International Journal of Research in Engineering and Science (IJRES)
ISSN (Online): 2320-9364, ISSN (Print): 2320-9356
www.ijres.org Volume 9 Issue 7 ǁ 2021 ǁ PP. 60-67

Removal Of Iron From Groundwater Using Ferric Hydroxide [$Fe(OH)_3$] Flocs, Rice Husk And Coconut Shell Charcoal: A Review

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Abstract

One of the problems related to groundwater is the reddish colour caused by the presence of ferrous and manganese. Initially, this colour cannot be seen but after it has been exposed to the air, the oxidation of groundwater will promote the precipitation of ferrous and manganese. Eventually, the groundwater turns into reddish in colour. The present study is focused on critical review of previous and current available information on potential of ferric hydroxides flocs, treated rice husk and coconut shell charcoal for the removal of iron(II) from groundwater. Various studies on adsorption efficiency of rice husk, coconut shell charcoal considering the parameters contact time, initial concentration of iron, pH, and contact time have been evaluated by many researchers. The present study analyzed those studies and compiled the adsorption efficiency of ferric hydroxide flocs, coconut shell charcoal and rice husk and concluded that coconut shell charcoal shows higher adsorption efficiency compared to other adsorbents.

Keywords: *Adsorption efficiency, Adsorption isotherm, ferric hydroxide flocs, rice husk, Coconut shell charcoal*

Date of Submission: 25-06-2021 Date of acceptance: 08-07-2021

I. INTRODUCTION

Groundwater is the major source of drinking water in most parts of the world. In India, 50% of urban water requirement and 85% of rural domestic water requirement are met by ground water use (World Bank, 2010). Groundwater is considered as the primary source of drinking water because of its convenient availability, naturally good quality and its relatively low capital cost. However, it is also vulnerable to all sorts of contaminations due to natural causes or various anthropogenic activities such as agricultural, domestic and industrial .One of the major concerns regarding the contamination of groundwater is the precipitation and accumulation of heavy metals. Natural sources of heavy metals may include weathering and erosion of bed rocks and ore deposits. Due to the rich availability of laterite soil, iron content in Indian states such as Kerala is high (DMG, 2016). When leaching occurs, the iron enters the aquifer affecting the water quality of the aquifer. As per the Central Groundwater Board status, about 13 districts of Kerala have iron content in the aquifers at a concentration exceeding the permissible limit of 1.0 mg/l (CPCB, 2007a, b). .The presence of iron results in a reddish colour and undesirable. It usually exists in two oxidation states comprising ferrous ion (soluble divalent form: Fe (II) or Fe(OH)+) and oxidized form (trivalent ion: Fe (III) or Fe(OH)3). A dose of 1500 mg/l iron can damage blood tissues in children while among adults, it can cause digestive disorders, skin diseases and dental problems. As per the drinking water standards in India, desirable limit of iron is 0.3 mg/l. High levels of iron make large volumes of water unavailable for drinking, and water scarcity ensues. So iron should be removed from ground water. Researchers have attempted to develop advanced technologies for the treatment of water, more effective and economic than the prevailing ones. Fe(II) present in groundwater are difficult to remove. So ferric hydroxide flocs can be used for the efficient adsorptive removal of Fe(II) from ground water. Agricultural residues such rice husk, coconut shell charcoal have a high potential to function as adsorbents so researchers are using these for adsorptive removal of iron from ground water. Considering initial concentration of adsorbate, pH and contact time, adsorption efficiency of rice husk has been reviewed in this study.

