

Comparative Study On The Adsorption Of Chromium Cr(VI) From Wastewater By Using Different Adsorbents.

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ABSTRACT:

Water that contains elevated levels of chromium, specifically the toxic form of elements generally refers to chromium-contaminated water. Chromium is a major source of aquatic pollution in the states of Tamil Nadu, Uttar Pradesh, and West Bengal of India. This experimental study is focused on the removal of chromium with the help of adsorption. Fly ash and activated carbon were used here as adsorbents. This study also checks the impact of several factors including initial concentration, adsorbent dose, contact time, and pH value of the solution on removal efficiency. To analyze the adsorption process, Langmuir and Fredundlich isotherm models were used. This study shows that activated carbon is the better adsorbent as compared to fly ash with an optimum removal efficiency of 85.6%.

Key Words: Chromium, Fly ash, Activated Carbon, Langmuir isotherm models, Fredundlich isotherm models.

Date of Submission: 05-09.2023

Date of acceptance: 16-09-2023

I. INTRODUCTION:

Pollution of water due to the presence of heavy metal ions is a significant environmental concern with far-reaching ecological and health implications. Heavy metals such as lead, mercury, cadmium, chromium, etc. can have harmful effects on ecosystems and human health depending on the specific metal and the level of exposure. These metals enter the environment through various sources, including industrial processes, mining activities, agricultural runoff, and improper electronic waste and battery disposal. When high concentrations of heavy metals leach into water bodies, they can alter the pH and chemical composition of water, making it toxic for aquatic organisms. In view of their toxicity mechanisms, non-biodegradability nature along with their health effects, their removal becomes absolutely necessary in order to protect our ecosystems and human wellbeing.

Chromium (Cr) is a chemical element with the atomic number 24. It is a transition metal known for its shiny, silver-grey appearance. Chromium is a naturally occurring element found in the Earth's crust, and it exists in different oxidation states, with the most common forms being trivalent chromium Cr(III) and hexavalent chromium Cr(VI). Both forms can contribute to water pollution, but hexavalent chromium is generally more toxic and of greater concern. Surface and ground water contamination due to metal chromium, especially hexavalent chromium, can have serious environmental and health consequences these days. Hence, it is essential to monitor and control the levels of chromium in water and take effective regulation and proper waste management to mitigate the impacts of chromium contamination in water.

The most common source of chromium pollution in water is from industrial processes. Industries such as electroplating, leather tanning, and metal manufacturing use chromium compounds, and the wastewater generated during these processes can contain high levels of hexavalent chromium (Cr (VI)). If not properly treated before discharge, this wastewater can contaminate nearby water bodies. Also, it can leach into groundwater and surface water when disposed of improperly in landfills or waste disposal sites. In some areas, chromium can be naturally present in rocks and soils at elevated levels. Under certain conditions, such as when groundwater comes into contact with these geological formations, it can pick up elevated levels of chromium, both Cr (III) and Cr(VI).

Exposure to hexavalent chromium, especially at high concentrations, can have adverse health effects. It is a known human carcinogen when inhaled, particularly in the form of hexavalent chromium compounds like chromium (VI) oxide or chromates. Ingesting or drinking water contaminated with hexavalent chromium can also have health implications. Prolonged exposure to high levels of hexavalent chromium in drinking water has been associated with an increased risk of lung, nasal, and sinus cancers. Contact with hexavalent chromium can lead to skin rashes and irritation. It can also cause gastrointestinal problems, such as stomachaches, vomiting, and diarrhea.

The U.S. Environmental Protection Agency (EPA) has set a Maximum Contaminant Level (MCL) for total chromium (which includes both trivalent and hexavalent chromium) in drinking water at 0.1 milligrams per

liter (mg/L) or 100 parts per billion (ppb). This limit is intended to protect against potential health risks associated with hexavalent chromium exposure.

