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Adsorptive Removal of Fluoride from aqueous solution by Alumina/ Commercial Activated Carbon Composite

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Abstract

Excessive exposure to fluoride in drinking-water may cause fluorosis which affects the teeth and bones. In this study, the potential of alumina/commercial activated carbon composite (ACAC composite) for adsorptive removal fluoride from aqueous solution has been investigated. The ACAC composite was prepared by precipitation of aluminium hydroxide on the surface of commercial activated carbon (AC) followed by calcination. Fluoride adsorption experiments were performed by batch method. Effects of various experimental parameters like pH, adsorbent dose, contact time and initial concentration on the fluoride removal were studied. The results shows that, the percentage of fluoride removal by the resultant ACAC composite was much higher (~70%) compared to the plain commercial AC (~30%). The maximum fluoride removal was observed at pH 2.0, adsorbent dose of 20 g/L and contact time of 180 min. The adsorption isotherms were analyzed using Langmuir and Freundlich isotherms. The adsorption data was fitted Langmuir better than Freundlich isotherm which confirms the monolayer coverage of fluoride onto the homogenous surface of the composite adsorbent. The maximum adsorption capacity (qm of Langmuir) of the ACAC composite for fluoride removal was 4.53 mg/g. The findings indicate that, loading of alumina (Al2O3) can enhance the fluoride adsorption efficiency of modified ACAC composite compared to unmodified plain AC. The composite can be used as an alternative adsorbent for removal of fluoride from contaminated water.

Keywords: *Alumina, commercial AC composite, adsorption, fluoride, water.*

1. Introduction

Fluoride is a nutrient, essential for proper development and maintenance of mineralized tissues like teeth and

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the bones. It has both beneficial and harmful effects on the human health depending upon its level. Upto 1.5 mg/L, it prevents against dental caries and promotes dental health but, higher levels of fluoride (> 1.5 mg/l) may result in dental or skeletal fluorosis. The dental fluorosis is characterized by discoloration or pitting of teeth and the skeletal fluorosis is marked by osteosclerosis, calcification of tendons and ligaments and deformities in bones. Besides, It can also lead to various diseases such as cancer, infertility, brain damage, alzheimer's syndrome, and thyroid disorder etc [1]. As set by WHO, the maximum permissible limit of fluoride in drinking water is 1.5 mg/L [2]. The major source of fluoride consumption is through drinking water.

The presence of elevated levels of fluoride in groundwater is due to natural or anthropogenic activities or a combination of both. Naturally, fluorides are released into groundwater through the break down and leaching of hardrocks, rich in fluoride such as sellaite, fluorite, cryolite, fluorapatite, apatite etc. The mobilization of fluoride in groundwater is also contributed by hydrogeological conditions. Volcanic ash and combustion of coal are the next major source for fluoride. Occurrence of fluoride bearing minerals and nature of the rocks determine the concentration of fluoride in ground water. Anthropogenic activities include phosphatic fertilisers, improper disposal of liquid waste from industries, alumina smelting, cement production and ceramic and brick firing etc [3]. Generally high alkalinity and low calcium favours high fluoride content in ground water [4].

Groundwater is already the preferred source of drinking water supplies globally. During the last few decades, fluoride contamination of groundwater and related health hazards has been recorded in several countries arround the world including Asia, Africa, Europe as well as USA and Australia. It is estimated that around 200 million people worldwide are thought to be exposed to drinking water with elevated levels of fluoride [2]. In India, the presence of excessive fluorides in groundwater is reported in 17 states, affecting more than 6 million people seriously [4]. Nepal's geography is similar to some parts of India and they are still responsible for excessive fluoride of their water supply. Fluorosis cases have been observed in the Terai region (low land) of Nepal [5]. However, the available data is quite unsystematic, incomplete and inconsistent. As such, it is necessary to remove the excessive fluoride from ground water before drinking if the fluoride concentration is higher than 1.5 mg/L.