II. THEORETICAL CONSIDERATIONS

2.1. Physicochemical characteristics of Ferric Hydroxide [Fe $(OH)_3$]

Nitha Abraham, Jenny James, 2019 reported that in ground water, iron exists mainly as Fe(II) due to lack of oxygen (Redman et al., 2002; Buschmann et al., 2006; Palmer et al., 2006; Benner and Fendorf, 2010). The water solubility of Fe(II) make its removal from groundwater a key concern for most water supply companies who use groundwater as their source. During the process, Ferric Hydroxide is formed on the surface of the media by hydration and the FeO is responsible for adsorption. Direct adsorption of Fe(II) from the groundwater onto the ferric hydroxide precipitate, its conversion to Fe(III), and its subsequent hydrolysis to ferric hydroxide, forming a new surface for further adsorption. The adsorption mechanism proposed in this study involved direct adsorption of Fe(II) from the groundwater onto the ferric hydroxide precipitate, its conversion to Fe(III), and its subsequent hydrolysis to ferric hydroxide, forming a new surface for further adsorption. Thus, there was no need to frequently replace or maintain the media and no backwashing process was required. This underscores the cost effectiveness of the propose method.

2.2. Physicochemical characteristics of Coconut Shell Charcoal

Liyanage, C.D. and Pieris .M (2015) reported that the chemical composition of coconut shell consists of lignin (29.35%), cellulose (24.20%), hemicellulose & others (38.56%), moisture (7.7-3%) and ash (0.68%).The shell carbon content increases up to 76.32 % as the shell was pyrolized into charcoal. The carbon structure of coconut shell charcoal has high porosity and large surface area and this properties are important in adsorption process.The iron removal results from the adsorption of oxygen followed by the oxidation of Fe2+, catalyzed by the adsorbent carbon surface. The Fe3+ precipitates on the solid, forming a hydrated iron oxide coated carbon that is also able to absorb iron. The present modified coconut shell charcoal has been found to be very effective and remove iron to below 0.3 ppm in water without increasing the pH above the acceptable limit. The rate of adsorption is closely related with the accessibility of the porous structure and, again, the molecular dimensions of the adsorbate. Moreover, the medium from which adsorption occurs (gas or liquid) is predominant. Generally, during adsorption from the gas phase in filters, the rate of diffusion is not limiting. This means that in these cases the maximum adsorption capacity is predominant, although for solvent recovery purposes the rate of desorption from the carbon is sometimes considered to be a bottleneck. For liquid applications however, the accessibility of the porous systems is of predominant importance, especially for large molecules that exhibit slow diffusion transport in narrow pores. This urges the need for a short distance between the wider pores that facilitate transport and more narrow pores in which effective adsorption takes place. In practice, this may be achieved by establishing a well-developed mesopore structure via the pore drilling mechanism. However this type of pore formation excludes large mesopore surface areas. This means that optimum carbons for this type of application are always a compromise between high adsorption capacities and good accessibility.

2.3. Physicochemical Characteristics of Rice Husk

Ye et al, (2012) reported physical properties of rice husk such as surface area of 438.05m2/g, bulk density of 0.3086 g/cm^3 and porosity of 0.38 by fraction. The chemical composition of rice husk by component analysis has been reported as 34.4% cellulose by weight, 29.3% hemicellulose by weight, 19.2% lignin by weight, 17.1% ash by weight, elemental analysis shows 49.3 % oxygen by weight , 44.6 % carbon by weight , and 5.6 % hydrogen by weight and by proximate analysis 59.5% volatiles by weight, 17.1 % ash by weight (composition of mineral ash SiO, K_2O,MgO,Fe_2O,Al_20_3,CaO)and 7.9 % moisture by weight was found (Williams and Nugranad, 2000). As rice husk is insoluble in water, having good chemical stability, structural strength due to high silica content so researchers are using it for treating heavy metal such as iron from ground water. . The adsorption of Fe(II) onto rice husk sample follows pseudo second order kinetics. The pseudo second order confirmed the chemisorption, involving ion exchange and valence forces through sharing or exchange of electrons between adsorbent and adsorbate.The surface of RH is porous, and the surface honeycomb holes can reach micron scale, about 10 microns. The internal structure possesses a number of irregular pieces of layered structure and reticulates. There is irregular holes distribution within the RH; this may be a result of the combined action of rice husk residue. The porous structure of RH has a relatively large specific surface area, and this morphological property is conducive to the uptake of metal ions.

