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Removal of Lead (II) Ions From Aqueous Solutions Using a Low Cost Adsorbent Obtained from *Centella Asiatica* Leaves

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Abstract: This work deals with the study of the removal of lead from aqueous solution using low cost adsorbent obtained from the leaves of centella asiatica . Effect of pH, adsorbent dose, dye concentration, contact time and temperature on the removal of the metal ions was investigated. Equilibrium data were analyzed using Langmuir and Frendlich isotherm models. The maximum removal was obtained for the adsorbent dose of 100mg at 10mg/l metal concentration at pH 5 and agitation speed of 120rpm. The result shows that Centella asiatica leaf could be employed as an effective low cost adsorbent for the removal of Lead.

Index Terms- Lead(II) ions, low cost adsorbent, *Centella asiatica* leaf, Isotherm models, Thermodynamic parameters.

I.INTRODUCTION

Heavy metals are one of the most predominant hazardous pollutants in the environment. Heavy metal ions are often found in the environment as a result of their wide industrial applications. They are the common contaminants of wastewater and most of them are known to be toxic or carcinogenic. In addition, they are not biodegradable and tend to accumulate in living organisms, causing various diseases and disorders[1]. Pollutant toxicity has forced industries and municipal authorities to treat wastewater before discharging to the natural water bodies[2]. In the human body very small amount of Pb(II) over a long period of time can lead to malfunctioning of certain organs and chronic toxicity[3]. It can damage nearly all tissues, particularly the kidneys as well as the immune and nervous system. The maximum permissible limit of lead in drinking water is upto 0.01mg/L.

Heavy metals refers to any element with the atomic weights between 63.5 and 200.6 and specific gravity greater than 5.0[4]. Cadmium, zinc, copper, nickel, lead, mercury and chromium are some examples of heavy metals which originates from activities of metal plating, mining, battery manufacture, petroleum refining smelting, tanneries, pesticides, pigment manufacture, printing, photographic industries and paint manufacuturing[5,6.]

There are various methods in practice for the removal of heavy metals such as chemical precipitation[7], ion exchange[8,9,10], co-precipitation[11], coagulation, reverse osmosis[12,13], adsorption[14,15] and filtration[16]. In recent years adsorption techniques for wastewater treatment have become more popular and efficient due to simplicity and low cost. Previous research shows that there is growing interest of searching for a variety of materials as low cost adsorbents including sawdust[17], cocoa shell[18], rice husk [19], modified sawdust of walnut[20], papaya wood[21], maize leaf[22], rice husk ash and neem bark[23], fly ash[24], and industry waste[25].

II. EXPERIMENTAL

Preparation of adsorbate

A stock solution containing 1000mg/l of Lead ions was prepared by dissolving an appropriate quantity of lead nitrate in double distilled water. The working solutions were prepared by diluting the stock solution. The concentration of the lead (II) ions in the solution was determined spectrophotometrically.

Preparation of adsorbent material

Centella Asiatic leaves were collected and washed with tape water several times to remove soil dust and finally washed with DD water. It is dried in sun shade. The dried leaves were powdered and soaked in con.H_2SO_4(1:1,w/w), for a day, then filtered and dried. The charred mass was kept in a muffle furnace at 400^0C,for 1 hour, it was taken out, ground well to fine powder and stored in vacuum desiccators. The Characteristics of the adsorbent is presented in the table-1

pH	6.5
Moisture Content, %	13.5
Ash Content, %	10.2
Volatile Matter, %	21.3
Water Soluble matter, %	0.45
Acid Soluble Matter, %	0.89
Porosity, %	48.1
Micropore volume cm^3/g	0.194194
Average pore width ^{0}A	28.2065
BET Surface Area, m^2/g	604.27

Table-1 Characteristics of the adsorbent

III. RESULTS AND DISCUSSION

Effect of pH

The pH is one of the important parameters influencing the adsorption process. The pH of feed solution was examined from solution a different pH levels, covering a range of 1.0-7.0. In the case of Pb(II) the maximum adsorption was obtained at pH 5.0 for both 10 and 50 mg/L. The pH of the working solution was controlled by adding 1NHCl or 1NNaOH solution. The uptake of the lead (II) ions at pH-5 was maximum (fig.1) at lower pH values, the H^+ ions compete with the metal cation for the adsorption sites. The heavy metal cations are completely released under extreme acidic conditions .With the increase in pH the adsorption also increases and reaches the maximum at pH 5. Hence the pH of the medium was maintained at 5 for further studies.