Removing chromium from wastewater is important to prevent environmental contamination and potential health hazards. There are various physicochemical treatments and biological treatments have been widely used for the removal of chromium from wastewater. Physicochemical treatments include ion exchange, adsorption, reverse osmosis, membrane filtration, electrocoagulation, and conventional sedimentation. Among these methods, adsorption is one of the most effective and versatile treatments for removing metals like chromium from wastewater. This process involves the attachment of heavy metal ions to the surface of a solid material, called an adsorbent. The adsorbent material has a high surface area and is typically porous, allowing it to attract and trap heavy metal ions from the wastewater. Over time, an equilibrium is reached as heavy metal ions continue to attach to the adsorbent's surface. At this point, the adsorbent becomes saturated, and its capacity to adsorb more heavy metal ions decreases. Once the adsorbent is saturated, it can be separated from the wastewater. This can be done through simple filtration or other separation techniques. The effectiveness of adsorption for heavy metal removal depends on several factors, including the choice of adsorbent, contact time, pH of the wastewater, temperature, and the initial concentration of heavy metals in the wastewater. It is a widely used and efficient method for treating industrial wastewater and mitigating the environmental impact of heavy metal pollution. The present study mainly focuses on removing chromium from wastewater by using fly ash and activated carbon as low-cost adsorbent. This study also checks the impact of factors like initial concentration of chromium, adsorbent dose, contact time, and pH of the solution on removal efficiency. There are several isothermal models conducted in this experimental work i.e. (Langmuir isotherm model, Freundlich isotherm) to analyze the adsorption process. Several calibration curves were also prepared to maintain high accuracy.

II. MATERIALS AND METHODS:

2.1 Adsorbent

2.1.1 Fly ash:

Fly ash is a fine powder consisting of spherical, glassy particles that are produced as a byproduct of burning pulverized coal in electric power generation plants. It is one of the most commonly generated industrial wastes globally, particularly in countries that rely on coal for electricity generation. Fly ash is primarily composed of silicon dioxide (SiO₂), aluminium oxide (Al₂O₃), iron oxide (Fe₂O₃), and calcium oxide (CaO), along with various other trace elements. Its chemical composition varies depending on the source of coal and combustion conditions. Fly ash particles are very small in size, which increases the surface area of fly ash. Due to this surface area, fly ash is very suitable as an adsorbent in the adsorption process. Table 2.1 and Table 2.2 show the physical and chemical properties of fly ash respectively.

Table 2.1- Physical properties of fly ash

PROPERTIES	RESULTS
Color	Grey
Specific gravity	2.22
ParticleSize	<300μ
pH	8-9

Table 2.2- Chemical properties of fly ash

COMPOUNDS	COMPOSITION(WT.%)
SiO ₂	63.63
Al ₂ O ₃	25.85
TiO ₂	0.46
Fe ₂ O ₃	5.13
MgO	0.66
CaO	2.54
K ₂ O	0.04
Na ₂ O	0.05
SO ₃	1.64

2.1.2 Activated carbon:

The activated carbon is a black residue which forms after burning carbonaceous source materials such as coconuts, coal, peat, wood etc. It is also known as activated charcoal, which is a highly porous form of carbon that is widely used for various applications due to its exceptional adsorption properties. This process removes volatile compounds and creates a network of small pores on the surface of the carbon, increasing its surface area and making it highly adsorbent. Table 2.3 and Table 2.4 show the physical and chemical properties of activated carbon respectively.

Table 2.3- Physical properties of activated carbon

PROPERTIES	RESULTS
Colour	Black
Specific gravity	1.25
Particle Size	<300µ
pH	7.54

Table 2.4- Chemical properties of Activated carbon

COMPOUNDS	CONTENT (%)
Carbon	99.84
Zinc	0.01
Ash	0.15

2.2 Preparation of raw adsorbents:

Activated carbon was purchased from the local market and then it was ground into powdered form. The particles were further sieved through a 300µm sieve and dried in the oven at 105⁰c for 1 hour. The fly ash was collected from the local power plant and then it was dehydrated with the help of an oven at 105⁰c for 1 hour.

2.4 Preparation of adsorbate solution:

The stock solution of hexavalent chromium Cr(VI) of concentration 1000 ppm is prepared by dissolving CrO₃ in distilled water. The stock solution was further diluted with distilled water to 100 concentrations to obtain the standard solutions for absorbance measurements. The initial pH is adjusted by using HNO₃ or NaOH solutions.