Various treatment methods such as adsorption [6], coagulation- precipitation [7], reverse osmosis [8], electrolytic treatment [9] ion-exchange [10] etc. have been investigated for removal of excess fluoride from water. Of all these treatment methods, adsorption [6] and precipitation/coagulation [11] have been commonly used to remove fluoride from water in developing countries. Coagulation-precipitation (also known as Nalgonda technique) is was preferable at all levels because of the low price and ease of handling. However, its main disadvantage is the generation of large quantity of sludge and uncontrolled pH of treated water. Alternatively, adsorption has been the most ideal method due to its simplicity and high efficiency and the availability of a wide range of adsorbents. The method can remove ions over a wider range of pH, even to a lower concentration [12]. Numerous adsorbents and their modifications have been explored for the fluoride removal from water which include activated carbon [6], activated alumina [13], calcite [14], bauxite [15], bone charcoal [16] etc. Among several, activated carbon (AC) and activated alumina are commonly used. Activated carbon (AC) is unique and versatile adsorbents for fluoride removal due to its large surface area and high porosity. These properties of AC causes a high metal dispersion with the relatively weak interaction with the support. Likewise, activated alumina is also preferred adsorbents due to its mesoporous structure, low toxicity, recyclability, and modifiability.

Commercial grades of AC, in both powdered (PAC) and granular (GAC) form, has been extensively used in water treatment. Generally, GAC has a larger particle size compared to PAC and consequently, presents a smaller external surface and diffuse rapidly. It is used for adsorption of gases and vapours [17]. Because of smaller size with larger surface area, PAC can adsorbs organic compounds to its surface more quickly. It is used in adsorption of liquid phase applications. Commercial ACs have BET surface areas ranging from 500 and 1500 m2/g or even as high as 3000 m2/g [18]. AC can be prepared by two activation methods: Physical and chemical activation.

Generally the physical activation requires high temperature and longer activation time as compared with chemical activation. ACs for liquid phase applications are generally produced commercially by chemical activation of wood peat, lignite etc [19].

Although activated carbons (ACs) are the most used adsorbents globally with high capacities, they are generally characterized by low selectivity for fluoride ions due to physical adsorption. ACs are thus, modified by oxidation/ subsequent impregnation with multivalent ions such as zirconium, titanium, iron, calcium, etc for fluoride removal. High electronegativity and small size make fluoride ion a hard base and displays a strong affinity for multivalent metal ions like Al3+, Fe3+, Ca2+, Zr4+, La3+, Ce4+ etc. [20]. The dispersion of these metals in the matrix of AC. can provide high fluoride adsorption capacity. Recently, considerable work has been conducted in developing metal loaded AC composite adsorbents for removal of fluoride from aqueous solution. For instance, the adsorption capacity of fluoride on aluminum impregnated carbon composite is found to be 3 to 5 times higher than that of plain AC [21]. Zirconium impregnated coconut shell carbon was found to have fluoride adsorption capacity, 25 to 30 times that of plain AC [22]. The adsorption capacity of Aluminium-iron-amended activated bamboo charcoal was 4.88 times higher than activated bamboo charcoal without Al2O3 [23]. Similarly, charcoal composite adsorbents that contain dispersed aluminum and iron oxides exhibited the best efficiency with a fluoride sorption capacity of 13.64 mg/g [24]. An alumina supported carbon composite, synthesized from eggshell waste was found to be more selective and efficient for fluoride removal [25]. To our knowledge, the literature contains few reports on the use of alumina supported AC as an adsorbent for fluoride removal. The aim of the present study, therefore was undertaken to explore the possibility of the alumina/commercial AC composite for the removal of fluoride from aqueous fluoride solution.

2. Experimental

2.1. Materials

The All the reagents used in this study were of analytical grade and used in this study without further purification. A commercial activated carbon and commercial activated alumina was purchased from Thermo Fisher Scientific India PVT.LTD. Stock fluoride solution was prepared by dissolving 0.221 g of anhydrous sodium fluoride in distilled water and diluted to 1000 ml. The standard solutions of appropriate concentrations were prepared by dilution of stock solution. The concentration of fluoride ion in the solutions is measured on Orion ionplus Fluoride Electrode Orion 94-09, 96-09, Thermo Electron Corporation, USA.

