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Use of Maize Husk Fly Ash as an Adsorbent for Removal of Fluoride from Water

Jadhav A S[1], Jadhav M V[2]

[1]M.E Civil (Environmental Engg.), Department of Civil Engineering, SRESCOE Kopargaon (M.S), India,
[2]Prof., Department of Civil Engineering, SRESCOE Kopargaon (M.S), India,

***Abstract*— In the present study batch adsorption process was conducted to evaluate the suitability of Maize husk fly ash (MH fly ash) as an adsorbent for removing fluoride from water. During the experimental work, effects of some of the major parameters of adsorption, viz. Contact time, p^H, Adsorbent dose and Stirring rate on removal efficiency were studied and optimized. The equilibrium was attained in 120 minutes, Maximum removal efficiency was obtained at pH value of 2, optimum adsorbent dose was found to be 2.0 g/50 mL, optimum stirring rate was obtained at 250 rpm. Maximum fluoride removal was observed to be 86% at optimum conditions. Freudlich, Langmuir, Temkin and Redlich-Perterson isotherms were plotted and best suited model was found.**

***Keywords*— Maize husk fly ash, Fluoride removal, Batch study, Adsorption isotherms.**

I. Introduction

Fluorine is the chemical element represented by the symbol F and atomic number 9. At standard pressure and temperature, fluorine is a pale yellow gas composed of diatomic molecules, F_2. It is the lightest element of the halogen column of the periodic table and has a single stable isotope, fluorine-19. Fluorine is rare compared to other light elements. In Earth's crust, fluorine is more common, being the 13th most abundant element. Fluorine, a fairly common element of the earth's crust, is present in the form of fluorides in a number of minerals and in many rocks. Excess fluoride in drinking-water causes harmful effects such as dental fluorosis and skeletal fluorosis. The permissible limit of fluoride level is generally 1 mg/l. The importance of defluoridation studies have increased due to high fluoride levels in drinking-water and its impact on human health in many parts of India.

One of the efficient method for the removal of fluoride from water is Adsorption. Up till now many adsorbent materials such as activated carbon, egg shell, bone-char, Tamarind seed, rice husk, limestone etc. have been used for removal of fluoride.

In this paper, for the removal of fluoride from water, a new low-cost adsorbent, namely, Maize husk fly ash has been used.

II. Methodology

A. Material collection and sampling

The maize husks were collected and air dried. The maize husk fly ash was then produced by simple open burns, which typically occur for a temperature range between 500ºc to 600ºc [nair et al, 2006]. The ash obtained was passed through 60 µm sieve to produce the final adsorbent.

Table 1 shows the chemical constituents of MH fly ash.

TABLE I
CHEMICAL CHARACTERISTICS OF MAIZE HUSK FLYASH

Constituents	Maize Husk Flyash
	Amount (%)
SiO_2	40.16
k_2O	25.52
CaO	6.90
MgO	5.91
P_2O_5	4.01
Cl	3.37
SO_3	1.45
LOI	12.68

Synthetic fluoride solutions of known concentrations were prepared to carry out the experimental work. The stock solution of 100 mg/l fluoride was prepared by dissolving 221 mg of anhydrous NaF in 1 L of distilled water. A calibration curve was prepared using standard solutions. Zirconyl acid reagent, Spands solution, NaF, reference solution etc., were among the chemicals used during the appropriate experimental procedure.

B. Methodology

The adsorbent used for the study was maize husk fly ash. All the experiments were carried out in 100 ml glass jar with 50 ml test solution at room temperature (29±2^{o}C). The jar, along with known volume of test solution of fixed concentration and 1.0 g of the adsorbent at neutral p^H, was shaken in mechanical stirrer at 400 rpm to study the equilibration time for maximum adsorption of fluoride. Synthetic fluoride solution of 7mg/l initial F^- concentration was used.

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The experimental work was carried for finding out the effect four varying parameters on fluoride removal efficiency, given as below:

1. Effect of contact time
2. Effect of p^H
3. Effect of adsorbent dose
4. Effect of stirring rate

III. Results and Discussion

Experimental work of this study has been divided into following phases. They are

• Adsorption procedure by Batch study.

1. Effect of contact time on fluoride removal.
2. Effect of p^H on fluoride removal.
3. Effect of Adsorbent dose on fluoride removal.
4. Effect of stirring rate on fluoride removal.
5. Effect of initial fluoride concentration (IFC) on fluoride removal.