2.3 Batch experiments:

All the adsorption experiments were carried out in a batch mode with the help of 100 ml conical flasks with a stopper containing 50 ml of standard chromium solution Cr(VI) at known concentration and pH. The orbital shaker was used to shake the solution uniformly agitated at 200 rpm after adding the adsorbents to the solution. All the batch experiments were conducted at room temperature. After completion of the adsorption process, the solutions were filtered with the help of watt man filter paper of size 45µm. After filtration, the final concentration of Chromium samples was determined with the help of UV-spectrophotometers. The absorbance of the light beam was measured with a wavelength of 540 nm after 45 to 60 min. The test was conducted at different values of parameters like pH values, initial concentration, temperature, adsorbent dosage, and contact time and observed the effect of parameters on the adsorption process. The parameters varied in the experiments are the Cr(VI) initial concentration ranging from 4 mg/l to 24 mg/l, the pH ranges from 2 to 12, the range of contact time was taken from 30 to 150 min, effect of different adsorbent dosage examined by varying its value i.e. (1 mg/l, 5 mg/l, 10 mg /l, 15 mg/l, 20 mg/l, 25 mg/l, 30 mg/l, 35 mg/l, 40 mg /l, 45 mg/l, 50 mg/l)

The following equation were used to evaluate the amount of Cr(VI) adsorbed per unit mass of the adsorbent's percentage of removal = $\frac{C_0 - C_e}{C_0} \times 100$

sorption capacity (mg/g)_e = $\frac{(C_0 - C_e) \times V}{M}$

C₀ = Initial concentration of chromium in (mg/l)

C_e = Equilibrium concentration of chromium in (mg/l)

V = Volume of chromium solution for adsorption in ml

M = Weight of adsorbent dose in gram

2.6 Adsorption Isotherms and Its Studies:

Adsorption isotherms are graphical representations or mathematical models that describe the relationship between the amount of a substance adsorbed onto a solid surface and the pressure of that adsorbate at a constant temperature. Several isotherms' models are available such as Langmuir, Freundlich, Redlich-Peterson, Koble-Corrigan, Tempkin, etc. In this experimental work, the analysis was conducted with the help of the Langmuir, Temkin isotherms model.

2.4 Langmuir isotherm model:

Langmuir adsorption isotherm model assumes that adsorption energies are uniform over homogenous sites (identical sites having an equal affinity for adsorbate). It presumes occurrence of monolayer adsorption of adsorbent on the outer surface and considers nil lateral interactions and transmigration amongst adsorbed molecules in the plane of the surface. Once adsorption takes place the adsorbate molecules become immobilized and no adsorption takes place after that. In this model, the energy of adsorption is considered to be constant. The Langmuir isotherm is expressed as:

$$\text{Adsorption capacity (mg/g)} q_e = \frac{(C_0 - C_e) \times V}{M}$$

The Langmuir isotherm express in a linear form i.e

$$\frac{C_e}{q_e} = \frac{1}{bQ_m} + \frac{C_e}{Q_m}$$

Where q_e = Adsorption capacity (mg/g)

b = Energy constant or Langmuir Isotherm constant

Q_m = Maximum adsorption capacity.

The Langmuir graph is plotted between $\frac{C_e}{q_e}$ value and C_e value. Here the C_e/q_e value taken along the y-axis and C_e value taken along the x-axis. The slope of the graph gives the $1/Q_m$ value.

There are certain assumptions in Langmuir isotherm, which are mentioned below.

1. Soluble molecules adsorbed on a fixed number of sites.
2. Maximum adsorption, the layer should be one molecule thick.
3. The rate of adsorption is directly proportional to the no of sites unoccupied.
4. The rate of desorption is directly proportional to the no of the site occupied.
5. The sorption energy is the same at all sites.
6. Adsorption may be reversible.

The essential characteristics of Langmuir isotherm can be expressed in terms of a dimensionless separation factor R_L , which describe the type of isotherms and is defined by,

$$R_L = \frac{1}{1 + bC_0}$$

The parameters indicate the shape of the isotherm accordingly, if the R_L value lies between 1 to 0, then favorable adsorption is indicated. If, R_L value is greater than 1, unfavorable adsorption, while a value of 1 represents linear & unfavorable and 0 represents irreversible.

