

IRON AND MANGANESE REMOVAL FROM GROUNDWATER USING CIGARETTE FILTER BASED ACTIVATED CARBON

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ABSTRACT.

Cigarette butts (CB) are considered one of the most hazardous litters in the world, containing toxic substances and non-biodegradable cellulose acetates. Correspondingly, recycling these wastes will be double edge solution for both groundwater treatment and a clean environment. The cigarette filter based activated carbon (CFAC) was made using a KOH chemical activating agent with the ratio of carbonaceous material/KOH= 1:2, 1:2.5, 1:1.5 denoted as CFAC-2, CFAC-2.5, and CFAC-1.5 with the temperature 500°C for four hours. Three different filters were installed using sand, CFAC-1.5, charcoal (made from CBs) whereas CFAC-1.5 had shown the highest removal efficiency (Iron-99%, Chloride-97%, Color-94%, Turbidity-77%, TDS-%, Hardness-36%, Manganese-33%). Analyzing the adsorption isotherm of CFAC-2 and CFAC-2.5 for methylene blue (MB) adsorption using the Langmuir and Freundlich isotherm model, the adsorption process was more biased towards Langmuir isotherm due to a higher correlation coefficient ($R^2 \approx 0.994$). CFAC-2.5 got the highest adsorption capacity after comparing it with CFAC-2. Compiling the findings, conclusions, and recommendations for future research had been outlined.

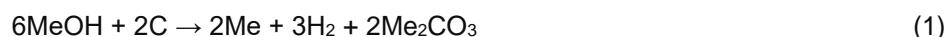
Keywords: cigarette butts, activated carbon, chemical activation, Langmuir isotherm, Freundlich isotherm

INTRODUCTION

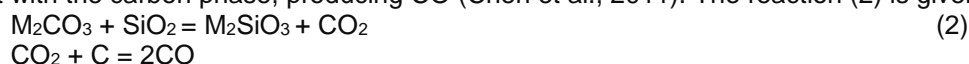
Cigarette Butts are commonly used to prevent smokers from inhaling harmful substances by blocking the largest tar particles as well as other toxic substances. Although CB wastes are non-biodegradable, they will not remain in the environment indefinitely. The sunlight will eventually degrade it into small pieces, releasing harmful and toxic substances into the environment, and causing harm to humans, animals, and aquatic life. In recent years, cigarette sales have reached 5.8 trillion cigarettes per year, resulting in 800,000 metric tonnes of discarded CB waste being thrown everywhere (Blankenship & Mokaya, 2017). Even if it is thrown in the garbage, it never mixes with the ground because of biodegradability. As a result, recycling CB wastes can help to alleviate the problem of waste disposal, which has piqued the interest of scholars. Previous research has found that CBs can be recycled as a raw material in brick production (Binti Abdul Kadir & Mohajerani, 2012), a biofilm carrier in an integrated fixed film activated sludge (IFAS) system (Sabzali, Nikaeen & Bina, 2012), a sound-absorbent (Gómez Escobar & Maderuelo-Sanz, 2017), and also cigarette butt remaining cellulose acetate can be used as cellulose membrane-based separator for high-performance lithium-based battery (Huang et al., 2015). Based on the industrial analysis, the carbon content of smoked cigarette filters is 58% (Zhang et al., 2020). As a result, cigarette butt can be a good raw material for activated carbon filters due to having high carbon content. Industrial activated carbon made from common precursors like coal, wood, coconut shell, and peat is very costly and also there is a problem associated with regeneration. As a consequence, low-cost precursor materials have become an attractive choice in the production of activated carbon. One of these low-cost precursors that have been used in the production of AC (activated carbon) in recent years is *Ulva Lactuca* (Ibrahim et al., 2016), bamboo (Lo et al., 2012), olive stone (Bohli et al., 2015), cattle bone (Cechinel et al., 2014), and orange peel. Cigarette butts being one

of the largest wastes, as well as having a large carbon atom, it can be used as a new raw material for making AC, opening up a new path for recycling cigarette filter wastes and utilizing them in the water treatment process. Used cigarette filters and $ZnCl_2$ as a chemical activating agent producing activated carbon got a specific surface area of $479\text{ m}^2\text{g}^{-1}$ (Conradi et al., 2019).

