

Studies on Removal of Arsenic from water by adsorption on Laterite and Ferric-Hydroxide coated Bentonite

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ABSTRACT:

The present work evaluates the suitability of two natural materials, viz., laterite and iron oxide coated bentonite as potential arsenic adsorbents. The materials were compared with respect to different parameters such as, pH, arsenic dosage, adsorbent concentration and contact time. The obtained experimental results were fitted to four different isotherms namely the Freundlich, Langmuir, Temkin and the Dubinin-Radushkevich isotherms. Optimum pH in the two cases was 2.13 and 4.1 for laterite and bentonite respectively. Arsenic removal was found to increase with an increase of adsorbent concentration throughout the range for bentonite, whilst for laterite, percentage arsenic removal decreased with increasing adsorbent concentration beyond maxima at 8g/l of adsorbent. Increase in arsenic concentration was found to cause a decrease in percentage arsenic removal in both cases. Increasing the contact time of the process was found to increase the amount of arsenic removed. The equilibrium removal of arsenic was found to be higher for the ferric-hydroxide coated bentonite. The equilibrium study yielded that the Temkin and the Dubinin-Raduskevich isotherms best fit the data for adsorption onto laterite and bentonite surfaces respectively. Kinetic data was best fitted by the pseudo second order rate equation in both cases.

Keywords: Adsorption, Arsenic, laterite, bentonite, adsorption models

INTRODUCTION

Arsenic, the 20th most plentiful element in the earth's crust, is also considered as one of the most toxic elements. It is a yellowish to white solid occurring in both metallic and nonmetallic forms. Chemically present in 2 oxidation states: +3(arsenite) and +5(arsenate), it enters the food chain due to leaching from rocks and minerals. Anthropogenic sources of arsenic include industrial discharge and agricultural run-off (mainly from pesticides)[20]. Arsenic may enter the human body through ingestion, inhalation, absorption through skin and mucous membranes [1, 2, 3]. Arsenic in drinking water is a serious problem in large part of *Gangetic* Bengal and asks for interventions from Governmental and non-governmental agencies.

Arsenic present in drinking water causes serious health hazards. The World Health Organization (WHO) recommends an upper limit of 10ppb of arsenic in groundwater. Conventional arsenic removal techniques include chemical precipitation, solvent extraction, dialysis, nano filtration and reverse osmosis, co-precipitation, polymer ligand exchange, ion exchange and adsorption [5, 6].

The presence of Arsenic in potable water with repeated consumption rate could invite severe health abnormalities in humans. Continuous consumption of arsenic –rich water may lead to vascular diseases like gangrene, reproductive abnormalities, elevated blood pressures etc [22].

Also it has been reported that hyperpigmentation, keratosis, cancers are the manifestations of chronic arsenicism towards the human body from any source [23]. In support to that, diseases like black-foot disease and skin cancers was found to be linearly related with the dosage concentration of arsenic in consumable water [24].

Thus if prevalent in the drinking water, there is an augmented probability that with the larger proportion of oral exposure of arsenic content in the water, the toxicological impact over the different body tissues are inevitable as because with the relatively higher proportions of oral exposure of arsenic rich water, methylation capacity may not be adequate to prevent the cytotoxic levels of Arsenic (III) from reaching the tissues. Arsenic reacts with the sulfhydryl groups of proteins, inactivates enzymes, and interferes with mitochondrial function by inhibiting succinic dehydrogenase activity and uncoupling oxidative phosphorylation [25]. Besides that, it has been proposed that arsenic may compete with phosphate during oxidative phosphorylation and may inhibit energy-linked reduction of nicotinamide adenine dinucleotide (NAD).