2.5 Freundlich Isotherm

The model is generally used to define adsorption over the non-homogeneous surface. The empirical equation predicts multilayer adsorption due to difference in affinity over adsorption sites. Because of such dissimilarity, the isotherm model likewise assumes variation in surface energy of adsorption also. An adsorption system that obeys this model indicates that the adsorption sites are larger in number as compared to pollutant molecules/ions to be removed. The linearized form of Freundlich isotherm is represented by following mathematical equation

$$q_e = K_f(C_e)^{1/n}$$

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e$$

Where, K_f is the Freundlich characteristics constants and $1/n$ the heterogeneity factor of adsorption, obtained from intercept and slope of $\ln(q_e)$ vs $\ln(C_e)$ linear plot respectively. The value of K_f is an indicator of adsorption capacity and thus can be used for relative measurement of the surface area. b and $1/n$ are related to enthalpy and intensity of the adsorption. $1/n$ value should be less than unity for high adsorption capacity.

III. Results and Discussion:

3.1 Comparison according to adsorbent dose:

In this analysis, the comparison of removal efficiency of chromium was checked by varying the dose of fly ash and activated carbon. The other parameters like initial concentration, pH value and contact time were maintained as constant. Here the initial concentration of the solution was maintained at 4 mg/L in both the cases. The pH was set at 6.5. The comparison result was plotted in a graph shown in figure 1.

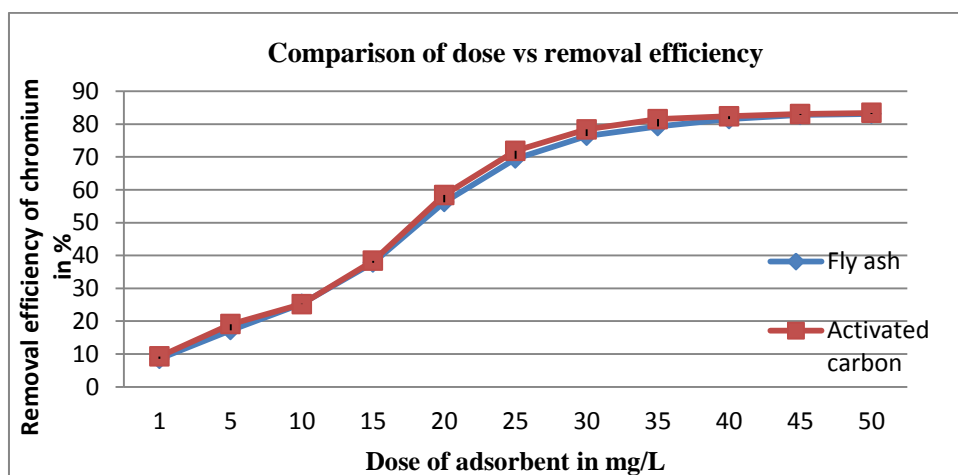


Fig 1: Comparison graph between the dose of adsorbent vs. removal efficiency of chromium

The above graph shows that the range of fly ash and activated carbon dose varied from 1 gm/L to 50 gm/L. The equilibrium removal efficiency of 79.32% and 81.51% was achieved at a dose of 35 gm/L by using fly ash and activated carbon as an adsorbent respectively. From this analysis, it was clear that the activated carbon gives better removal efficiency as compared to fly ash.

3.2 Comparison according to initial concentration:

The comparison of the removal efficiency of chromium was conducted by varying the initial concentration of chromium in synthetic water solution. Here the dose of fly ash and activated carbon was taken as 35 gm/L solution. The pH was maintained at 6.5 in both cases. The result of this comparative analysis was plotted in figure 2.

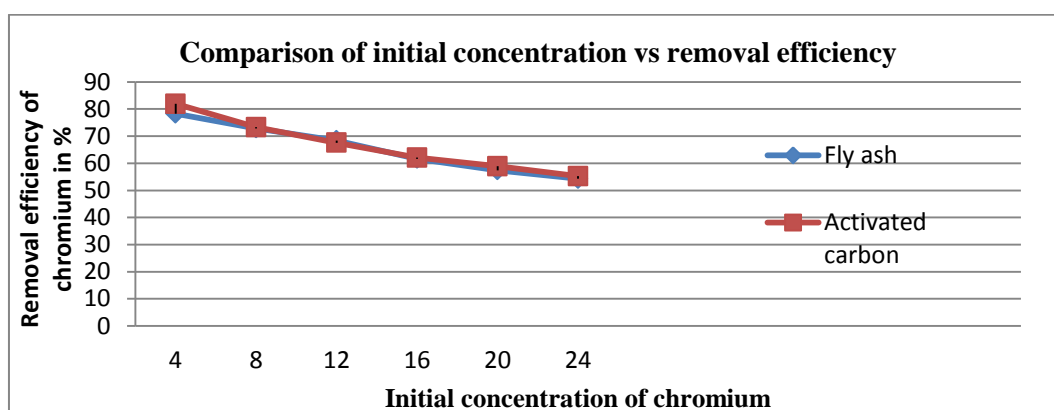


Fig 2: Comparison graph between the initial concentration vs removal efficiency of chromium