The CFAC can be obtained either by physical activation or by chemical activation after physical activation. Activated carbon made by physical activation has a lower specific surface area compared to the chemical activation process. During physical activation, the CBs are carbonized in a tabular furnace at certain temperatures which usually range from 65°C - 900°C under an argon atmosphere. During chemical activation, the CMs (carbon materials) are impregnated into chemical constituents like KOH, NaOH, $ZnCl_2$, and HNO_3 which increases the porosity and surface to make the desired activated carbon. Previous research has revealed that among all chemical activating agents, alkali hydroxides (specifically NaOH and KOH) can be appealing activating agents for the preparation of microporous carbon due to the production of valuable materials (Freitas et al., 2007). Active carbon from cigarette filter waste using KOH activation obtained a high specific surface area of $1892\text{ m}^2\text{g}^{-1}$ (Polarz et al., 2002). The overall process is given in reaction (1).



Overall, the reaction produces alkali carbonate, hydrogen, and alkaline metal, where $\text{Me} = \text{K}$ or Na (Guo et al., 2002). The detection of metal traces $\text{Me} = \text{K}$ or Na (Guo et al., 2003; Lillo-Ródenas et al., 2004) confirms the proposed reaction (Lillo-Ródenas et al., 2003; Daz-Terán et al., 2003). Depending on the reactivity of the carbon precursor, hydrogen can be detected at temperatures ranging from 200°C to 600°C (Lillo-Ródenas et al., 2004). CO or CO_2 can be detected due to the decomposition of Me_2CO_3 and the devolatilization of the carbon precursor itself at temperatures above 800°C . Carbon material's porosity increases due to interaction with SiO_2 , which produces alkaline silicates and CO_2 . The CO_2 will then react with the carbon phase, producing CO (Chen et al., 2011). The reaction (2) is given below:



Meanwhile, in many countries around the world, groundwater is one of the potential natural resources for human consumption. However, drinking water sources in developing and underdeveloped countries are seriously threatened by contamination issues caused by both man-made and industrial activities. Groundwater is the primary source of drinking water in Bangladesh, but there are some issues with groundwater in the southern part of the country due to excess Fe, Mn, As, and salinity. One of the most serious issues with groundwater is its reddish and blackish color due to high levels of iron (Fe) and manganese (Mn) (Du et al., 2017; Dou et al., 2018). The primary sources behind the presence of Fe and Mn are due to weathering process of natural minerals in the soil, sediment, and bedrock in groundwater. Furthermore, excessive Fe and Mn levels in groundwater can occur as well as a result of rain filtration through soil, sediment, and rocks. Though iron is not a dangerous element, high levels of Fe associated with water quality issues can lead to plumbing fixtures, bad-tasting water, and hemochromatosis (Hussin et al., 2014). Excess Fe in drinking water and water supply systems causes a problem with the brown-reddish color of the water. Furthermore, chronically consuming large amounts of iron can result in iron overload. The permissible limit for iron in water is 0.3 mg/L according to WHO and $0.3\text{-}1.0\text{ mg/L}$ according to Bangladesh standards. Depending on the iron concentration, removal is necessary for good drinking water quality. Several heavy metal removal techniques have been described by many researchers, including adsorption, precipitation, oxidation/filtration, and electrochemical and ion exchange methods (Chaturvedi and Dave, 2012; Patil et al., 2016). Among them, adsorption is the most convenient and inexpensive method for removing iron and manganese. Therefore, considering the cost-effective method, using cigarette butts as activated carbon can be a double-edged solution for both groundwater treatment and cigarette butt recycling.

The main purpose of this article is to determine the effectiveness of cigarette butts as activated carbon for groundwater treatment using a filter made from cigarette butts based activated carbon and compare it with both sand and charcoal filter to know the removal efficiency of iron, manganese, and other pollutants. In addition, the adsorption capacity and adsorption isotherm of CFAC for methylene blue was also determined. Finally, the adsorption isotherm was analyzed to know the sorption process of adsorbent, and compiling all the work, conclusions, and recommendations were outlined.

