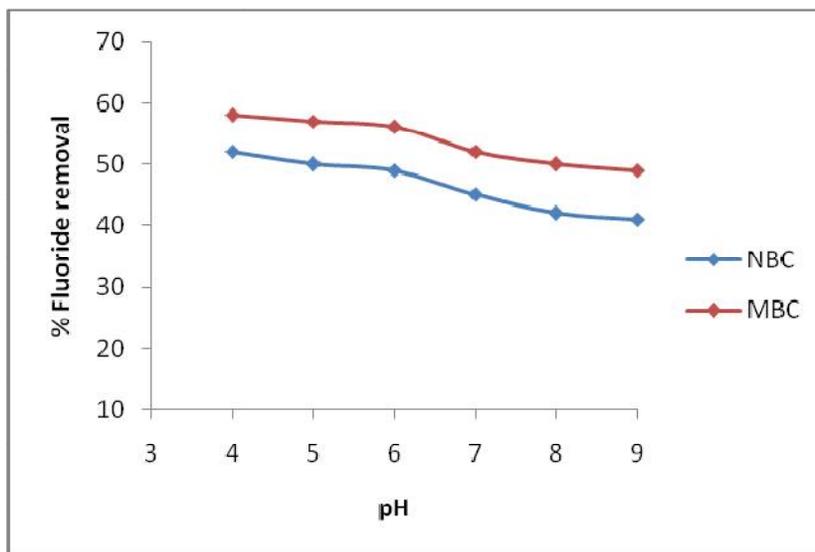


Fig. 1. The effect of pH on fluoride removal

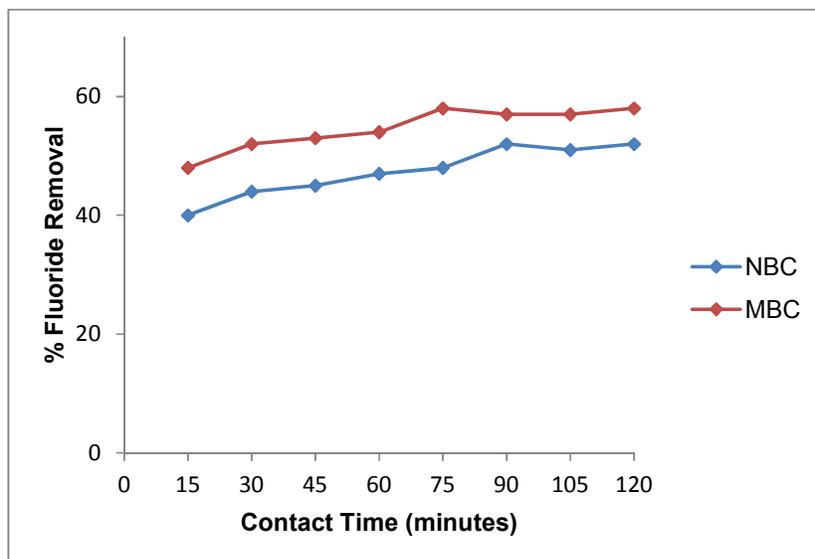


Fig. 2. Effect of contact time on fluoride removal efficiency

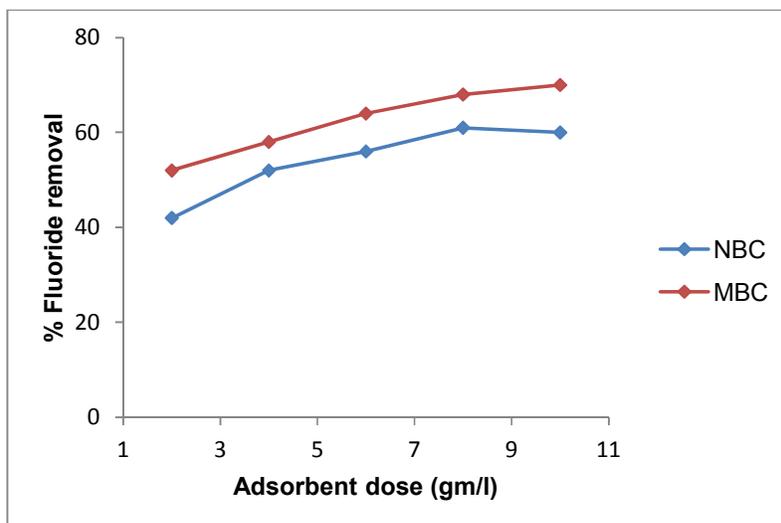


Fig. 3. Effect of adsorbent dose on fluoride removal

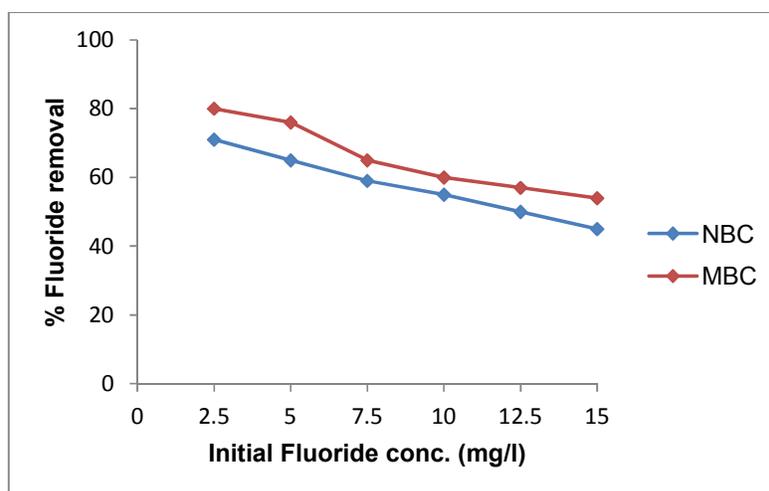


Fig. 4. Effect of fluoride concentration on fluoride removal efficiency

The results illustrated that fluoride removal efficiency was decreased, by increasing the initial fluoride concentration. The reason was that the capacity of the adsorbent materials gets exhausted sharply with the increase in initial fluoride concentration. This was due to the fact that for a fixed adsorbent dose, the total available adsorption sites were limited, which became saturated at a higher concentration [37].

The amount of fluoride adsorbed per unit adsorbent (q_e) (mg/g) was calculated according to following Equation (1).

$$q_e = \frac{(C_i - C_e)}{m} V \quad (1)$$

where C_i and C_e are initial and final fluoride concentrations respectively, V is the volume of the solution (L), and m is mass of the adsorbent (gm). The results of Fig. 5 demonstrate that the amount of adsorbed fluoride increased with the increase in fluoride initial concentrations. It also led to an increase the equilibrium sorption, until sorbent saturation was achieved.

3.5 Adsorption Isotherms

The experimental data was applied in accordance with the linearized form of the Freundlich isotherm model equation (2).

$$\log \left(\frac{x}{m} \right) = \frac{\log K_f + 1}{n \log C_e} \quad (2)$$

where x is the amount of solute adsorbed (mg), m is the mass of adsorbent used (gm), Ce the equilibrium solute concentration in solution (mg/L), and K_f and 1/n are the constants, which represent adsorption capacity and adsorption intensity respectively. The values of K_f and n were obtained from the slope and intercept of the plot between log (x/m) and log Ce. The Freundlich equation deals with physicochemical adsorption on heterogeneous surfaces. Linearized form of the Freundlich equation is demonstrated in Fig. 6. In this study, values of K_f and n were found to be “1.15 and 2.68” and “1.935 and 3.102” for NBC and MBC respectively (Table 2). Adsorption data fitted well in linearized form of model equation (R² > 0.92) which indicated the acceptability of the model.

3.6 Adsorption Kinetics

Adsorption kinetics was tested in order to understand the mechanism of fluoride adsorption on studied adsorbents. For this, pseudo-first-

order and pseudo-second-order models were used. The rate constant (k) for sorption of fluoride, for all of the considered adsorbents was studied by applying the Lagergren rate equation eq. (3). For pseudo second-order model, eq. (4) was applied:

$$\log (q_e - q) = \frac{\log q - k_1 t}{2.303} \quad (3)$$

$$\frac{t}{q_t} = \frac{1}{q_e} t + \frac{1}{k_2 q_e^2} \quad (4)$$

Table 2. Isotherm parameters of adsorbents

Isotherm	Adsorbents	
Freundlich	NBC	MBC
K _f (mg/g)	1.155	1.935
n	2.68	3.102
R ²	0.93	0.98

where q_e and q (both in mg/g) are the amount of fluoride adsorbed at equilibrium and at time t, respectively. k₁ (/min) and k₂ (g/mg/min) are the kinetics rate constants for the pseudo first- and second order models, respectively. Fig. 7 shows the pseudo-second order kinetic model for

Table 3. Comparison of the defluoridation capacities of different biomass based sorbents

Adsorbent	pH	Adsorption capacity (mg/g)	References
Powdered biomass <i>Tinospora cordifolia</i>	7.0	25	[39]
Biomass carbon prepared at 300°C	5.8	0.52	[40]
Used tea leaves	1.5	0.51	[41]
Activated carbon derived from rice straw	2.0	0.04	[42]
Ammonium carbonate activated carbon of <i>Tamarindus indica</i> fruit shells	7.05	22.33	[43]
zirconium (iv)-impre-gnated groundnut (<i>Arachis hypogia</i>) shell carbon	-	2.32	[13]
Modified Bagasse Carbon	7	3.91	Current work