Nature has given human body protective resources against intruders by chelating the toxic intruders out. Metallothionein, a natural antioxidant in human systems can chelate the produced free radicals in the human body caused due to the intake of arsenic-rich water and thus preventing the inception of dreaded diseases like cancers etc. The present study is designed to find out a low cost chelator suitable to emerging economy like India and its applicability to rural backgrounds to ensure supply of consumable water without arsenic contamination and by designing an effective chelator mechanism made up of laterite and ferric hydroxide coated bentonite.

With the aid of this effective chelator mechanism of the present study, the management of safe drinking water could be achieved in the arsenic polluted environment. Arsenic removal by adsorption has gained immense popularity due to its wide applicability (can be used in domestic as well as large scale operations) and simplicity of operation. Adsorption also provides economic advantages over the other methods of arsenic removal [3, 4, 18, 11]. Materials used in the study are cheap and easily available.

Conventional adsorbents include activated carbon, zeolites or molecular sieves, silica gel and activated alumina [13]. However these adsorbents are expensive. In contrast laterite and bentonite are naturally available. Their ion exchange potential and high adsorption properties exhibit efficacy of these adsorbents [8, 15, 16, 17].

Furthermore, natural laterite is a composite material of iron, aluminum and silicon (Table 1), formed due to intensive chemical weathering under oxidizing conditions [21, 7]. Iron oxides present impart a reddish colour to laterite. It is widely available in many areas of West Bengal, especially in the districts of West Midnapore, Bankura and Purulia. In our study, laterite collected from Bankura district was applied for adsorption of As (V) from stock solution.

Bentonite is a type of aluminum phyllosilicate containing mostly montmorillonite [14], the composition of typical bentonite samples shown in Table 2. It usually forms from weathering of volcanic ash in presence of water [9, 10, 12]. In this study the efficiency of bentonite was compared with laterite and as the major constituent of laterite is iron oxide bentonite was chemically modified by impregnating with iron oxide salts.

Present study encompasses a comparison between the performances of these two adsorbents in Arsenic removal from stock solution. The study also emphasizes on the removal of arsenic (As(V)) at varying conditions of pH, adsorbent dosage, metal ion concentration and contact time.

Table 1 : Chemical composition of laterite [21]

Sl. No.	Name of Chemical	Percentage
1	AL ₂ O ₃	19,4%
2	Fe ₂ O ₃	25.75
3	TiO ₂	2.55
4	SiO ₂	34.40
5	CaO	0.45
6	Na ₂ O	0.30
7	K ₂ O	8.80
8	P ₂ O ₅	0.45
9	MgO ₃	4.60

Table 2: Chemical composition of bentonite [21]

Sl. No	Name of chemical	Percentage
1	AL ₂ O ₃	20.27%
2	Fe ₂ O ₃	9.08%
3	TiO ₂	0.78%

4	SiO ₂	54.82%
5	CaO	2.10%
6	MgO	3.02%
7	Na ₂ O	1.31%
8	K ₂ O	0.06%

2. EXPERIMENTAL SECTION

2.1 REAGENTS

All chemicals used were of analytical grade and used without further purification. Deionized water was used to prepare all aqueous solutions. Concentrated H₂SO₄, HNO₃, NaOH pellets were used as received. (NH₄)₆Mo₇O₂₄.4H₂O (100mg/ml was used as stock solution). Na₂S₂O₅ was used in solid form. Stock solutions (1000ppm) were prepared using Na₂HAsO₄.7H₂O. Appropriately diluted solutions were made from the stock as and when required.

2.2 EXPERIMENTAL SET UP

All glass wares were washed in deionised water and dried in a hot air oven. Digital pH meter (Hanna Instruments) was used for all pH measurements. A high precision electrical balance was used for weighing. Magnetic stirrer (REMI 1MLH) was used for agitation purpose. Final arsenic concentration was determined by spectrophotometric method.