The above figure shows that chromium's maximum removal efficiency, i.e., 78.29 %, was achieved at an initial concentration of 4 ppm when fly ash was used as an adsorbent. Similarly, chromium's maximum removal efficiency, i.e., 81.92 %, was achieved at an initial concentration of 4 ppm, when activated carbon was used as an adsorbent. In both cases, the removal efficiency of chromium was decreased by increasing the concentration of chromium in the synthetic water solution.

3.3 Comparison according to contact time:

The removal efficiency of chromium was examined by different contact time intervals. The range of contact time varied from 30 minutes to 150 minutes in both the case. Here the dose of adsorbent was maintained

at 35 gm/L synthetic solution. The initial concentrations of chromium were maintained 4 ppm for both fly ash and activated carbon. The pH value of the synthetic solution was maintained at 6.5 in both cases. The result of the above analysis was plotted in a graph, which shown in figure 3.

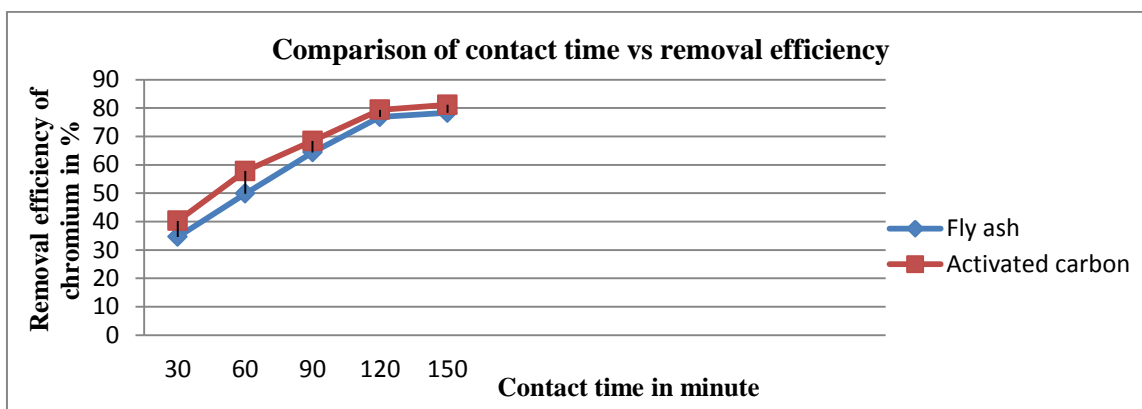


Fig 3: Comparison graph between contact time vs removal efficiency of chromium

The above graph shows that the maximum removal efficiency of chromium was achieved at 150 minutes of contact time in both cases. In the case of fly ash, 78.32 % of chromium was removed from the synthetic solution at 150 minutes of contact time. Similarly, in the case of activated carbon 81.13 % removal efficiency of chromium was achieved at 150 minutes of contact time.

3.4 Comparison according to pH value of the wastewater:

The compression of the removal efficiency of chromium was checked by varying the pH value of the synthetic solution. The initial concentration of the solution 4 ppm was maintained for both fly ash and activated carbon. A dose of 35 gm/L solution was used in both the case. The range of the pH of the solution was varied from 2 to 12. The result of this comparative analysis was plotted in a graph, which shown in figure 4.