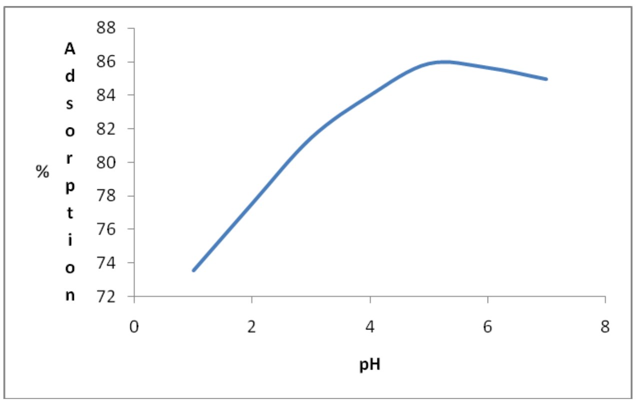

Fig:1 Effect of pH on the adsorption of Lead(II) ions on to the adsorbent

Effect of Adsorbent dose

The effect of adsorbent dosage was studied by varying the dosage from 25mg to 150mg keeping the other parameters constant. The result is shown in fig.2. The adsorption percentage increased as the adsorbent dosage increased. The adsorption percentage increases and reaches the maximum at 100mg of adsorbent dosage thereafter, there was no appreciable increase in the percentage of lead (II) ions removal. This is due to greater availability of the sites for adsorption on the adsorbent surface. Therefore the adsorbent dosage was maintained at 100mg for further studies.

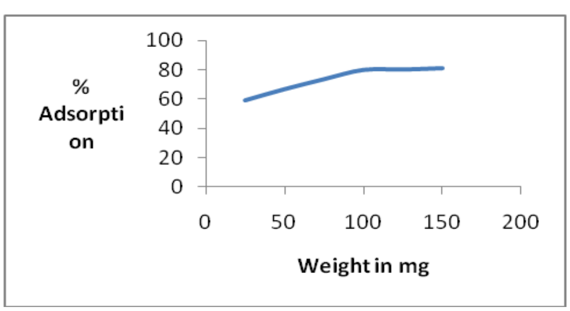

Fig:2 Effect of adsorbent dose on the adsorption of Lead (II) ions on to the adsorbent

Effect of metal ion concentration

The experiment was carried out with 10mg/l to 50mg/l solutions with adsorbent dosage 100mg, for a contact time of 90min at pH-5, and the result is shown in fig.3. From the figure it can be seen that the percentage of adsorption decreases with increase in the lead (II) ions concentration. This is due to less availability of sites on the adsorbent surface.

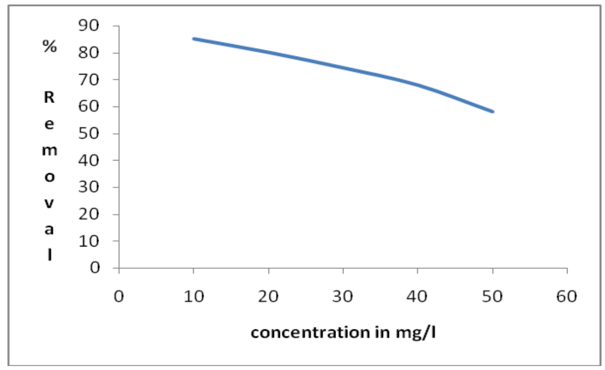

Fig:3 Effect of metal ion concentration on the adsorption of Lead (II) ions on to the adsorbent

Effect of contact time

The effect of contact time on adsorption of lead (II) ions removal is shown in fig.4. 100mg of adsorbent was used for this experiment with a contact time of 10 to 120minutes. It is observed that the percentage removal of metal increases with time and reaches the maximum at 90min, thereafter, there was no appreciable increase in the percentage of lead (II) ions removal. This is because at the initial stage, the rate of removal of lead (II) ions was higher due to availability of more number of active sites on the surface of the adsorbent and become slower after 90 minutes, due to availability of lesser number of active sites. Hence all the experiments were conducted for a period of 90 minutes.