2.4 PREPARATION OF THE ADSORBENT

Locally available laterite soil procured from the Bankura district of West Bengal was used as an adsorbent. The soil was first washed several times with deionized water and then sun dried for 2-3 days. It was then crushed in hammer mill and then sieved to obtain a size range of - 0.211+0.152 mm. The soil was then washed with 1M H₂SO₄, followed by 1M NaOH and finally with distilled water. Final drying of soil was carried out in a hot air oven at 393K for 4 hours.

Bentonite clay available in granular form was grounded. Clay samples were weighed and dried for 72hrs at 313K until it is completely dry to prevent any alteration of sample mineralogical properties. The soil was then washed with 1M H₂SO₄, followed by distilled water. To impregnate bentonite samples with Ferric Hydroxide, 5.44gm of ferric chloride (FeCl₃.6 H₂O) was added to 200mL of distilled water. To this mixture ammonia was added drop wise to precipitate ferric hydroxide [Fe(OH)₃]. Solution was stirred at 500 rpm to ensure uniform coating on surface of bentonite particles. Solution was then filtered and air dried at 393K for 4 hours. Dry coated bentonite was then grounded, collected in the size range of 0.211+0.152 mm and stored in air tight containers for further use.

2.5 EXPERIMENTAL STUDIES

Batch experiments were performed at 303 ± 1 K at an average speed of 180 rpm. To find optimum adsorbent dose, arsenic contaminated water (50 mL) with varying adsorbent dosage (2-20gm/L) was agitated for 30 mins. Samples were allowed to settle overnight and then centrifuged at 10000 rpm to collect the supernatant. The aliquot was used for the analysis of the remaining arsenic in the solution.

2.6 ANALYTICAL METHODS

Quantification of Arsenic was made by the following method: filtrate (1mL) was added to $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24} \cdot 4\text{H}_2\text{O}$ (4.5mL, 100mg/mL) followed by addition of 0.705gm of $\text{Na}_2\text{S}_2\text{O}_5$. Resultant solution was warmed using hot water giving a molybdenum blue color after 10 min. Absorbance was measured at 690nm using reagent blank as reference.

2.6 BATCH ADSORPTION STUDY

2.6.1 EFFECT OF PH

To study the effect of pH on adsorption of As (V) on both modified bentonite and laterite was performed with an initial arsenic concentration of $100 \mu\text{g}/\text{mL}$. pH was varied from 2.1 - 8 using 0.1M HNO_3 and 0.1M NaOH . A digital pH meter (HANNA instruments, USA, Model No. H198107) was used to check pH measurements. Samples were stirred in a glass bottle using a magnetic stirrer at an average speed of 180 rpm for 30 minutes. Contact time of 24 hrs was allowed which is considered to be sufficient to reach equilibrium. Samples were then doubly centrifuged and analyzed for residual arsenic content by method described previously.

2.6.2 EFFECT OF ADSORBENT DOSAGE

Batch study on the effect of adsorbent dosage on adsorption of As (V), both on laterite as well as bentonite was performed with initial arsenic concentration of $5 \mu\text{g}/\text{mL}$. pH was maintained at optimum values for adsorption on both laterite and modified bentonite. Samples were stirred as before for 30 mins and a contact time of 24hrs was allowed. Samples are analyzed using the above mentioned method.

2.6.3 EFFECT OF ARSENIC CONCENTRATION

Batch study on effect of adsorbate dosage on adsorption was performed with adsorbent concentration of 2gm/L. Concentration of arsenic solution was varied from $1 \mu\text{g}/\text{mL}$ - $5 \mu\text{g}/\text{mL}$ in both cases. For each adsorbent pH was maintained constant at obtained optimum value. Studies were performed according to batch adsorption experiment. Samples were analyzed for residual arsenic content.

2.6.4 EFFECT OF CONTACT TIME

Batch study on effect of contact time on adsorption was performed with initial adsorbent concentration of 2gm/L. pH was kept constant at optimum values. Adsorbate concentration was

kept constant at 5µg/L. Samples were stirred in glass bottle using magnetic stirrer at average speed of 180 rpm for 3mins. Contact time varying from 30mins to 3hrs was allowed. Samples were then filtered using regular filter paper and analyzed.