2.4. Adsorption isotherm

In adsorption modelling, the distribution of solute between the liquid and the solid phase is often described in terms of isotherms. The presentation of the amount of solute adsorbed per unit of adsorbent as a function of the equilibrium concentration of solute in bulk solution, at a constant temperature is called adsorption isotherm. The Langmuir and Freundlich model are the two commonly used adsorption isotherms in aqueous system. The Langmuir model is valid for single layer adsorption. It is based on the assumption that maximum adsorption corresponds to a saturated monolayer of solute on the adsorbent surface, the adsorption energy is same at all sites and there is no transmigration of adsorbate in the plane of the surface.

The Freundlich isotherm is the most widely used mathematical description of adsorption in aqueous systems. It is based on the assumption of heterogeneous adsorption surface comprising of different classes of adsorption sites and energies. The Freundlich equation is an empirical expression but it can often as a means of data description. Dzombak and Morel (1996) reported that cation sorption on to hydrous oxides can usually be approximated by the Freundlich isotherm. Freundlich isotherm is mathematically represented as

(1)

Where q_e is the amount of solute adsorbed per unit weight or surface area of the adsorbent (g/g or g/m^2), C_e equilibrium concentration of solute (g/m^3), K_f and n are isotherm constants. On linearization the equation takes the form

(2)

Isotherm constant K_f is the measure of adsorption capacity and constant n is the measure of adsorption intensity. For fixed values of C_e and n, the higher value of K_f, higher is the adsorption capacity. For fixed value of K_f and C_e, the lower the value of n, the stronger is the adsorption bond. As n becomes very small, the capacity tend to be independent of C_e, and the isotherm plot approaches the horizontal. If the value of n is large, the adsorption bond is weak, and the value of q_e changes markedly with small changes in C_e.

2.5. Adsorption capacity

To estimate the adsorption capacity in terms of removal efficiency, which means the amount of iron uptake. Equations shown below are used to calculate amount of iron uptake (adsorption capacity) and uptake % or adsorption efficiency (Thanakrit Neamhom,2019)

= (3)

Uptake % or Adsorption Efficiency =×100 (4)

Where the amount of iron uptake, V is the volume of solution, W is the weight of adsorbent in Kg, and C_i and C_f are the initial and final iron concentration in g/L.

III. MATERIAL AND METHODS

3.1. Materials used

Ferric hydroxide [Fe(OH)$_3$] flocs, coconut shell charcoal, rice husk are used as adsorbents. Chemicals such as 2 M potassium hydroxide (KOH) solution, 0.5 M HCL and 0.5M sodium hydroxide(NaOH) solution were used for treating coconut shell charcoal and rice husk. Ferric chloride was used for the preparation of Ferric hydroxide flocs.

3.2. Preparation of adsorbents

3.2.1. Ferric Hydroxide: 15 g of $FeCl_3$ was dissolved in 1 L of distilled water to get $FeCl_3$ solution, into which 20 ml of ammonia solution was added to initiate the precipitation of Fe $(OH)_3$. The adsorbent was filtered using Whatman Grade 42 Filter Paper after washing it thoroughly with distilled water such that its pH was maintained around 7(Nitha Abraham, Jenny James,2019).

3.2.2. Coconut shell charcoal: Coconut shells which were collected from local markets were crushed and sieved to 20 to 30 mesh and dried at 400 ^{0}C for 1 h in a vacuum. After that, dried CS was mixed with a 2 M potassium hydroxide (KOH) solution (1:40 w/v) overnight. Finally, CS was dried at 105 ^{o}C for 1 h and sieved to 60 mesh (Thanakrit Neamhom, 2019).