EXPERIMENTAL

Materials

The abandoned smoked cigarette butts (CBs) were collected from random streets and waste bins. For the experiment, two sources of contaminated water were used where the iron water was collected

from a shallow tube well of Arang Ghata, Khulna, and chlorine water was collected from tap water. All other chemicals including potassium hydroxide solution (KOH), methylene blue solution (MB), silver nitrate, lead acetate, and other reagents were collected from Environment Engineering Laboratory, KUET. Instruments like a magnetic stirrer, muffle furnace, pH meter, spectrometer, and turbidimeter are also collected from that lab.

Preparation and Modification of CFAC

Figure 1 depicts a schematic illustration of the CFAC synthesis process. To begin, the smoked cigarette butts were cleaned with water after being removed from the wrapping paper to avoid contamination with tobacco or soot. After that, the CBs were oven dried for one day at 105°C. The CBs loaded in a steel chamber were then placed in a muffle furnace for carbonization at 500°C for 4 hours in an argon atmosphere. After carbonization, the desired charcoal was made due to physical activation. To create the desired activated carbon, the carbonized materials (CM) were impregnated for 24 hours in various KOH solutions (mass ratio of CM/KOH = 1:1.5, 1:2, 1:1.2.5. Then the solution with the carbonized materials was filtered and washed with distilled water. The carbonized materials were again heated in the oven at 105°C. Finally, the desired activated carbon had been obtained by this procedure and designated by CFAC-1.5, CFAC-2, and CFAC-2.5 as shown in Figure 1.

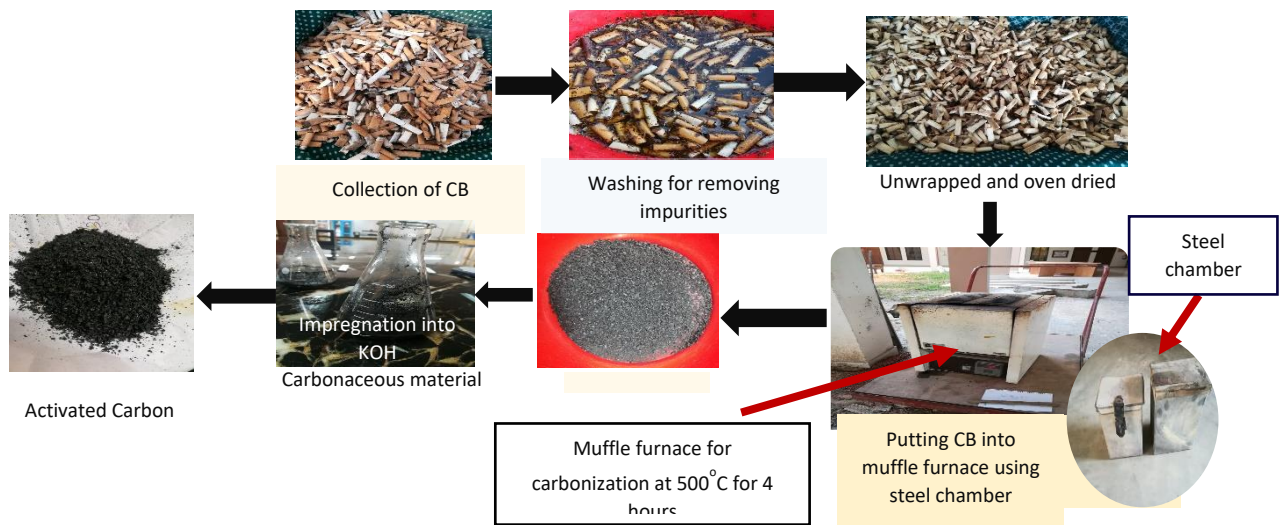


Figure 1: Schematic diagram of the preparation of activated carbon from cigarette butts

In Figure 2, the overall research strategy is depicted.