2.6.5. EQUILIBRIUM STUDIES

In order to carry out the equilibrium studies for both cases, the initial concentration of arsenic solution was varied from (1-5µg/ml). Adsorbent dose was kept constant at 2g/L and the pH of the solution was maintained at the optimum values. Stirring speed was maintained at 180 rpm. Temperatures 303±1K and an equilibrium time of 24 hrs were applied to the study. The equilibrium data was modeled by five adsorption isotherms viz., Langmuir, Freundlich, Temkin and the Dubinin-Radushkevich isotherms.

2.6.5.1. LANGMUIR ISOTHERM

The Langmuir model assumes monolayer adsorption.

The linearized form of the Langmuir isotherm is given by:

$$\frac{1}{q_e} = \frac{1}{a \cdot b} \times \left(\frac{1}{C_e} \right) + \frac{1}{b} \quad (1)$$

a and b are Langmuir constants indicating the energy of adsorption and the maximum adsorption capacity respectively. The efficiency of adsorption can be determined by the equilibrium parameter (r), a dimensionless parameter defined by

$$r = \frac{1}{1 + bC_0} \quad (2)$$

A value of $r < 1$ indicates favorable adsorption [19].

2.6.5.2. FREUNDLICH ISOTHERM

The linearized Freundlich isotherm can be represented as:

$$q_e = k_f C_e^{\frac{1}{n}} \quad (3)$$

Hence, a plot of $\log q_e$ vs $\log C_e$ would yield a straight line with slope equal to $1/n$ and intercept equal to $\log k_f$.

2.6.5.3 TEMKIN ISOTHERM

The Temkin isotherm enumerates the relation between the amount of adsorbate adsorbed and the equilibrium concentration of the adsorbate. The linearized Temkin isotherm is expressed as:

$$q_e = \frac{RT}{b_t} \ln(a_t \times C_e) \quad (4)$$

Here, R is the universal gas constant in J/mol.K, T is the absolute temperature at which the experiment was conducted, a_t and b_t is constants.

2.6.5.4. DUBININ–RADUSHKEVICH(D.R) ISOTHERM

The D.R isotherm can explain the mechanism of adsorption. The linearized form of this isotherm is given by:

$$\ln q_e = \ln q_m - k\varepsilon^2 \quad (5)$$

where ε is the Polanyi potential $\left[RT \ln \left(1 + \frac{1}{C_e}\right)\right]$, q_m is the maximum adsorption capacity in mol/g, q_e is the amount of arsenic adsorbed at equilibrium per unit weight of adsorbent in mol/g. Free energy change can be evaluated using the expression,

$$E = -(2k)^{-0.5} \quad (6)$$

Value of E ranges from 1-8 kJ/mol for physisorption; from 8-16 kJ/mol for chemisorptions [15].

3. RESULTS AND DISCUSSIONS

3.1 EFFECT OF PH

In order to observe the effect of pH on adsorption of As (V) percentage removal of As (V) as a function of pH was studied.

Arsenic removal by both laterite and bentonite is completely governed by pH of the solution. From Fig. 1 it is evident that maximum removal of arsenic was achieved at pH 2.13 for laterite and 4.1 for bentonite. As (V) exists in anionic forms H_2AsO_4^- or HAsO_4^{2-} . Hence arsenic species get attracted to positively charged laterite and bentonite surfaces. pH_{zpc} of laterite soil is 6.96 as obtained from literature. Thus above this pH value, the laterite surface acquires negative charge and repels As (V) anions. To explain the optimum pH of surface modified bentonite, it can be attributed to the fact that at high pH values (>8) As (V) exists as an anion H_2AsO_4^- and the bentonite surface becomes increasingly negatively charged which finally decreases adsorption capacity.