3.2.3. Rice Husk: Collection of rice husk may be done from local rice mill and may be prepared as an adsorbent by sieving through different sieve sizes to get the various particle size of rice husk. Cleaned rice husk was soaked in 0.5 M HCl solution (1:20 w/v) at room temperature for 4 h in permanent agitation on a 200 rpm shaker. To increase the adsorption capacity, RH was washed and soaked in a 0.5 M sodium hydroxide (NaOH) solution (1:20 w/v) for 4 h at the same above mentioned conditions. The pH value was adjusted using HCl acid solution. Purified RH was repeatedly washed with distilled water and dried at 40 ^{o}C for 48 h. After dry with high temperature, they were ground and sieved using a 60-mesh screen. (Thanakrit Neamhom,2019).

Table 1. Parameters considered and adsorption isotherm of adsorbents

Adsorbents	Parameters considered	Adsorption isotherm	References
Ferric hydroxide flocs	pH, contact time	_	Nitha Abraham, Jency James ,Tuhin Banerji, and Ratish Menon,2019
Coconut shell charcoal	pH, contact time	Freundlich isotherm,	Ahmad bin Jusoha, W.H. Chenga, W.M. Lowa, Ali Nora'ainia, and M.J. Megat Mohd Noor,2005
Rice husk	pH, contact time	Freundlich isotherm	Sunita Shrestha, Anita Kumari Dhami and Armila Rajbhandari,2021

IV. RESULTS & DISCUSSION

4.1. Effect of pH

4.1.1. Ferric hydroxide [Fe $(OH)_3$] Flocs: N. Abraham et al. (2019) reported that as the pH increased from 5 to 7, the Fe(II) removal efficiency by the adsorption process also increased to double the initial value. The variation of Fe(II) removal efficiency with pH by the adsorption process in graph (figure 1). At pH 7, 86% of iron removal efficiency was obtained, whereas further increase in pH initiated the oxidation of Fe (II) to Fe(III) and subsequent formation of ferric hydroxide. At acidic pH, Fe (III) reduces to Fe (II) there by reducing the removal efficiency. Also, under alkaline pH conditions, Fe (II) conversion to ferric hydroxide is anticipated immediately after its preparation. At higher pH, the precipitate settles down even before it could pass through the ferric hydroxide flocs contained in the adsorption unit. Hence, the precipitation was found to work effectively within the narrow pH range of 6.5 and 7.

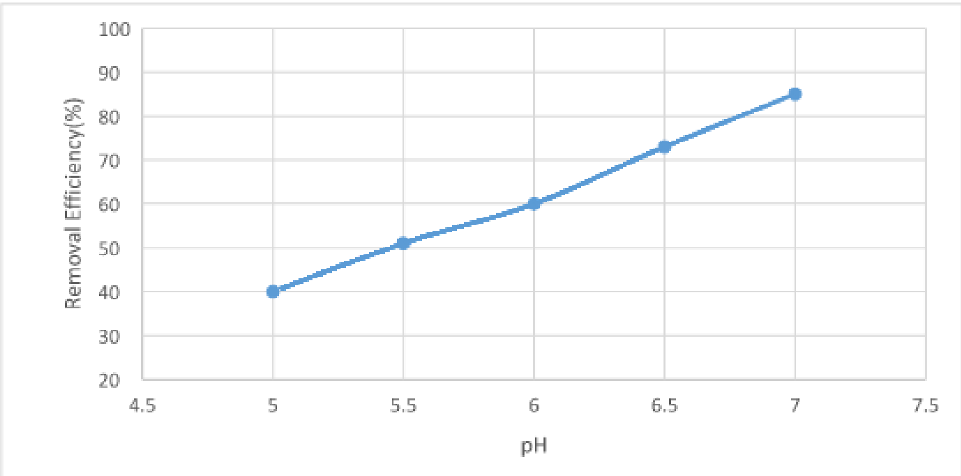

Figure 1.Graph showing variation of Fe(II) removal efficiency with pH

4.1.2. Coconut shell charcoal: Ketsela G, et al. reported maximum removal efficiency at optimum pH for Fe (II) is about 90.08% (shown in Figure 2) because at pH values below 4.0 the surface of the adsorbent is surrounded by more H^+ ions which decrease metal ion interaction with binding sites of coconut shell charcoal by greater repulsion force or the electrostatic force of repulsion between adsorbent and adsorbate is constant, therefore, the removal efficiency is low. At pH above 6.0, the competing effect of H^+ ion is decreased or OH^- ion increased and adsorption sites could be available, there is a opportunity of adsorbate precipitation on the surface of the adsorbents by nucleation.