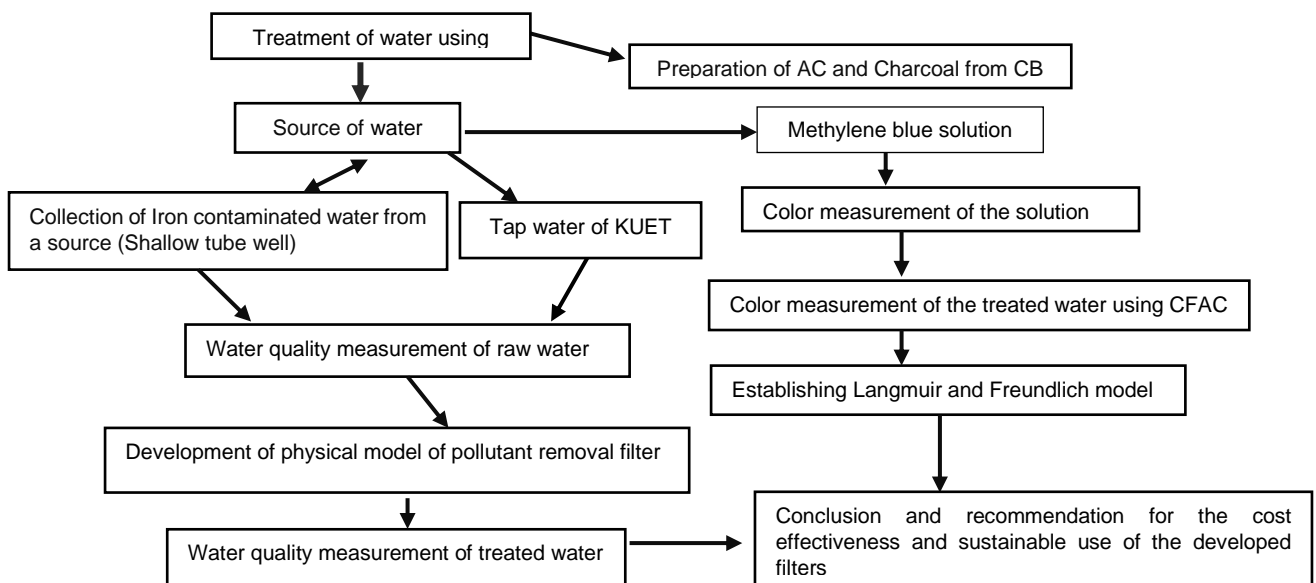


Figure 2: Schematic diagram of overall research strategy

Filtration Model Development and Various Water Quality Parameters

There were 3 designed filter media. Filter media using

- 1) Filter 1: Sylhet sand (2.5 cm) and Kustia sand (2.5cm)
- 2) Filter 2: Sylhet sand (2.5 cm), charcoal (5cm) from cigarette filter, and Kustia sand (2.5cm)
- 3) Filter 3: Sylhet sand (2.5 cm), activated carbon from cigarette filter (5cm), and Kustia sand (2.5 cm)

A simple cylindrical filtration tube had been developed with the following dimension:

- Length of the tube= 100 cm
- Inside diameter of the tube = 0.5 cm
- Outer diameter of the tube = 0.75 cm
- The base is covered with a clean piece of cloth.
- The upper portion of the cylindrical tube is open.

The setup procedure had described below:

The first filter was made with Sylhet and Kustia sand. The other two filter media was made of charcoal and CFAC-1.5 to compare the effectiveness of these three filter media. Filter 2 and filter 3 also had Sylhet and Kustia sand but the difference was one had the charcoal and the other had activated carbon with the same thickness which is 5 cm.

For the analysis, the iron, manganese, and chloride water had been passed through the three filters where the rate of flow was measured at 100mL/min. Different water quality parameters (related to the research) of the collected raw water before filtration and treated water after filtration such as pH, Color, Turbidity, Iron, Chloride, Arsenic, Manganese, Hardness, Total dissolved solids, Total coliform, and Faecal coliform were measured.

Batch Adsorption Experiments

CFAC batch adsorption experiments were carried out using methylene blue solutions. For this analysis, a 0.002 mg/ml methylene blue solution was prepared and mixed with various concentration ranges (0.2-1.4g) of CFAC-2.5 and CFAC-2 before being placed in a magnetic stirrer until equilibrium was reached. After reaching equilibrium, the solution was separated from the CFAC, and the color was determined using a HACH Spectrophotometer, model DR 2700. The adsorption equilibrium capacity was calculated using equation (1) (Xu et al., 2016; Hu et al., 2019).

$$q_e (mg/g) = \frac{(C_0 - C_e)V}{M} \quad (1)$$

where q_e (mg/g) was equilibrium adsorption capacity, C_0 (mg/L) is the initial concentration of adsorbate, C_e (mg/L) is the adsorbate's concentration after adsorption by CFAC, V (L) is the solution's solution volume, M (g) is the mass of CFAC.