3.2. EFFECT OF ADSORBENT DOSAGE

The two adsorbents exhibit markedly different behavior when adsorbent dosage is varied from 2-20g/L. Whilst the percentage removal of arsenic by bentonite enhances continuously throughout the range, laterite shows a maximum removal at 8g/L of adsorbent dose and after that it decreases which closely corroborates the previous work [22].

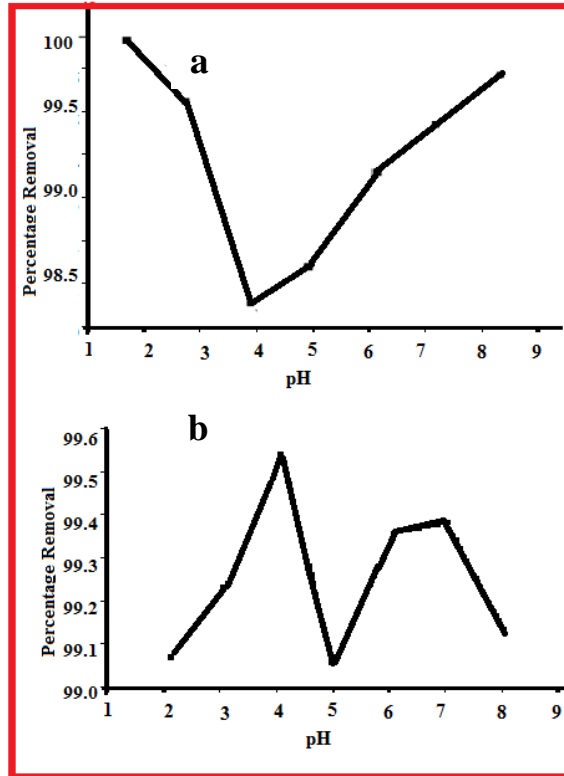


Fig.1 Variation of The %ge removal of Arsenic with pH of the solution for laterite (a) and bentonite (b) adsorbent. Conditions: $C_0 = 5.0$ mg/L; pH = 4.1; shaker speed = 180 rpm; contact time = 24 hrs.

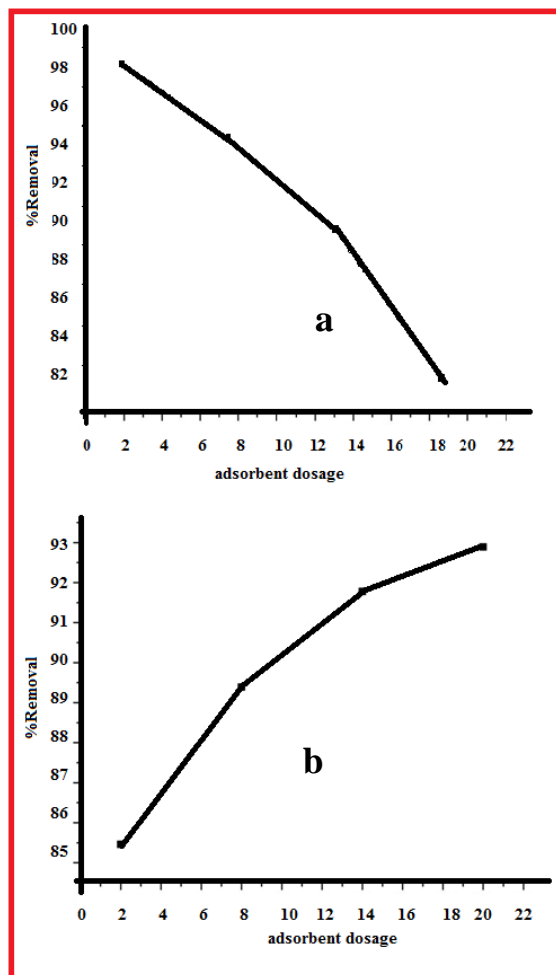


Fig. 2 Variation of Percentage removal of Arsenic with dosage of Laterite (a) & bentonite (b) in solution. Conditions: $C_0 = 5.0$ mg/L; pH = 4.1; shaker speed = 180 rpm; contact time = 24 hrs.