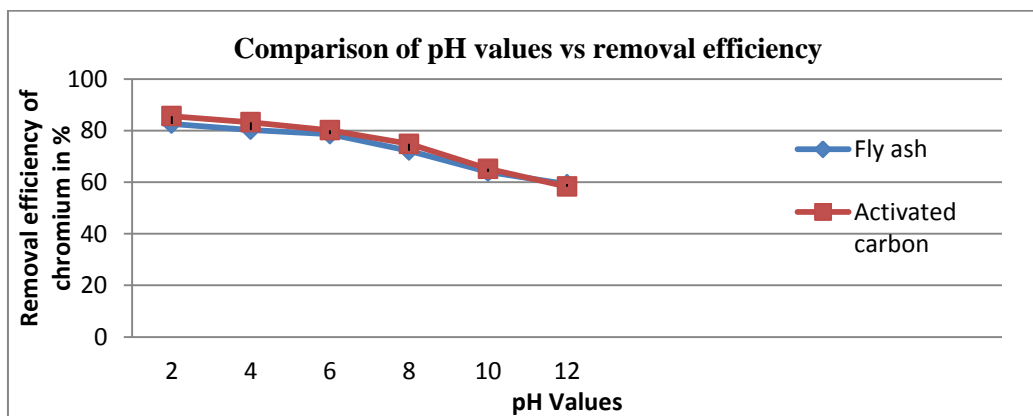


Fig 4: Comparison graph between pH value vs. removal efficiency of chromium

The graph shows that the maximum removal efficiency of chromium 82.63 % was achieved at pH value 2 when fly ash was used as an adsorbent. Similarly, in the case of activated carbon, the maximum removal efficiency of 85.6 % was achieved at pH value 2. It seems that by increasing the pH value up to 10 the removal efficiency was decreased in both cases.

3.5 ADSORPTION ISOTHERM STUDY:

An equilibrium study on adsorption provides information on the capacity of the adsorbent. At a fixed temperature, the ratio of the quantity adsorbed to that remaining in the solution is called adsorption isotherm and it best describes the relationship between the adsorbent and the adsorbate. The Langmuir isotherm Freundlich adsorption isotherm for both fly ash and activated carbon is represented in figure number 5, 6, 7 and 8 respectively. The details values are listed in table 3.1.