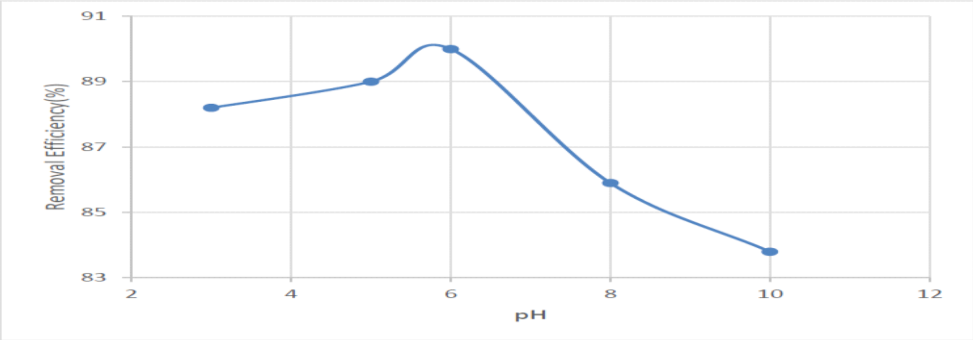

Figure 2. Graph showing Effect of pH on iron adsorption on charcoal

4.1.3. Rice Husk: Monic et al. reported the pH parameter has been identified as one of the most important parameter that is effective on metal adsorption. It is directly related with competition ability of hydrogen ions with iron ions to active sites on adsorbent surface. The effect of pH on the adsorption of iron onto Rice Husk showed in the graph (Figure 3) which notices that the removal of iron on RH became higher as the pH value increased. The removal became constant at pH value between 7 and 9 or in neutral or alkali solution.

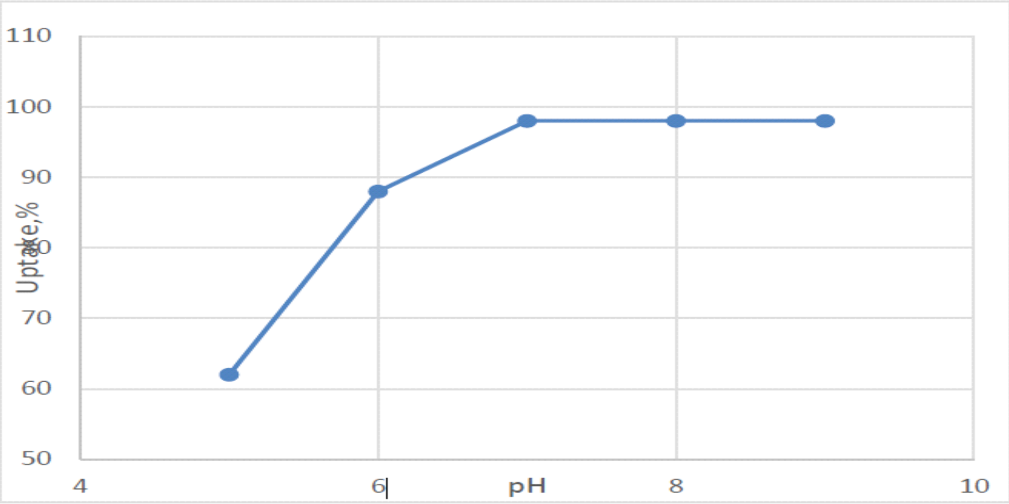

Figure 3. Graph showing effect of pH on iron adsorption on Rice Husk

4.2. Effect of contact time

4.2.1. Ferric hydroxide: N. Abraham et al. (2019) reported that as the contact time increases, the removal efficiency also increases. At 15 min contact time, Fe(II) removal efficiency obtained for 20 mg/l initial concentration was 100% and the variation in contact time, from 1 min to 30 min, resulted in the increase of Fe(II) removal efficiency by 5% is shown in Figure 4.Thus, contact time emerged as a dominating factor in the adsorption process and variation in contact time positively influenced the performance of the process.