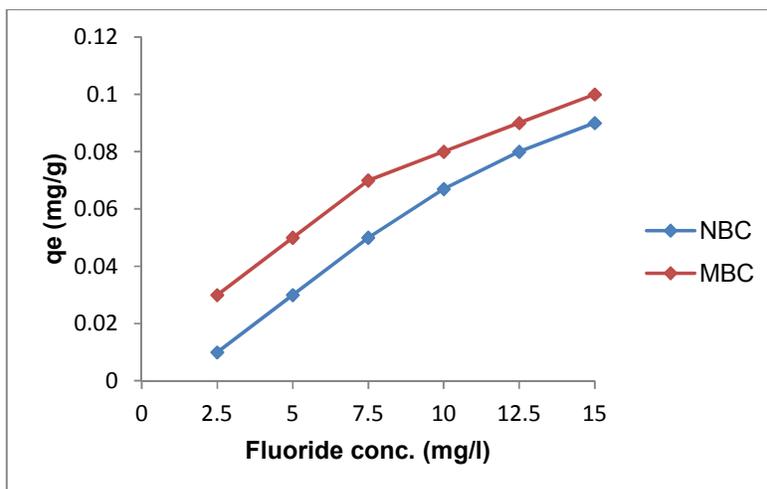


Fig. 5. Effect of fluoride concentration on fluoride adsorption

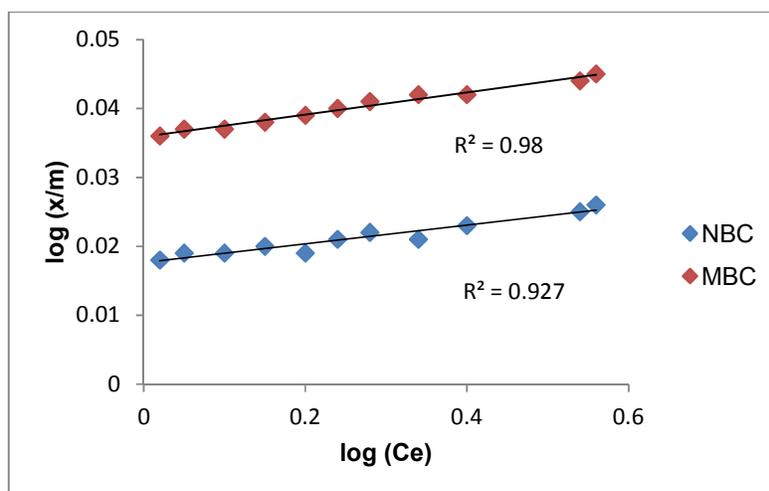


Fig. 6. Freundlich isotherm for adsorption of fluoride by NBC and MBC

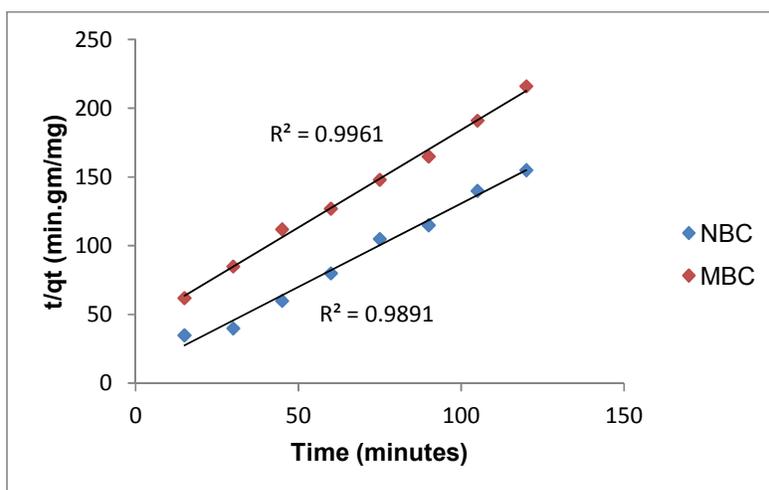


Fig. 7. Pseudo second order kinetic model for fluoride adsorption on NBC and MBC

studied adsorbents. The results demonstrated that pseudo second-order model ($R^2 = 0.989$, $k_{2NBC} = 0.082$ g/mg/min) and ($R^2 = 0.996$, $k_{2MBC} = 0.114$ g/mg/min) was better than the first-order model ($R^2 = 0.91$, $k_{1NBC} = 0.035$ /min) and ($R^2 = 0.95$, $k_{1MBC} = 0.021$ /min) to correlate the adsorption kinetics data. This suggested that the adsorptions of fluoride onto the considered adsorbents were mainly dominated by chemical adsorption reactions.

4. CONCLUSION

In the present study, agricultural biomass based adsorbents were studied for removal of fluoride from synthetic, as well as from groundwater

sample. The conclusions drawn from this study are given below:

1. The NBC and MBC removed 51.8 and 58.4% respectively from an aqueous solution of 5 mg/L fluoride at pH 6.0, with the contact period of 75 min and a dose of 4 gm/L.
2. Adsorption of fluoride on all studied adsorbents from aqueous solution followed pseudo-second order reaction, and the mechanism of fluoride removal on adsorbents was found to be complex.
3. Significant amount of fluoride can be reduced in the neutral pH range by these adsorbents, which makes them suitable for utilize in drinking water treatment,

especially in rural areas where minimum facilities are available.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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