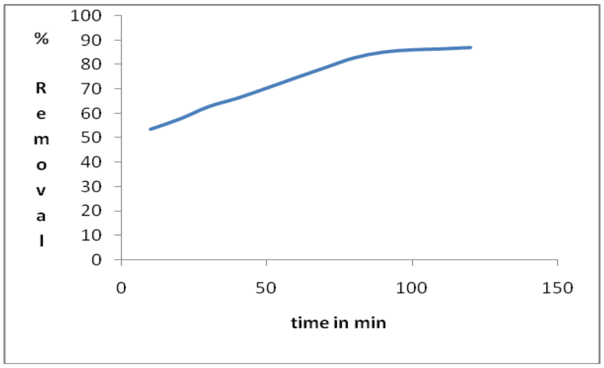

Fig:4 Effect of contact time on the adsorption of Lead (II) ions on to the adsorbent

Effect of Temperature

Effect of temperature on the adsorption of lead is shown in fig.5. As the temperature was increased from 30C^0 to 60C^0 the percentage of adsorption also increased. The increase in temperature may be accompanied with an increase in the porosity and in the total pore volume of the adsorbent. This may also be attributed an increase in the mobility of the metal ion with the increase of temperature.

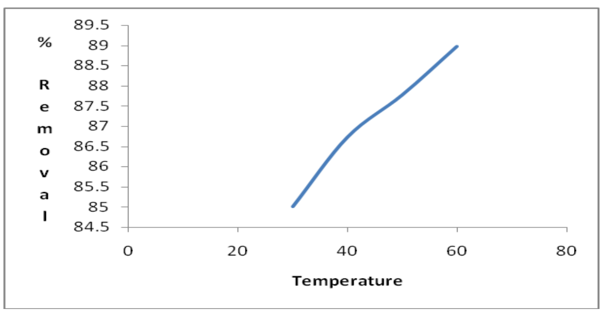

Fig:5 Effect of temperature on the adsorption of Lead(II) ions on to the adsorbent

IV. ADSORPTION ISOTHERMS

Langmuir Adsorption Isotherm

The equilibrium adsorption isotherms help to understand the mechanism of the adsorption. The Langmuir and Freundlich isotherms have been tested in this study.

The Langmuir isotherm was based on the assumption that maximum adsorption corresponds to a saturated monolayer of adsorbate molecules on the adsorbent surface, the energy of adsorption was constant and there was no transmigration of adsorbate in the surface.

The linear form of Langmuir equation[26] is expressed as follows

$$\frac{Ce}{Qe} = \frac{Ce}{Q_0} + \frac{1}{Q_0 b}$$

The values of Q_0 and b were calculated from the slope and intercept respectively, of the linear plots of C_e/Q_e verses C_e. Langmuir adsorption isotherm is presented in fig.6. Higher value of correlation co-efficient (R^2-0.999) indicates that the experimental data fits well with the Langmuir equation. The values of Q_0 and b are given in Table-2.

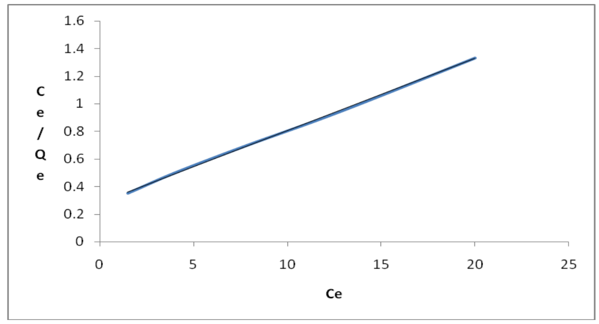

Fig:6 Langmuir isotherm for the adsorption of Lead (II) ions on to the adsorbent

Qo(mg/g)	b	R^2
19.2307	0.1850	0.999

Table-2. Langmuir constants

The essential characteristics of the Langmuir adsorption isotherm are expressed by a dimensionless constant called separation factor. This value indicates whether the adsorption is favorable or not. R_L is defined by the following equation

$R_L = 1/(1+bCo)$

Where,R_L _ dimensionless separation factor[27], Ci – intial concentration , b – Langmuir constant (Lmg-^{1}),The parameter $R_L$indicates the type of the isotherm.

Values of R_L	Types of isotherms
$R_L>1$	Unfavourable
$R_L = 1$	Linear
$0<R_L<1$	Favourable
$R_L = 0$	Irrevisible

The R_L value obtained using the above equation for 10mg/l Lead (II) ions concentration is 0.3508. This R_L value lies between 0 and 1 indicating the favorability of the adsorption.