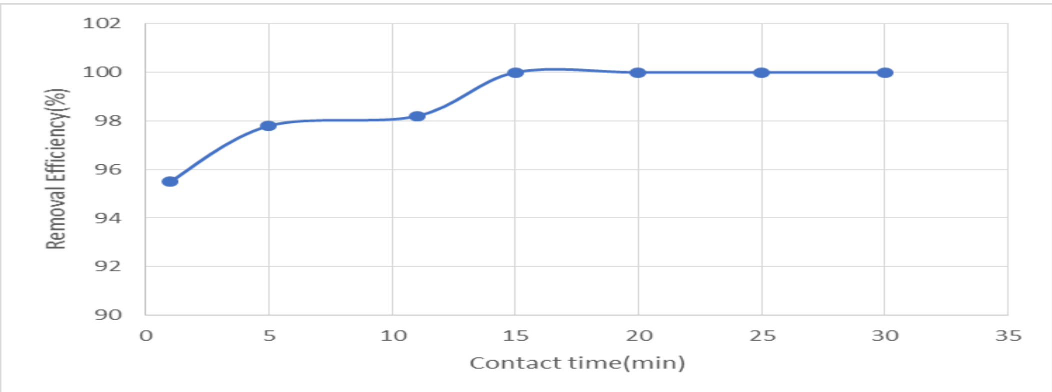

Figure 4. Graph showing Effect of contact time on iron adsorption on Ferric Hydroxide

4.2.2. Coconut shell charcoal: Figure 5 reveals that the rate of percent Fe^{3+} removal is higher at a contact time of 40 min. This may be due to larger surface area of the activated Carbon powder being available at the beginning for the adsorption of Fe^{3+}. As the surface adsorption sites become exhausted, the uptake is controlled by the rate at which the adsorbate is transported from the exterior to the interior sites of the adsorbent particles. (Jetafew Ketsela et al,2020).

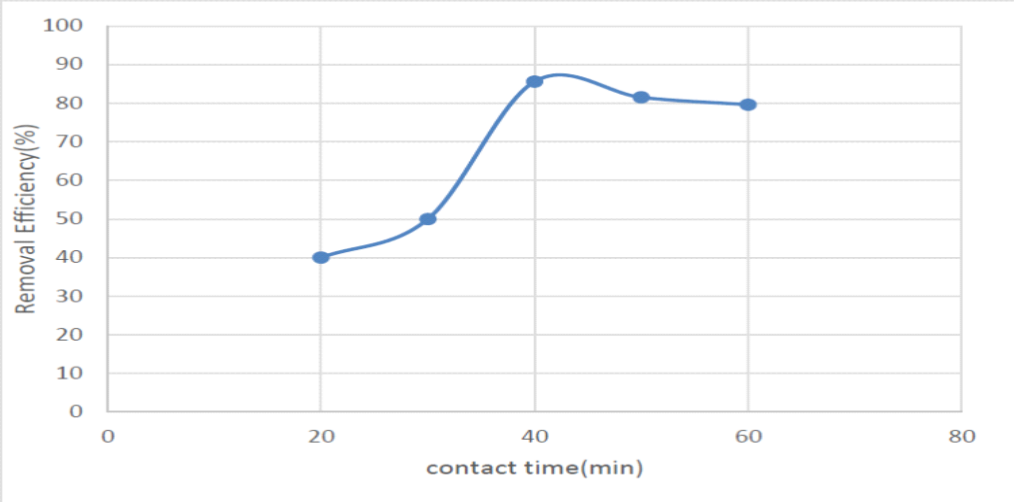

Figure 5. Graph showing effect of contact time on iron adsorption on coconut shell charcoal