2.2 Preparation of ACACC adsorbent

The ACAC composite was prepared by precipitation method. 36.8 g of aluminium nitrate (Al(NO3)3.9H2O) g was dissolved in 400 ml. distilled water and 5 g AC was added. Subsequently, 5M NaOH was added dropwise, precipitating out Al(OH)3. The adding of NaOH was continued till the precipitation of Al(OH)3 was completed. The product was dried in oven at 110 oC for 12 hrs and then calcined in the tube furnace at 300 oC for one hr. The prepared ACAC composite was washed with distilled water till it became neutral and dried at 100 oC for 24 hrs. The ACAC composite was sieved to obtain a particle size of 212 μm and used for removal of fluoride from water.

Reaction of Al(NO3)3 with NaOH occurs in pores of the AC to form Al(OH)3. The Al(OH)3 was precipitated under alkaline medium.

$$Al\,(NO_3)_3 + 3NaOH \rightarrow Al(OH)_3(s) + 3NaNO_3$$

2.3 Adsorption experiments

Batch adsorption procedure was used to determine the effect of various operating conditions on the adsorption process. A definite amount of ACAC composite was suspended in 50 ml of fluoride solution in 100 ml plastic conical flasks. The suspension was agitated on Digital VDRL Rotator at 225 rpm at room temperature. After equilibrium, the suspension was filtered. Then, the residual fluoride ion concentration in the filtrate was measured. The fluoride ion concentration in the sample was determined by using fluoride ion selective electrode (ISE) according to the procedure using calibration curve [26].

The adsorption studies were conducted for the optimization of various experimental conditions such as pH, contact time, initial concentration, adsorbent dose and adsorption isotherm.

2.3.1 Effect of adsorbent dose

The effect of adsorbent dose on percentage of fluoride removal was studied by varying adsorbent dose from 2.5 g/L to 30 g/L. Other parameters were kept constant such as initial fluoride concentration 19 mg/L, contact time 3 hrs and pH 6.5.

2.3.2 Effect of adsorbent dose

The effect of contact time on the percentage of fluoride removal was studied at different contact time from 5 min to 300 min. During the study, other parameters constant kept constant such as the adsorbent dose 2.5 g/L, initial fluoride concentration of 19 mg/L and pH 6.5.*4.3 The Future of Online to Offline (O2O) Services*

2.3.3 Effect of pH

The Effect of pH on the percentage of fluoride removal was studied over a pH range of 2 to 10 by keeping other parameters constant such as adsorbent dose 2.5 g/L, initial fluoride concentration 19 mg/L and contact time 3 hrs. pH of the solution was adjusted by adding 0.01 N HCl and 0.1 N NaOH.

2.3.4 Effect of initial concentration

The Effect of initial fluoride concentration on percentage removal was studied by varying fluoride concentration from 5 to 160 mg/L while keeping other parameters constant such as pH 6.5, contact time 3 hr, and adsorbent dose of 5 g/L.

The percentage removal of fluoride ions was calculated from the following equation (1).

$$\text{Removal}\ (\%) = \frac{(C_o - C_e) \times 100}{C_o} \tag{1}$$

where, C_o is the initial fluoride ion concentration in solution in mg/L, C_e is the equilibrium fluoride ion concentration in mg/L.

2.3.5 Adsorption isotherms

Adsorption isotherms were studied using solutions of different initial fluoride concentrations varying from 5 to 100 mg/L. Other parameters were kept constant such as adsorbent dose 5 g/L, contact time 3 hrs and pH 6.5. The amount of fluoride ions adsorbed in milligram per gram (qe) was determined by using the following equation (2).

$$q_e\,(mg/gm) = \frac{(C_o - C_e)\times V}{M} \quad (2)$$

where, V is volume of fluoride solution in liter and m is the mass of the ACAC composite in gram.

3. Result and Discussion

3.1 Adsorption studies

Percentage of fluoride removal of Alumina (Al_2O_3), commercial AC and ACAC composite were compared and shown in Figure: 1.