• Validation of results through Modeling (Langmuir, Freundlich, Temkin, Redlich-perterson Models) for fluoride removal.

A. Effect of contact time on percentage fluoride removal

The effect of contact time on percentage fluoride removal was studied/ determined by taking 1g of adsorbent dose, stirring rate of 300 rpm at neutral p^H as shown in Figure 1.

The percentage removal efficiency of fluoride was found to be function of contact time. With the increase in contact time, there was increase in the removal efficiency. An equilibrium time of 120 min was found and used for further studies. Though the efficiency obtained was less, it may be due to the low adsorbent dosage used.

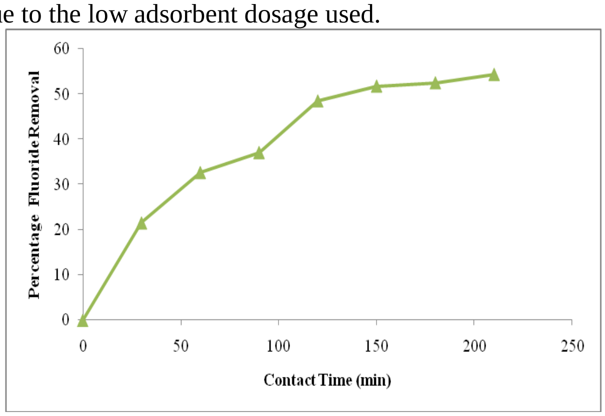

Figure 1: Graph for effect of contact time on fluoride removal

B. Effect of p^H on percentage fluoride removal.

The effect of p^H on percentage fluoride removal was studied by varying the p^H from 2 to 12 and by taking 1g of adsorbent dose, stirring rate of 300 rpm at an equilibrium contact time of 120 minutes as shown in Figure 2.

Removal efficiency was found to be decreasing with an increase in p^H values. Maximum removal efficiency was obtained at a p^H value of 2 which was around 71.71%.

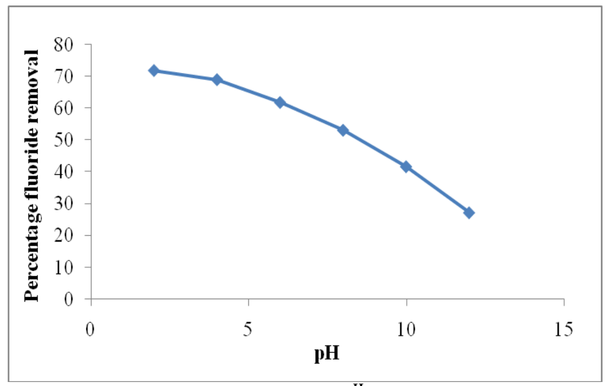

Figure 2: Graph for effect of p^H on fluoride removal

C. Effect of Adsorbent dose on percentage fluoride removal.

The effect of adsorbent dose on percentage fluoride removal was studied by varying the adsorbent dose from 0.25g to 2.5g with a stirring rate of 300 rpm at neutral p^H and an equilibrium contact time of 120 minutes as shown in Figure 3.

With the increase in adsorbent dose, the fluoride removal efficiency also increased. Also after a adsorbent dose of 2g used, there was no marginal increase in the efficiency. Hence a optimal dosage of 2g was taken for further studies.Maximum removal efficiency was found to be around 86%.

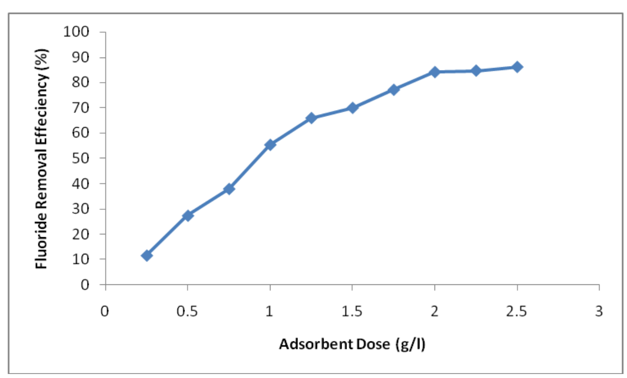

Figure 3: Graph for effect of adsorbent dose on fluoride removal

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D. Effect of Stirring rate on percentage fluoride removal.