3.5.1 Langmuir adsorption isotherm:

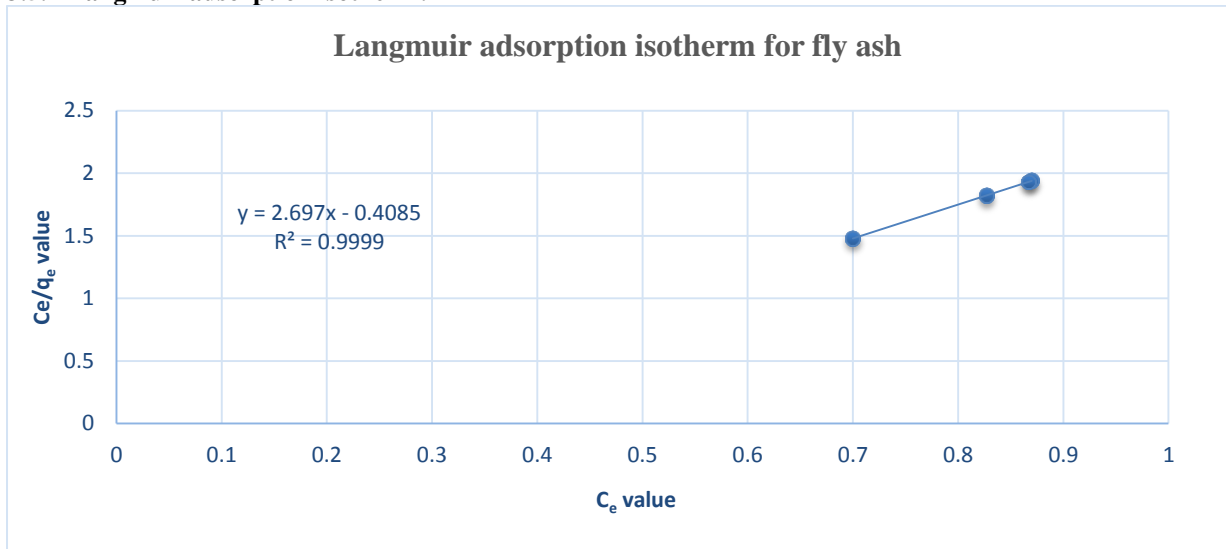


Fig no 5: Graphical representation of Langmuir adsorption isotherm for fly ash

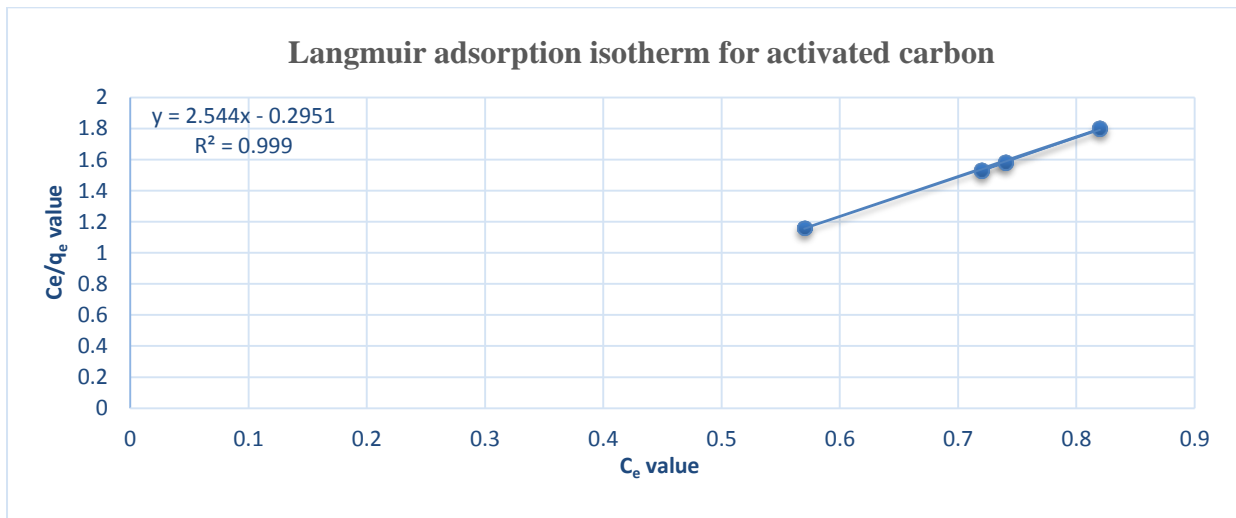


Fig no 6: Graphical representation of Langmuir adsorption isotherm for activated carbon