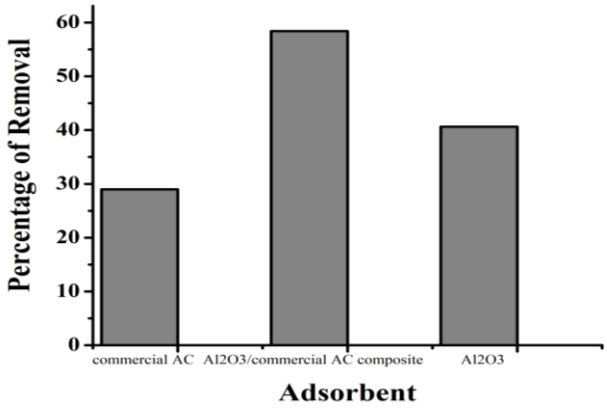


Figure 1. Percentage of fluoride removal by Alumina (Al2O3), commercial AC and ACAC composite

The percentage of fluoride removal by both Alumina and commercial AC was found to be less than 40 %. When ACAC composite is used, the adsorption capacity increased to ~60 %. The adsorption capacity of the ACAC composite is significantly greater than the individual components for removal of fluoride from water. It indicates that, the synergistic effect of the alumina and commercial AC in the composite enhanced adsorption of the fluoride from water.

Mechanism of fluoride adsorption was inferred in terms of ligand exchange reaction between hydroxyl ion of the co-ordination sphere on the ACAC composite surface and fluoride ions of water. The ligand exchange was considered as chemical adsorption which is characterized by fast, strong and less reversible. In water, the surface loaded Al(III) ions on the ACAC composite tends to coordinate with H_2O molecules and chemically adsorbed on the surface of the composite. In the process, the H_2O molecules are dissociated, forming adsorbed OH groups. Now, Al(III) cation on the composite adsorbent surface behaves as a Lewis acid and exchanged their adsorbed hydroxyl ligand [27]. A covalent chemical bond was formed between the Al(III) cation (Lewis acid) and fluoride anions of aq. solution on the adsorbent surface which results in the release of OH^- anions, previously bonded to the Al(III) cation. In other words, Al(III) ions exchanged its structural hydroxyl ligands against fluoride ions and an inner sphere complex was formed at the adsorbent surface. During the process, Al(III) ion acted as a bridge between composite adsorbent surface and fluoride ions.

It is suggests that the loaded Al(III) ions tend to extensively polymerized and hydrolyzed with OH^- ions as well as water molecules for ligand exchange with fluoride species even at very low concentration and converted to $[Al(H_2O)n]^{3+}$ species. Schematic diagram for ligand exchange reaction between hydroxyl ion on the ACAC composite surface and fluoride ions is shown in Figure 2.

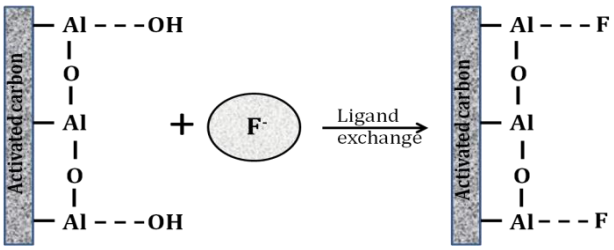


Figure 2. Schematic diagram for ligand exchange reaction between hydroxyl ion on the ACAC composite surface and fluoride ions.

The mechanism can be represented by the following reaction [28]. The active sites of the ACAC composite were the following surface groups such as ≡Al−OH2+, ≡Al−OH and ≡Al−O- where ≡ represents the surface of the Al (III) loaded composite adsorbent. The first and last groups are formed when the ≡Al−OH group is protonated and deprotonated according to the following reactions.

$$\equiv Al–OH + H^+ \rightleftharpoons \equiv Al–OH_2^+$$

$$\equiv Al–OH_2^+ \rightleftharpoons Al–OH$$

$$\equiv Al–OH \rightleftharpoons Al–O^- + OH$$

The fluoride lignd exchange mechanism may occur on the surface of the ACAC composite according to the following reaction:

$$\equiv Al–OH + F^- \rightarrow \equiv Al–F + OH^-$$

$$\equiv Al–OH_2^+ + F^- \leftrightarrow \equiv Al–OH_2\text{-----}F$$

$$\equiv Al–OH_2^+ + F^- \rightarrow \equiv Al–F + H_2O$$

3.2 Effect of pH

The initial pH of the solution is a major factor in the adsorption process since it affects the surface charge of the adsorbent, the degree of ionization and the specification of the surface functional groups [29]. Effect of pH on the percentage of fluoride removal by the ACAC composite is shown in Figure: 3.