3.3. EFFECT OF ARSENIC SOLUTION CONCENTRATION

In both cases, the percentage removal of As (V) decreases with an increase in initial concentration of arsenic solution. This is due to the fact that the adsorption capacity and the amount of adsorbent remains the same, in spite of the increase in As (V) concentration.

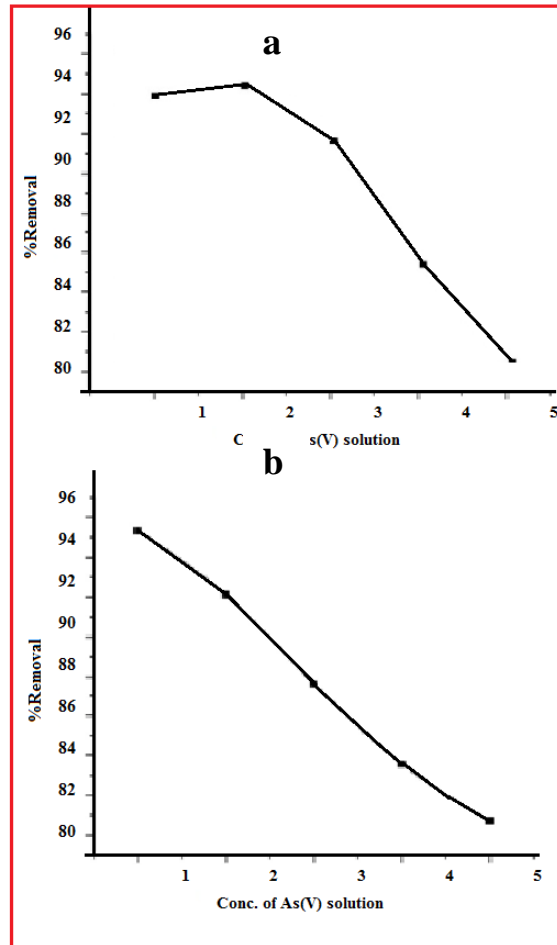


Fig. 3 Variation Of % removal of arsenic with arsenic concentration in stock solution for adsorption onto laterite(a) and bentonite(b). Conditions: $C_0 = 5.0$ mg/L; pH = 4.1; shaker speed = 180 rpm; contact time = 24 hrs.

3.4. EFFECT OF CONTACT TIME

Both laterite and bentonite exhibit an increase in uptake of As (V) ion with contact time.

However, for bentonite, a sharp decrease in the % removal is observed after 180 min. This may be due to saturation of the adsorbent sites.