The adsorption isotherm of CFAC was analyzed by two models:

- Langmuir Isotherm
- Freundlich Isotherm

RESULTS AND DISCUSSIONS

Adsorption Capacity and Effect of Adsorbent Dosage

The adsorption capacity means the amount of adsorbate taken by the adsorbent per unit mass of adsorbent. Table 1 represents the comparison of the adsorption capacity of CFAC-2 and CFAC-2.5 for methylene blue adsorption where the adsorbent dosage and initial concentrations were the same.

Table 1 Adsorption capacity of CFAC-2 and CFAC-2.5

Sample	Amount of CFAC	Initial Concentration	Final Concentration	Adsorption Capacity	Sample	Initial Concentration	Final Concentration	Adsorption Capacity	
CFAC-2(CM:K OH=1:2)	0.2		17180	660	CFAC-2 (CM:K OH=1:2.5)		16500	1000	
	0.5		15500	600			14500	800	
	0.8	18500	14100	550			18500	13700	600
	1.1		13550	450				12989	501
	1.4		12270	445				12900	400
	1.7		11700	400				12414	358

The result found from the table is that CFAC-2.5 has the highest adsorption capacity due to a higher concentration of chemical activating agent KOH which has increased the porosity. Furthermore, it was notable that, the higher the CFAC dosage, the lower the adsorption capacity, as the adsorbent dosage is inversely proportional to adsorption capacity according to equation (1). The effect of adsorbent dosage on adsorption capacity had been graphically represented in Figure 3.

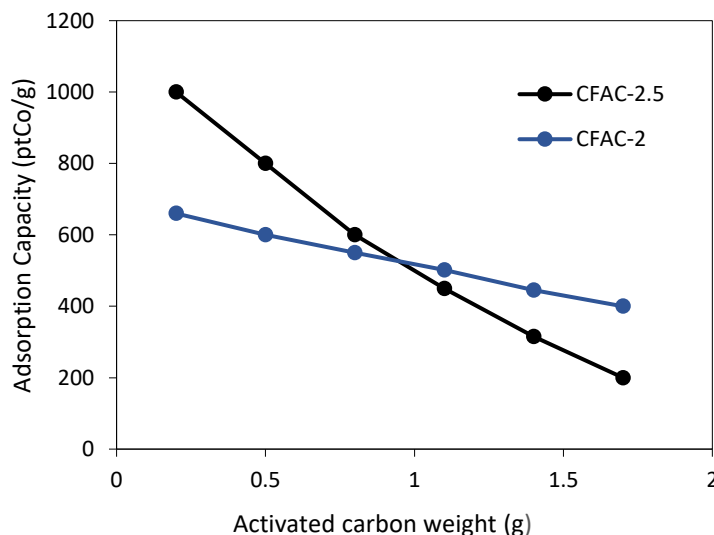


Figure 3: Effect of concentration of CFAC on Adsorption Capacity

Treatment of Groundwater Using Different Filter Media

Table 2 compared the water quality parameter of raw water to the groundwater sample after being run through three different filter media, Filter-1 (Sand filter), Filter-2 (Charcoal filter from CBs), and Filter-3 (CFAC-1.5) to demonstrate the efficiency of the filters. According to Table 1, the charcoal and activated carbon filters from CBs remove more of these parameters than the sand filter, with the activated carbon filter having the highest removal efficiency. After passing through an activated carbon filter, only iron (0.02mg/L), turbidity (6.97 mg/L), pH (7.16), chloride (67.5 mg/L), manganese (0.4 mg/L), and hardness (450 mg/L) were within the allowable limits according to Bangladesh standards, and the rest of the parameters were close enough to that limit.