A gradual increase in the fluoride uptake was observed with rise in pH from 2 to 10 and highest percentage removal of ~ 88 % was obtained at pH 2 (Figure: 2). Maximum percentage of adsorption by the ACAC composite was observed at low pH. This may be due to an increase in H+ on the adsorbent surface which makes the surface of the ACAC composite positively charged. It resulted in electrostatic attraction between positively charged adsorbent surface and fluoride ions. So fluoride adsorption was high at low pH levels. The removal efficiency decreased with increasing pH. It can be attributed to (i) the competition for the active sites by OH− ions and (ii) the electrostatic repulsion of anionic fluoride by the negatively charged adsorbent surface. Thus at higher pH, the fluoride removal by the ACAC composite was found to decrease. The same trend of the adsorption of fluoride at different pH was also observed in AC prepared from banana (Musa paradisiaca) peel and coffee (Coffea arabica) husk [30].

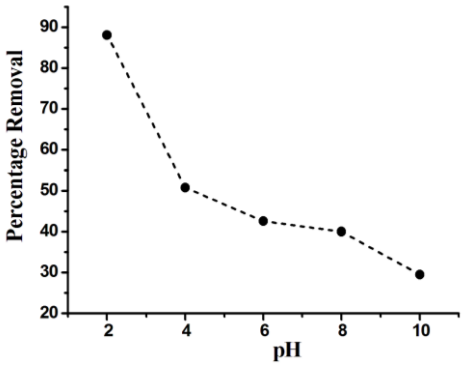


Figure 3. Effect of pH on the percentage of fluoride removal.

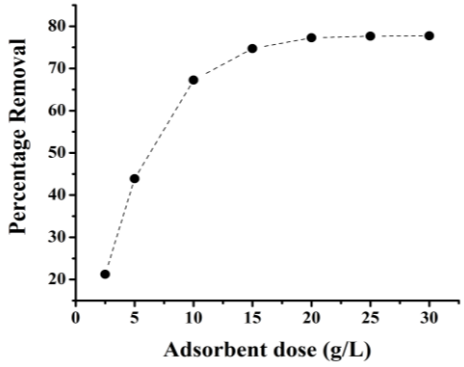


Figure 4. Effect of adsorbent dose on the percentage of fluoride removal.

3.3 Effect of adsorbent dose

The optimum adsorbent dose is a key parameter to control both availability and accessibility of adsorption sites. The effect of adsorbent dose on the percentage removal is shown in Figure: 4. As the adsorbent dose increased from 2.5 g/L to 20 g/L, the fluoride removal percentage also increased. Beyond this dose, the percentage removal did not increase considerably. As such, an optimum dose of 20 g/L was required for a maximum fluoride removal. The increased fluoride uptake with the increase of adsorbent dose could be explained by high surface area and the availability of more active binding sites for fluoride adsorption on the surface of the ACACC adsorbent. Further addition of the adsorbent did not show any significant increase in percentage removal. This could be due to aggregation of the particles, resistance to mass transfer and repulsive forces between binding sites at higher adsorbent dose. Similar results were obtained on fluoride adsorption from water by Al2O3/Areca nut AC composite [31].

3.4 Effect of contact time

The contact time indicates the rate of adsorption process. It is thus an important parameter used to evaluate the kinetics of adsorption process [32]. The effect of contact time on the percentage of fluoride removal is shown in Figure: 5.

The percentage of fluoride removal increased with the increase in time from 5 minutes to 180 minutes and then it became almost constant denoting attainment of equilibrium. Therefore, minimum contact time for the maximum removal by the ACAC composite was considered at 180 minutes. The fluoride removal was very rapid in less than 60 min indicating that, the adsorption of fluoride probably took place due to the diffusion into the pores on the surface of the composite adsorbent. The adsorption of fluoride was further increasing with increase in time which might be due to the availability of abundant active sites on the surface. The decrease in fluoride removal beyond 180 min might be due to less availability of vacant sites on the composite adsorbent. It lowered mass transfer of the fluoride ions to the exterior surface of ACAC composite from the liquid bulk. This result is in very good agreement with the effect of contact time on fluoride removal from water by zirconium-impregnated ground-shell carbon [33].