4.2.3. Rice Husk: Monic et al reported rate of adsorption is important to design batch experiment. Thus, the effect of contact time on iron adsorption was investigated. The experiment was done for 240 minutes (4 hours) and sample was collected after 60 minutes, 120 minutes, 150 minutes, 180 minutes and 240 minutes processes is shown in the Fig 6. The removal of iron was decreased the contact time increased but become constant after 180 minutes. For further experiment, 60 minutes is considerably suitable for iron adsorption process as reported previous report for similar result in reducing heavy metal using different kind of adsorbents.

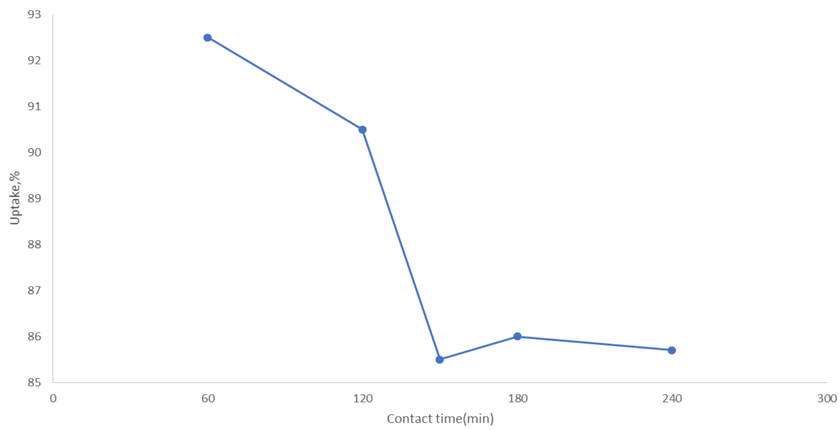

Figure 6: Effect of contact time on iron adsorption on Rice Husk

4.3. Adsorption isotherm

Freundlich isotherm for adsorption of iron onto coconut shell charcoal was plotted (Figure 7). K_f indicates adsorption capacity which was found to be 3.162mg/g for charcoal. The slope 1/n ranging between 0 and 1, is favorable adsorption condition and was found to be 0.1122 for charcoal which suggests that the adsorption is linear and uniform throughout the adsorbent surface, however the correlation behavior of Fe(II) from aqueous solution by charcoal coefficient value obtained from Freundlich isotherm for charcoal was found to be 0.9925.

Freundlich isotherm for adsorption of iron onto rice husk was plotted (Figure 8). K_f indicates adsorption capacity which was found to be 2.907 mg/g for rice husk. The slope 1/n ranging between 0 and 1, is favourable adsorption condition and was found to be 0.2574 for rice husk respectively which indicates the favourable adsorption behaviour of Fe(II) from aqueous solution by rice husk sample however the correlation coefficient value obtained from Freundlich isotherm for rice husk was found to be 0.8797.