Table 2 Comparison of three different filter media for groundwater treatment

Properties	Raw Water	Sand Filter (Filter-1)	Charcoal filter made from cigarette butts (Filter-2)	Activated carbon filter made from cigarette butts (Filter-3)	WHO Guideline Values	Bangladesh Standard Values
Color (PtCo)	300	158	71	19	15	15
Turbidity (mg/L)	30.1	29.6	8.58	6.97	5	20
Total Dissolved Solid (mg/L)	1810	1200	900	700	500	500
pH	7.52	7.45	7.17	7.16	6.5-8.5	6.5-8.5
Iron (mg/L)	1.65	0.08	0.05	0.02	0.3	0.3-1.0
Chloride(mg/L)	435	433.3	70	67.5	250	150-600
Manganese(mg/L)	1.2	1.2	0.9	0.4	0.3	1.0
Hardness(mg/L)	703.76	675.98	685.24	450.26	120-170	200-500
Total Coliform(TC)	0	0	0	0	-	-
Fecal Coliform	0	0	0	0	-	-

Removal Efficiency of Different Filter Media

Removal efficiency means how much organics wastes can be removed by that particular material. The removal efficiency of different filter media used such as CFAC-1.5, charcoal from cigarette butts, and sand in % had described here in Table 3.

Table 3 Removal efficiency (%) of different filter media used

Filter Media	Removal Efficiency(%)							
	Water Quality Parameters	Color	Turbidity	TDS	Chloride	Iron	Manganese	Hardness
Sand filter		47.3	1.66	11	0.39	95	0	4
Charcoal filter		76	71	50	84	96	25	2.63
Activated Carbon Filter		94	77	61.3	97	99	33	36

The primary goal of this study was to use CFAC to remove iron and manganese from groundwater. According to Table 3, the activated carbon filter from Cigarette Butts was 99% effective at removing iron from groundwater, while the charcoal filter and sand filters were 96% and 95% effective, respectively. As shown in Table 3, the activated carbon filter from CBs was also more effective than the other two filter media in removing other parameters such as manganese, color, turbidity, TDS, and hardness.

Methylene Blue Adsorption Isotherm of CFAC:

The significance of the adsorption isotherm is to express the relation between adsorbent and adsorbate about how the adsorbate accumulated on the surface of adsorbents. Furthermore, the adsorption isotherm data is important for developing an equation that accurately represents the results and can be used for design purposes, where the equation is dependent on the nature and type of the system (Chen,2011).

To explore the adsorption mechanism, the adsorption isotherm of MB was fitted to Langmuir and Freundlich isotherm. The Langmuir equation (2) is based on the assumptions of homogenous adsorption on the adsorbent surface with the same adsorption site and no transmigration of adsorbate in the plane of the surface. Moreover, there will be no steric hindrance between the adsorbed molecules and incoming ions. The linear equation of Langmuir isotherm can be shown as:

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m K_L} \tag{2}$$

Where q_e (mg/g) is the equilibrium adsorption amount, C_e (mg/L) is the equilibrium concentration of the solute, q_m (mg/g) and K_L (L/mg) are constants related to the adsorption capacity and energy of adsorption. The values of q_m and K_L is calculated from the slope and intercept of the Langmuir plot of C_e versus C_e/q_e .

The Freundlich equation (3) is based on the assumption of surface heterogeneity and an exponential distribution of active sites of adsorbents along with their energies towards adsorbate (Clayton, 1926).

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_e \tag{3}$$

Where K_F is roughly an indicator of the adsorption capacity and n is the heterogeneity factor.

As shown in Figure 4, the obtained experimental data for MB were fitted into Langmuir and Freundlich isotherm linearly where the Langmuir and the Freundlich constants are determined from the graph.