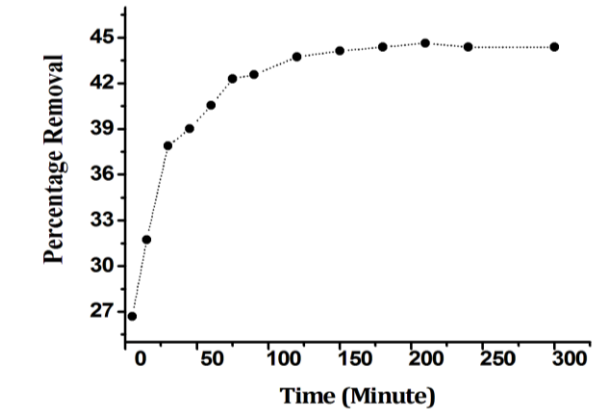


Figure 5. Effect of contact time on the percentage of fluoride removal

3.5 Effect of initial concentration

The adsorption rate of adsorbate onto adsorbents is a function of the initial concentration of the fluoride ions. This makes it a major factor to be determined for an effective adsorption. The effect of initial concentration on the percentage of fluoride removal is shown in Figure 6.

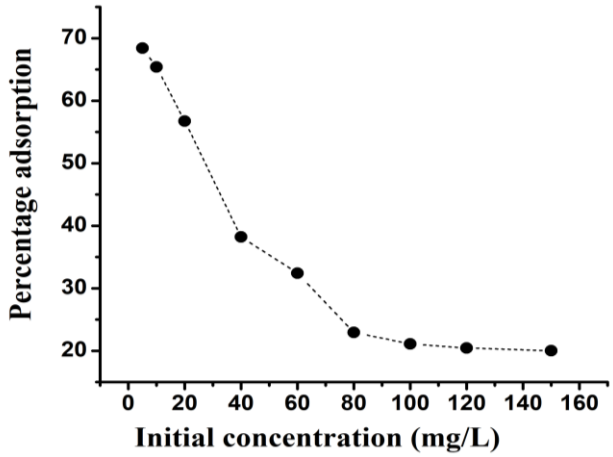


Figure 6. Effect of initial concentration on the percentage of fluoride removal

As seen from Figure: 5, the percentage removal of the fluoride ion has decreased with an increase in initial concentration of the fluoride ion from 5 to 150 mg/L after which it became fairly stable. At low concentrations of the fluoride ion, sufficient numbers of active sites were available on the ACAC composite and most of the fluoride ions interacted with these active sites. Thus, percentage removal of fluoride was more. This decrease was due to the fact that all adsorbents have a fixed number of active sites. At higher initial concentrations, the active sites were saturated and incoming fluoride ions from water was repelled by repulsive forces between the fluoride ions on the solid surface and in solution. Hence, an increase in initial concentration of fluoride led to a decrease in its percentage removal. Similar study [34] also pointed out that, there was a decrease in fluoride removal efficiency of the adsorbents with increasing initial concentrations of fluoride.

3.6 Adsorption Isotherms

The equilibrium data, commonly known as adsorption isotherm are basic parameters for the design of adsorption systems to remove contaminant from solutions. Adsorption isotherm represents the relationship between the concentration of a solute in solution and the quantity adsorbed at the solid surface at constant temperature. The parameters of the isotherm provide information on, adsorption mechanism as well as affinity of the adsorbent. It is therefore important to determine the most suitable correlations of equilibrium curves to optimize the condition for designing adsorption of fluoride ions [35]. Fluoride adsorption by the ACAC composite was analyzed using the Freundlich and Langmuir isotherms. These isotherms are most frequently used isotherms for the application of AC in water treatment. Langmuir adsorption isotherm describes the saturated monolayer adsorption of solutes on the homogenous adsorbent surface without interaction between adsorbed molecules whereas Freundlich adsorption isotherm applied to non-ideal sorption on heterogeneous surfaces by multilayer sorption. The linear form of the Langmuir isotherm can be expressed as