3.5.2 Freundlich adsorption isotherm:

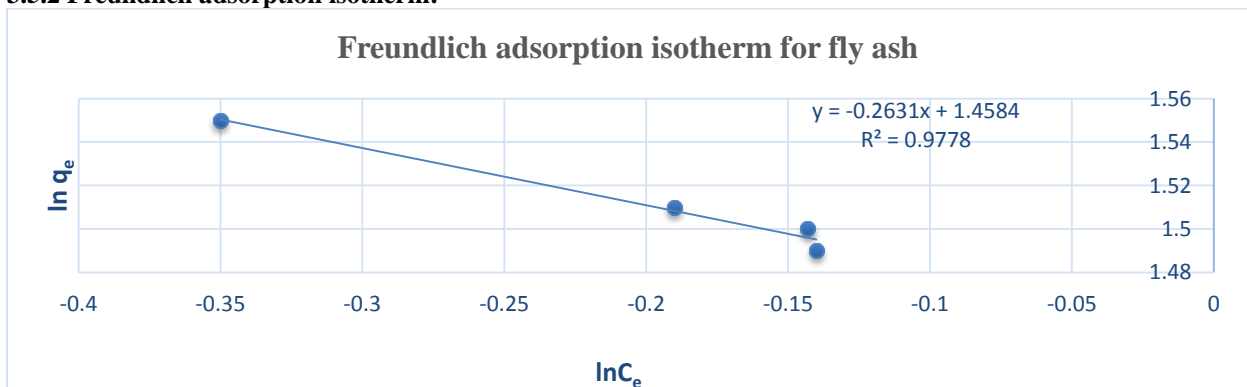


Fig no 7: Graphical representation of Freundlich adsorption isotherm for fly ash

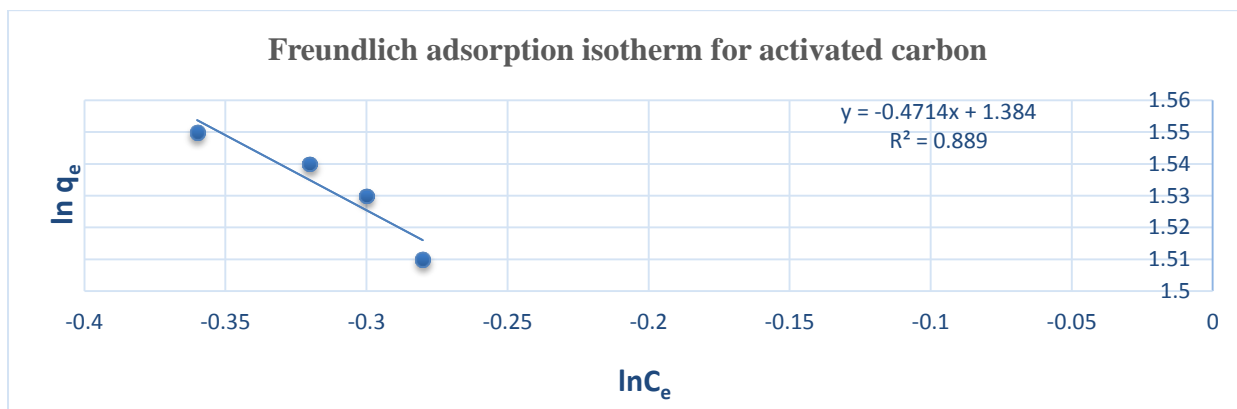


Fig no 8: Graphical representation of Freundlich adsorption isotherm for activated carbon

Table 3.1: Langmuir and Freundlich adsorption isotherm

Type of adsorbent	Langmuir adsorption isotherm				Freundlich adsorption isotherm		
	Q_m	b	R_L	R^2	K_f	n	R^2
Fly ash	0.370	6.66	0.036	0.999	5.15	3.80	0.9778
Activated Carbon	0.393	8.84	0.027	0.999	10.38	2.12	0.889

The separation factor R_L represents the nature of adsorption according to the following characteristics. If $R_L > 1$ then it represents the unfavorable condition of adsorption, if $0 < R_L < 1$ then it represents the favorable condition and if $R_L = 0$ then it represents the irreversible condition for adsorption. In this experiment, R_L was found 0.036 for fly ash and 0.027 for activated carbon. Here in both the case R_L is between 0 to 1, this indicates that fly ash and activated carbon are good adsorbents for chromium ion removal. It also seems that the R_L value of activated carbon is lesser than fly ash, which indicates that activated carbon is a better adsorbent than fly ash. Similarly, in the Freundlich model “n” is the Freundlich exponent that represents the adsorption intensity and k_f (mgg^{-1}) is the Freundlich constant which represents the adsorption capacity. K_f is related to temperature and the physiochemical characteristics of the adsorbent. Here “n” is an indicator of the change of intensity of the adsorption process and also a measure of the deviation from linearity of the adsorption. If the n value is greater than 1, it is indicated that the condition is favorable for adsorption. Similarly, if n value is less than 1, it is indicated that the condition is poor for adsorption. In this experimental investigation, the n value was found 3.80 for fly ash and 2.12 for activated carbon. Here both the value greater than 1, which indicates that the favorable condition of adsorption.

IV. Conclusion

In this experimental study, the effect of different parameters like concentration of adsorbate, contact time, the dose of adsorbent, and pH of the solution have been investigated. This study shows the result of the suitability of adsorbent for the removal of chromium from aqueous solution. It is concluded that waste materials like fly ash and activated carbon can be used as an adsorbent in case of the removal of chromium from wastewater. The optimum removal efficiency was achieved at an initial concentration of 4 mg/L for fly ash and activated carbon. The optimal removal efficiency of 83.12 % and 83.42 % was achieved in the case of fly ash and activated carbon respectively. The equilibrium adsorbent dose was achieved at 35 mg/L for both the cases. The equilibrium time for chromium adsorption is 120 min for both the case of the adsorbent. The maximum percentage of removal is achieved at pH 2 for both fly ash and activated carbon.

In the above four parameters activated carbon always gives the maximum percentage of removal efficiency as compared to the fly ash. The removal efficiency decreases with increasing the concentration of chromium and removal efficiency increases with increasing the dose of adsorbent with constant concentration. The analysis was carried out by using the Langmuir model and Freundlich model. The outcome shows that the R^2 value of the Langmuir model is greater than the Freundlich model, which concludes the predominance of monolayer over inter-molecular interaction amongst the adsorbed chromium in both cases of the adsorbent. These analyses also concluded that both fly ash and activated carbon are suitable for the adsorption of chromium from wastewater.

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