Freundlich Adsorption Isotherm

The Freundlich model can be applied for non-ideal sorption onto heterogeneous surfaces involving multilayer sorption.

The linear form of the Freundlich equation[28] is as follows

$$\log Qe = \log K_F + \frac{1}{n}\log Ce$$

Where Qe , amount of metal ion adsorbed (mg/g), K_f ,adsorption capacity, n, adsorption intensity. By ploting logQe Vs logCe, the values of n and K were calculated from slope and intercept respectively (fig.7). The values of K_f and n are given in table-3. The value of linear regression co-efficient (R^2) was found to be 0.974. This indicates that the adsorption process follows Langmuir adsorption isotherm more than Freundlich adsorption isotherm.

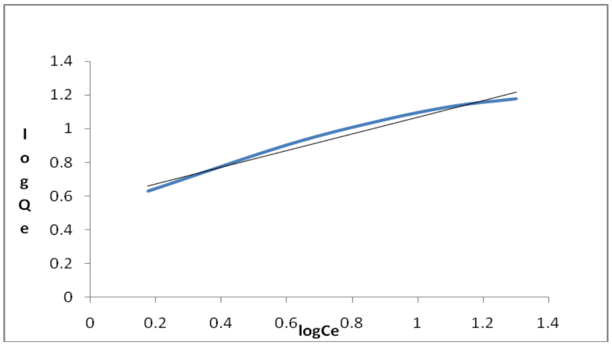

Fig:7 Freundlich isotherm for the adsorption of Lead (II) ions on to the adsorbent

n	K_f(mg/g)	R^2
2.0283	3.7583	0.974

Table-3. Freundlich constants

The value of n in the range 2-10 represents favourable adsorption.

V . THERMODYNAMIC PARAMETERS

Thermodynamic studies related to the adsorption process is essential to conclude whether a process will occur spontaneously or not. The fundamental criteria for spontaneity is the standard Gibbs free energy change ΔG^o, if the ΔG^o value is negative, the reaction will occur spontaneously. The thermodynamic parameters, standard free energy (ΔG^o), change in standard enthalpy change (ΔH^o) and change in standard entropy(ΔS^o) for the adsorption of lead (II) ions onto the adsorbent were calculated using the following equations.

$$K_0 = \frac{C_{solid}}{C_{liquid}}$$

$$\Delta G^0 = -RT \ln K_0$$

$$\log K_0 = \frac{\Delta S}{2.303R} - \frac{\Delta H}{2.303RT}$$

Where Ci is the concentration of the metal ion at equilibrium and Ce is the amount of metal ion adsorbed on the adsorbent

The values of ΔG^o (KJ/mol), ΔH^o(KJ/mol^{-1}) and ΔS^o (J/K/mol) can be obtained from the slope and intercept of a linear plot of log K_o verses 1/T and are presented in table-4.

Conc. of Pb (mg/l)	$-\Delta G^o$				ΔH^o	ΔS^o
	30^0 C	40^0 C	50^0 C	60^0 C		
10	4.3732	4.8867	5.2951	5.7817	9.5716	46.04
20	3.4937	4.0027	4.5450	4.9018	10.8908	47.52
30	2.6681	3.0468	3.3625	3.7675	8.1854	35.80
40	1.8820	2.2054	2.5690	2.9049	8.4324	34.00
50	1.0210	1.1081	1.4756	1.7087	6.2611	23.83

Table-4. Thermodynamic parameters for the adsorption of Lead (II) ions on to the adsorbent.

The negative values of ΔG^o indicates that the adsorption process is spontaneous and highly favorable . The positive values of ΔS^o indicates the increased randomness at the solid solution interface. The values of ΔH^o indicates that the adsorption process is endothermic and physical in nature.

VI. CONCLUSION

The adsorption behavior of Lead (II) ions on the adsorbent obtained from the *Centella asiatica leaves* was investigated in batch equilibrium method. The adsorption was found to be highly dependent on various parameters like contact time, pH, initial concentrations, adsorbent dose and temperature. The optimum pH for Lead (II) ions adsorbent was found to be 5. The experimental data were correlated well by the Langmuir adsorption isotherm. The result of this study indicates that this adsorbent can be successfully utilized for the removal of Lead (II) ions from aqueous solution.

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