$$\frac{C_e}{q_e} = \frac{1}{b\,q_m} + \frac{C_e}{q_m} \tag{3}$$

where q_e is the monolayer adsorption capacity of the adsorbent (mg/g), b is the Langmuir adsorption constant (L/mg) related to the energy of adsorption and q_m is the maximum monolayer adsorption capacity of adsorbent (mg/g) and C_e (mg/L) is the equilibrium concentration of the fluoride in solution. The Langmuir constant (b) and maximum adsorption capacity (q_m) can be calculated from slope and intercept of the linear plot. Similarly, the linear form of the Freundlich isotherm can be expressed as

$$\log q_e = \log K + \frac{1}{n} \log C_e \tag{4}$$

where, K (mg/g)(mg/L)^n or (mg/g (L/mg)$^{1/n}$) and dimensionless constant, 1/n are Freundlich adsorption constants related to the adsorption capacity and intensity of the adsorbents respectively. It also indicates the relative distribution of the energy and the heterogeneity of the adsorbate sites. The value of adsorption intensity (n) and Freundlich constant (K) can be determined from slope and intercepts of the linear plot. In general, the Freundlich constant, K value increased with increasing the adsorption capacity of adsorbent for a given adsorbate. The applicability of the isotherm equation was evaluated by judging coefficient of determination (R^2) value. The isotherm is favored by the adsorption process if the value of R^2 of graph is close to unity. A Langmuir isotherm for the adsorption of fluoride on the ACAC composite was plotted with C_e/q_e v$_s$ C_e and is shown in Figure: 7. Freundlich isotherm for the adsorption of fluoride on the ACAC composite was plotted with log q_e vs. log C_e which is shown in Figure: 8.

Both the Langmuir and Freundlich adsorption isotherms fitted well for the fluoride adsorption onto ACAC composite with the coefficient of determination (R^2) of 0.990 and 0.942 respectively. Moreover, the R^2 value obtained for Langmuir isotherm model (0.990) was slightly higher than that for the Freundlich isotherm (0.942). Hence, the fluoride adsorption on ACAC composite followed Langmuir isotherm suggesting that, fluoride ions formed a monolayer surrounding the structurally homogenous solid surface of the ACAC composite and the number of active sites on the carbon surface was limited. Langmuir and Freundlich parameters for the adsorption of Fluoride onto the ACAC composite are shown in Table: 1.

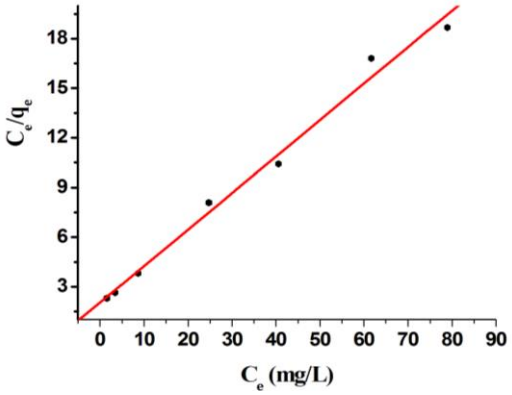


Figure 7. Plot of C_e/q_e vs C_e for Langmuir isotherm.

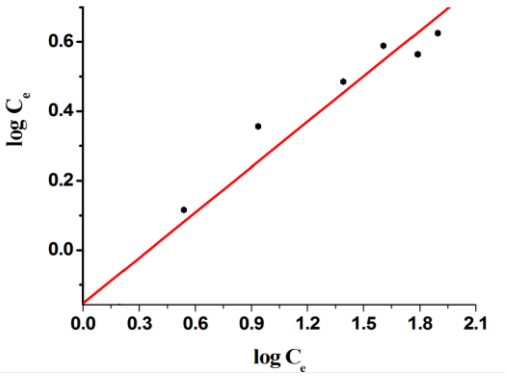


Figure 8. Plot of log q_e vs C_e for Freundlich isotherm.

Table 1. Langmuir and Freundlich parameters for the adsorption of Fluoride onto the ACACC adsorbent.