The effect of Stirring rate on percentage fluoride removal was studied by varying the stirring rate from 50 rpm to 400 rpm with a adsorbent dose of 2g at neutral p^H and an equilibrium contact time of 120 minutes as shown in Figure 4.With the increase in stirring rate, the fluoride removal efficiency also increased. Also after a stirring rate of 250 rpm, there was no marginal increase in the efficiency. Hence an optimal stirring rate of 250 rpm was considered for further studies. Maximum removal efficiency was found to be around 84%.

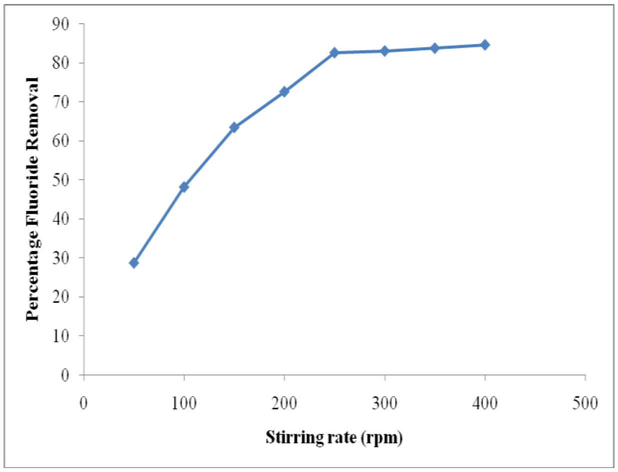

Figure 4: Graph of effect of stirring rate on fluoride removal

E. Validation of results using adsorption isotherms.

The results obtained were validated by using adsorption isotherms like Langmuir model, Freundlich model, Temkin model and Redlich - Perterson model

1) Langmuir Model

The linear form of the Langmuir isotherm is expressed as follows.

$$1/q_e = 1/a + 1/ab\ c_e$$

Where q_e (mg/g) is the ratio of fluoride adsorbed to the dosage of the adsorbent, Ce (mg/l) is the equilibrium concentration of adsorbate, a is the adsorption capacity of adsorbent and b is a constant related to the energy adsorption.

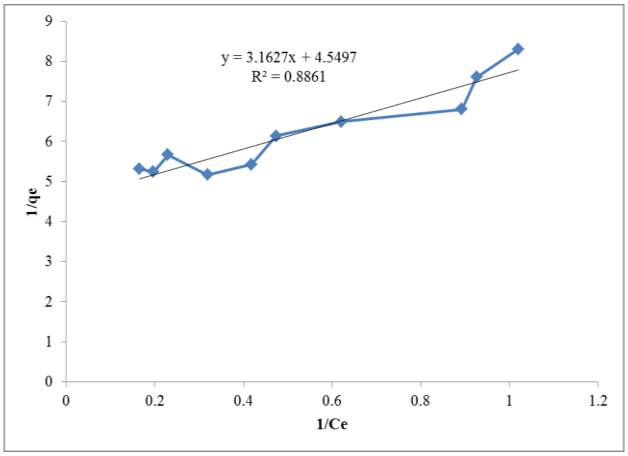

Figure 5: Langmuir model for removal of fluoride using MH fly ash.

Figure 5 shows the graph for Langmuir model for which the R^2 value is equal to 0.8861.

2) Freundlich model

The linear form of Freundlich model is represented by the following equation.

$$\ln q_e = \ln K_f + 1/n \ln c_e$$

Where, K_f is the adsorption capacity of adsorbent and n is a constant related to the intensity of adsorption.

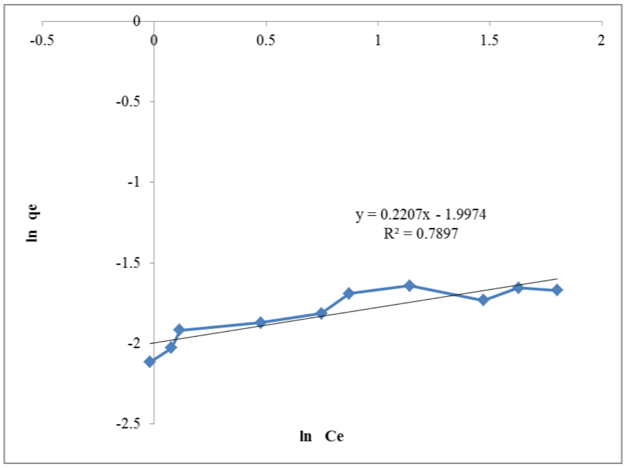

Figure 6: Freundlich model for removal of fluoride using MH fly ash.