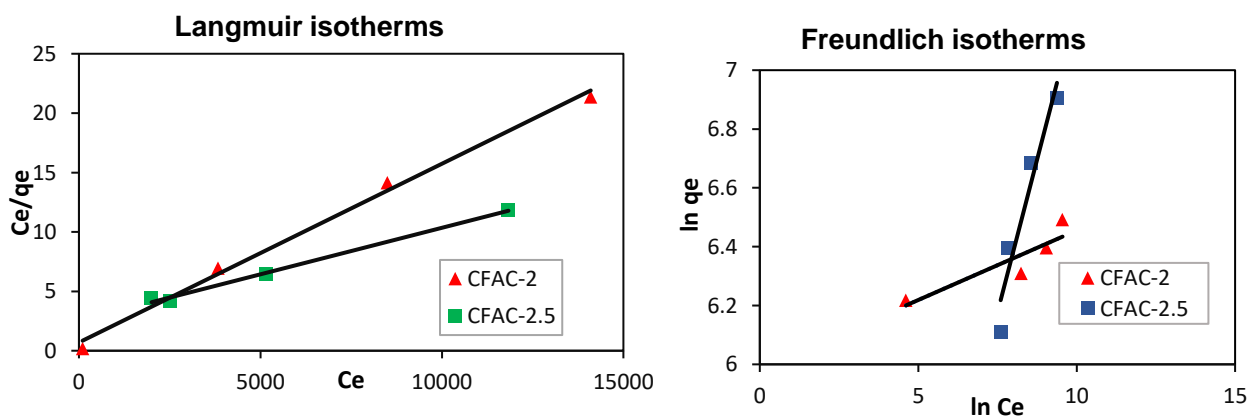


Figure 4 Adsorption isotherms (a) Langmuir isotherms (b) Freundlich isotherms

The data of Langmuir and Freundlich isotherm were also summarized in Table 4. According to the table, it was depicted that the methylene blue adsorption process of CFAC was more biased towards Langmuir isotherms than the Freundlich isotherms since the correlation coefficient value of Langmuir isotherm ($R^2 \approx 0.99$) was higher than Freundlich isotherm. As a result, it could be finalized from the table that the methylene blue adsorption on the CFAC surface could be a monolayer sorption process.

Table 4 The parameters of the Langmuir and Freundlich isotherm equation

Adsorbate	Samples	Equations	Langmuir model			Equations	Freundlich model		
			q_m (mg/g)	K_L	R^2		K_F	n	R^2
Methylene blue	CFAC-2	$q_e/C_e=0.0015$ $C_e-0.6947$	667	0.0021 5	0.994 2	\ln $q_e=0.0473\ln C_e+5.9$ 822	39 6	21. 1	0.811 5
	CFAC-2.5	$q_e/C_e=0.0008$ $C_e-2.5158$	1250	0.0003 2	0.996	$\ln q_e=0.41561\ln$ $C_e+3.0588$	21	2.4	0.925 6

CONCLUSIONS AND RECOMMENDATIONS

In the case of filter media, activated carbon prepared from cigarette butts, the removal efficiency for treatment of water was found iron-99%, manganese-33% color-94%, turbidity-77%, TDS-61.3%, chloride 97%, hardness-36%. According to WHO guidelines and Bangladesh standards, only iron (0.02mg/L), turbidity (6.97 mg/L), pH (7.16), chloride (67.5 mg/L), manganese (0.4 mg/L), and hardness (450 mg/L) were within the allowable limit after passing through the activated carbon filter. In the case of filter media charcoal prepared from cigarette butts, the amount of manganese was found 0.9 mg/L which is not within the allowable limit but close to it. From the filter media of charcoal, only iron(0.05 mg/L), and pH (7.17) were within the allowable limit. In the case of filter media sand, only iron (0.08 mg/L), and pH (7.45) were within the allowable limit. From the adsorption isotherm analysis, the adsorption isotherm of CFAC for methylene blue adsorption process was more biased toward Langmuir isotherm due to a higher correlation coefficient ($R^2 \approx 0.99$) than the Freundlich isotherm, indicating monolayer adsorption process of CFAC. Finally, the overall study found that activated carbon filters made from cigarette butts removed more pollutant parameters than the others. As a result, it could be a better low-cost treatment option for groundwater treatment and in that way, cigarette butts wastes could also be recycled.

Based on the performance study of a developed filter, the following recommendations can be made for future work:

- Since adsorption capacity depends on a variety of factors, a study needs to be done on factors like surface area, pH, time, and adsorption kinetics on CFAC.
- The Study needs to be done on the modification of CFAC.
- The Study needs to be done on the performance of the developed filters by designing a new treatment unit for use at the household or community level. The double filtration process can be adopted for obtaining better efficiency.
- The manganese and hardness removal efficiency is relatively low. So, high saline and hard water should not be used.
- The Study needs to be done to know how the filter can be reused.
- A future study should be done on clogging after replacing filter bed materials is needed.
- The long term research should be required to ensure the filtration rate as well as filter run time.
- Last but not the least, a study needs to be done on the use of CFAC at an industrial scale. Though the amount of CFAC produced at an industrial scale will be small in number, it is worth producing CFAC at an industrial scale to keep the environment safe. So, the Government should take the necessary steps on it.

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