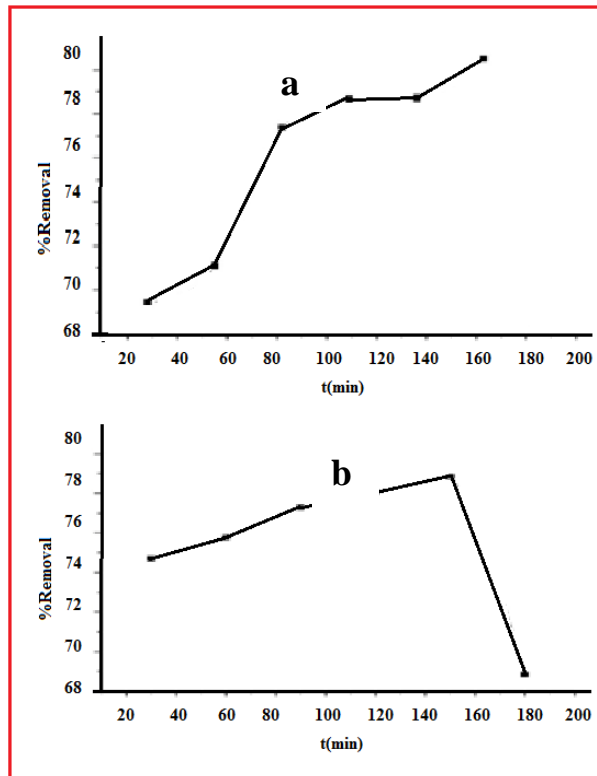


Fig.4 Variation Of %removal of arsenic with contact time (min) for laterite(a) and bentonite(b). $C_0 = 5.0$ mg/L; pH = 4.1; shaker speed = 180 rpm; contact time = 24 hrs

3.5. EQUILIBRIUM STUDIES

The equilibrium data obtained was fitted onto four adsorption isotherms, namely the Langmuir, Freundlich, Temkin and the Dubinin-Rasdushkevich (DR) isotherms.

The parameters for each isotherm model as observed in the adsorption of As (V) on laterite and bentonite are given in the following tables (3 and 4).

In case of Langmuir isotherm, $r = 0.0546$ & 0.0924 for adsorption onto laterite and bentonite surfaces respectively. Hence, adsorption of As (V) onto both laterite & bentonite surfaces is favorable in nature. Value of n obtained from Freundlich isotherm also supports favorable adsorption of As (V) on both laterite and bentonite.

Table 3: Comparative study of the four adsorption isotherms for adsorption of arsenic onto laterite surface

Sl. No	Name of the Isotherm	Parameter 1	Parameter 2	R^2
1	Langmuir		$b = 3.460$	0.978
2	Freundlich	$a = 2.449$ $k_f = 0.1919$	$1/n = 0.597$	0.933
3	Temkin	$a_t = 2.556 \times 10^6$	$b_t = 3.0643 \times 10^8$	0.995
4	Dubinin – Radushkevich	$q_m = 5.389 \times 10^{-4}$	$k = 3 \times 10^{-9}$	0.945

Table 4: Comparative study of the four adsorption isotherms for adsorption of arsenic onto bentonite surface

Sl. No	Name of the Isotherm	Parameter 1	Parameter 2	R^2
1	Langmuir	$a = 6.7867$	$b = 1.9646$	0.987
2	Freundlich	$k_f = 2.07508$	$1/n = 0.469$	0.996
3	Temkin	$a_t = 1.6 \times 10^6$	$b_t = 3.5987 \times 10^8$	0.96613
4	Dubinin-Radushkevich	$q_m = 2.735 \times 10^{-4}$	$k = 2.891 \times 10^{-9}$	0.99813

It is evident from the experiments, that the data for adsorption onto laterite was the best fitted by the Temkin isotherm whilst that for adsorption onto bentonite surface was most well fitted by the D-R isotherm. Linearized plots of both the best fitted isotherm data were shown in the following figures (Figure 5 and 6). Additionally, for arsenic adsorption E comes out to be 12.909 kJ/mol & 13.15 kJ/mol on laterite & bentonite surfaces respectively. Thus, we can conclude that adsorption of arsenic on laterite & bentonite takes place via chemisorptions route.

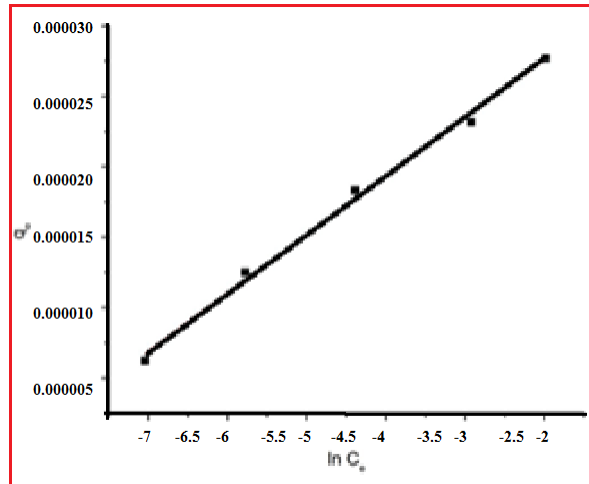


Fig 5. Plot of q_e versus $\ln C_e$ for adsorption onto laterite. Conditions: $C_0=5.0$ mg/L; pH = 4.1; shaker speed = 180 rpm; contact time = 24 hrs