Figure 6 shows the graph for Freundlich isotherm model for which the R^2 value is equal to 0.7897.

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3) Temkin Model

The Temkin isotherm, the simple form of an adsorption isotherm model, has been developed considering the chemisorption of an adsorbate onto the adsorbent, is represented as:

$$q_e = a + b \log c_e$$

where q_e and c_e have the same meaning as noted previously and the other parameters are called the Temkin constants. The plot of qe versus log ce will generate a straight line. The Temkin constants a and b can be calculated from the slope and intercept of the linear plot.

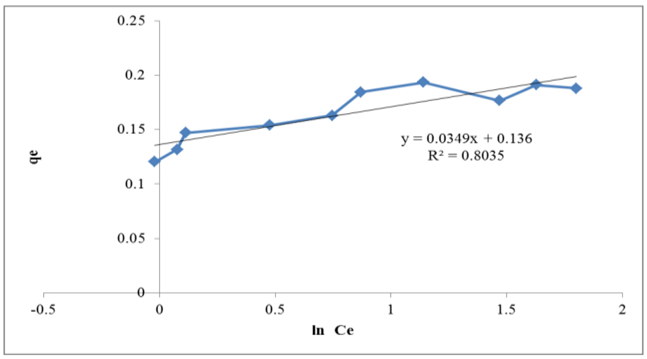

Figure 7: Temkin model for removal of fluoride using MH fly ash.

Figure 7 shows the graph for Temkin isotherm model for which the R^2 value is equal to 0.8035.

4) Redlich-Perterson model

The Redlich–Perterson isotherm contains three parameters and incorporates the features of the Langmuir and Freundlich isotherms.

The linear form is represented by the following equation.

$$Ln((A\ C_e/q_e)-1) = g \ln C_e + \ln B$$

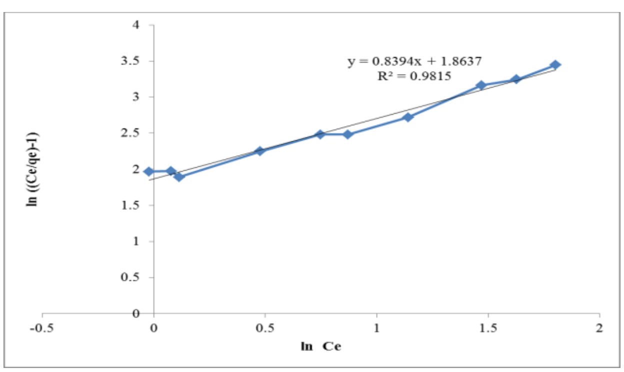

Figure 8: Redlich - perterson model for removal of fluoride using MH fly ash

Figure 8 shows the graph for Redlich - Perterson isotherm model for which the R^2 value is equal to 0.9815.

IV. CONCLUSIONS

- The efficiency of maize husk fly ash as an adsorbent for removal of fluoride from water using batch process has been recognized fruitfully.
- In the first and most important phase, batch study was carried out. In this phase, the removal efficiency of adsorbent used was checked for four varying parameters, viz. Contact time, p^H, adsorbent dose and stirring rate, to find the optimum conditions or equilibrium data.
- For the effect of contact time, equilibrium was attained for 120 minutes.
- For the effect of p^H, equilibrium was attained for p^H value 2.
- The percentage of fluoride removal was found to be a function of adsorbent dose and contact time at a given initial fluoride concentration. In case of effect of adsorbent dose, equilibrium dosage of 2g was determined. While the maximum efficiency was found to be 86%.
- For Stirring rate variation, the equilibrium stirring rate of 250 rpm was found. The maximum efficiency obtained was found to be 84.71% for MH fly ash.
- As for MH fly ash, the removal increased with time, adsorbent dose and stirring rate, but for varying p^H and the removal efficiency decreased with increased p^H value.
- The second and the last phase of the study validated the results obtained from batch study by using adsorption isotherm modelling. The present study on defluoridation using MH fly ash as an adsorbent reveals that the equilibrium data fits better to Redlich-Perterson model (R2 > 0.981) than Langmuir, Freundlich and Temkin model.

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