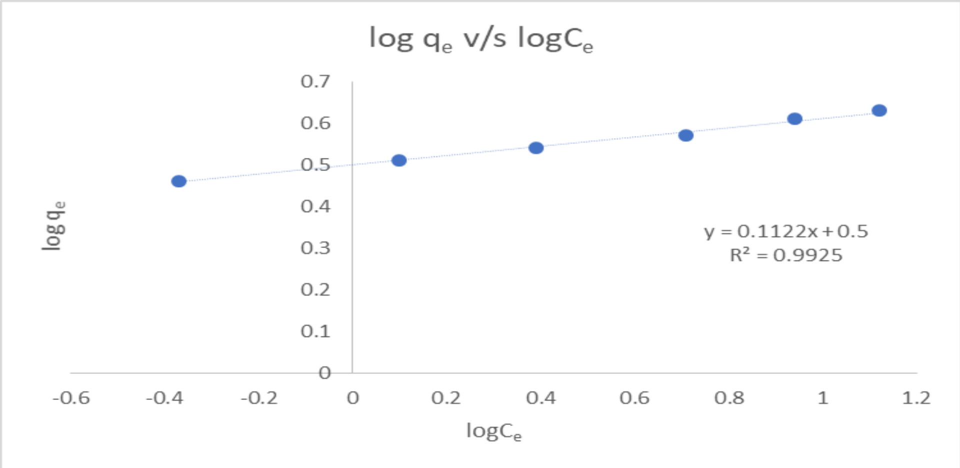

Figure 7: Freundlich isotherm plot for the adsorption of iron onto charcoal

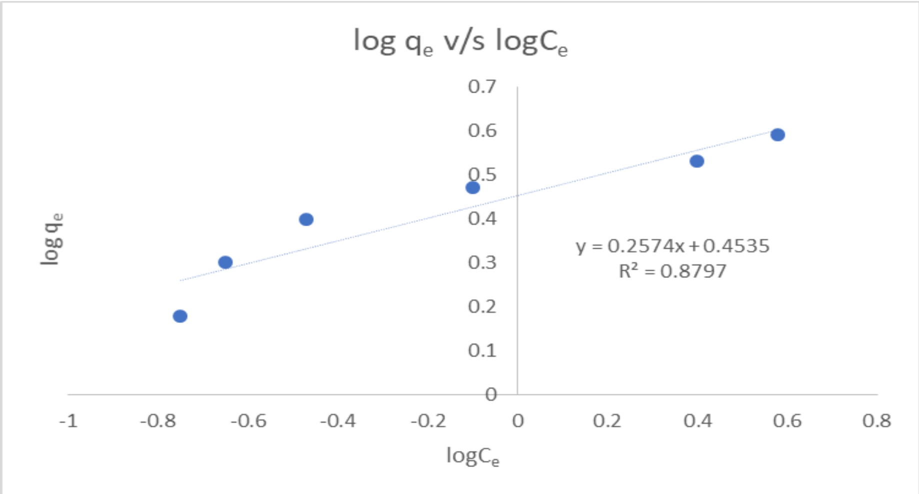

Figure 8: Freundlich isotherm plot for the adsorption of iron onto rice husk

The adsorption data was tested through Freundlich Adsorption Isotherm model. Table 2 shows the data obtained from adsorption isotherm of charcoal and rice husk.

Table 2. Data obtained from adsorption isotherm of charcoal and rice husk

Adsorbent	1/n	K_F	R^2
Rice husk	0.2574	2.907	0.8797
Activated carbon	0.1122	3.162	0.9925

From Freundlich adsorption isotherm, efficiency was calculated and is shown in the table 3. From the table it is clear that coconut shell charcoal shows higher adsorption efficiency compared to rice husk and the efficiency was decreasing with increase in iron concentration.

Table 3. Efficiency Calculation

	CHARCOAL						RICE HUSK					
INITIAL CONCENTRATIO N	0.2	0.4	0.6	0.8	1	1.2	0.2	0.4	0.6	0.8	1	1.2
EFFICIENCY	86.5	48.5	34.67	28.75	24.5	21.33	38	25	21	18.75	17.3	16.25

V. CONCLUSION

This review summarizes the various studies on removal of iron from water using ferric hydroxide flocs, coconut shell charcoal, and rice husk. The use of rice husk and coconut shell charcoal as potential bio-adsorbents for metal remediation and environment management technologies has increased in recent past due to their easy availability, low cost, easy processing, application and recovery without any adverse impact on the environment. Ferric hydroxide flocs are efficient for adsorptive removal of dissolved iron in groundwater. Based on the study conducted adsorption efficiency of various adsorbents obtained. The proposed method of adsorption mechanism showed remarkable efficiency in the removal of Fe (II) from water. In the narrow pH range between 6.5 and 7, the adsorbent worked effectively during the adsorption process. Coconut shell charcoal shows higher adsorption capacity when compared to other adsorbents.

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