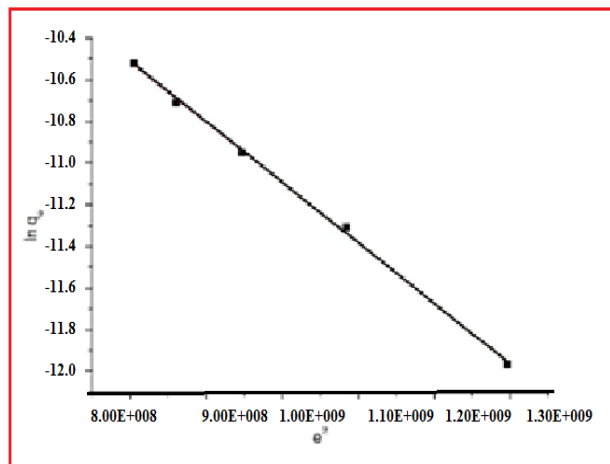


Fig 6. Plot of $\ln q_e$ versus e^2 for bentonite. Conditions: $C_0 = 5.0$ mg/L; pH = 4.1; shaker speed = 180 rpm; contact time = 24 hrs

3.6. KINETIC STUDIES

The kinetics of sorption of As (V) was studied using pseudo 1st order equation by Lagergen. It can be represented as

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (7)$$

Where, q_e and q_t are the values of sorption capacities at equilibrium (in mg/g) and at time (t) and k_1 is the rate constant(/min).

The rate of sorption, if it follows a second order mechanism, can be modeled by the pseudo-second order rate equation:-

$$\frac{dq_t}{dt} = k(q_e - q_t)^2 \quad (8)$$

Where q_e is the equilibrium adsorption capacity, q_t is the adsorption capacity at time t and k is the rate constant (g/mg min).

For the present study, the kinetic data was fitted with the first and the second order models, for both laterite and bentonite, it was observed that, the experimental kinetic data shows the best fit with second order kinetics (correlation coefficient R^2 values =0.99 in both cases). This is evident from the correlation coefficient (R^2) values for the second order rate equation which is significantly higher than the correlation coefficient for the first order model (=0.7675 for laterite and 0.855 for bentonite).

The results indicate that the kinetic behavior of adsorption is governed by the whole duration of the process. The first order equation is generally pertinent only for the first few minutes of the process and hence, cannot be effectively used to predict the kinetic behavior.

An indigenous apparatus with modified bentonite and another with laterite is prepared for house hold use in arsenic affected area to get safe consumable water. The results show in normal conditions at house hold in rural Bengal the apparatus is effective in removing arsenic; the different conditions of the experiment will set the efficiency.

4. CONCLUSION

The adsorption of arsenic onto laterite and bentonite surfaces was thus studied. Effect of pH, adsorbent dosage, metal ion concentration and contact time on the adsorption process was also investigated. The equilibrium data was fitted to four isotherms, viz. the Freundlich, Langmuir, Temkin and the D-R isotherm. The Temkin isotherm shows the best fit for adsorption onto laterite and the D-R isotherm showed the best fit for adsorption onto bentonite. For both cases, the pseudo second order kinetic equation showed the best fit for the experimental data. It has also been observed that surface modified bentonite exhibited the maximum removal of As (V) and thus proved to be superior in comparison with laterite. Thus these natural existing substances can have some applied significance in home-made use with indigenous support to provide safe and portable consumable water.

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