Species	Adsorbent	Langmuir Isotherm			Freundlich Isotherm		
		q_m (mg/g)	b (L/mg)	R^2	K (mg/g)(mg/L)^n	1/n	R^2
Fluoride	Al_2O_3/CAC composite	4.533	0.107	0.990	1.422	0.436	0.942

The maximum adsorption capacity (qm of Langmuir) for adsorption of fluoride by the ACAC composite was found 4.53 mg/g. The small value of the Langmuir constant, b (0.107 L/mg) indicated a low heat of adsorption. Likewise, the value of 1/n derived from Freundlich isotherm was 0.436 (between 0−1). The value of 1/n was lower than 1, confirmed the stronger interaction between ACAC composite and the fluoride ions. The higher value of adsorption capacity, K suggesting that, fluoride ions had a greater affinity for the ACACC adsorbent. The values of both Freundlich parameters K and 1/n were greater than 0 confirmed the chemi-adsorption of fluoride by the ACACC adsorbent. A similar result has been obtained with powdered activated charcoal [36].

The features of a Langmuir isotherm can be expressed in terms of dimensionless constant called separation parameter (R_L, also called equilibrium parameter) which was calculated from following equation. The value of the separation factor R_L describes the nature of adsorption of the isotherm [37].

$$R_L = \frac{1}{1+bC_o} \quad (5)$$

where, C_o is the initial fluoride ion concentration (mg/L) and b is the Langmuir equilibrium constant (L/mg). There four possibilities for the R_L value are (i) $0 < R_L < 1$ for favorable sorption, (ii) $R_L > 1$ for unfavorable sorption, (iii) $R_L = 1$ for linear sorption, and (iv) $R_L = 0$ for irreversible sorption. It was observed that the value of R_L lying between 0 and 1 confirmed the favorable uptake of fluoride onto the ACACC adsorbent.

In adsorption process, the performance of the adsorbent is evaluated from its maximum adsorption capacity (q_m), obtained from the fitted adsorption isotherm. So, the value for maximum adsorption capacity for adsorption of fluoride ions onto the ACAC composite was also compared to that of other adsorbents. Maximum adsorption capacity (q_m) for adsorption of fluoride by various adsorbents is shown in Table: 2.

Table 2. Maximum adsorption capacity (qm) for adsorption of fluoride by various adsorbents.

Fluoride Adsorbents	**Maximum adsorption capacity**	**Source**
	q_m (mg/g)	
Diatomite Modified with Aluminum Hydroxide	1.67	Akafu etal., 2019
Zr imp. groundnut shell carbon	2.3	Alagumuthu and Rajan, 2010
Aluminum impregnated coconut fiber ash	3.19	Kumar et al., 2015

Al_2O_3/Areca nut AC composite	7.62	Sahira Joshi, 2018 (Previous study)
Alumina/ commercial activated carbon composite	4.53	Present study

This adsorption capacity of the ACAC composite was significantly higher compared to other adsorbents such as Zr imp. groundnut shell carbon, Aluminum impregnated coconut fiber ash and diatomite modified with aluminum hydroxide. Conversely, the adsorption capacity was lower than Al_2O_3/Areca nut AC composite. Therefore, as prepared ACAC composite was proved to be effective for the removal of fluoride from aqueous solution.

4. Conclusion

In this study, an alumina/commercial carbon composite has been synthesized by compositing commercial AC with alumina by precipitation method and evaluated for removal of fluoride ions from water. The percentage removal of the ACAC composite was much higher (~70%) as compared to the plain unmodified commercial AC (~30%). Optimum conditions of removal of fluoride from water by the ACAC composite were (i) pH-2 (ii) adsorbent dose-20 g/L and (iii) contact ime-180 min. The adsorption data were fitted more to Langmuir isotherm than Freundlich isotherm. The maximum monolayer adsorption capacity of ACAC composite was 4.53 mg/g. Adsorption capacity of the ACAC composite is found to be comparatively higher than Zr imp. groundnut shell carbon, Aluminum impregnated coconut fiber ash and diatomite modified with aluminum hydroxide. Based on the results, it can be concluded that, the Alumina/commercial AC composite can be promising adsorbent for removal of fluoride from contaminated drinking water.

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