



Laterite-A Potential Alternative for Removal of Groundwater Arsenic

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ABSTRACT : Arsenic removal by heat treated laterite from contaminated water was investigated through batch adsorption experiments. The removal rate was dependent on the initial arsenic concentrations and a high initial rate of removal followed by a slower subsequent removal rate with a gradual approach to a steady-state condition. Rate kinetics was studied using both first-order and pseudo-second order models, and intraparticle diffusion from the solution to the adsorption sites was considered as major rate controlling step. Adsorption equilibrium data pointed to favorable adsorption of arsenic onto laterite and fitted with both Langmuir and Freundlich models. Thermodynamic data suggested chemical nature of the adsorption. Experimental data were used to estimate the life-time of laterite as a column packing-adsorber with the simplest assumptions and for typical initial concentration ($\sim 0.30 \text{ mg L}^{-1}$ arsenic) it was 74 days maximum while to achieve WHO safe limit (0.01 mg L^{-1}). Laterite is a natural substance and can be collected very cheaply, so its utilization for arsenic removal is expected to be economical and feasible. It might be a promising alternative of other proposed arsenic removal media for the arsenic-affected region of the world. @ JASEM

Elevated levels of groundwater arsenic, that can have severe human health implications, are reported from all over the world (Pontius et al., 1994, Lian and Jian, 1994, Jekel, 1994, Berg et al., 2001). However, the scale of contamination is grave in the Bengal Delta Plain (West Bengal of India, Bangladesh) in terms of toxicity and geographical distribution (Chowdhury et al., 2000). Arsenic toxicity has no known effective treatment, but drinking of arsenic free water can help arsenic affected people at early stage of ailment to get rid of it. There is, therefore, an urgent need to provide low-cost as well as small-scale treatment system that could be implemented at household or community levels. A number of treatment options are available with demonstrated efficiency for arsenic removal which include coagulation (Shen, 1973, Sorg and Logsdon, 1978, Hsia et al., 1994, Cheng et al., 1994, Edwards, 1994, Scott et al., 1995), softening (McNeill and Edwards, 1995), adsorption on alumina (Belleck, 1971, Gupta and Chen, 1978, Ghosh and Yuan, 1987, Hathaway and Rubel, 1987) or activated carbon (Gupta and Chen, 1978, Huang and Fu, 1984), anion exchange (Clifford, 1990, Ramana and Sengupta, 1992) and reverse osmosis (Fox and Sorg, 1987, Fox, 1989). Hardened paste of Portland cement (Kundu et al. 2004), activated red mud (Altundogan et al., 2002) are examples of some recent efforts proposed to remove arsenic from water. The aforementioned arsenic treatment technologies have been successfully applied to remove trace arsenic concentrations from contaminated water, but most of them are cost-prohibitive for communities or developing countries. An iron-aluminum-complex rich red soil-laterite would be a potential candidate for cheap arsenic removal agent since it has showed sufficient capacity of arsenic sorption in our preliminary studies. Present work focused on the different aspects of arsenic contaminated water treatment by laterite.

MATERIALS AND METHODS

Collection and Treatment of Laterite

Laterite-soil collected from Ogasawara Island (N 27°24', E 142°13'), Tokyo, Japan was used in the study. Total 56%, 15% and 29% of oxygen, carbon and iron-aluminum-silica (Fe-Al-Si) content were explored through surface analysis of laterite by ESCA. Compositions of Fe-Al-Si at surface were Fe, 2%, Al, 43% and Si, 55%. Laterite with particle size 355-710 μm showed better efficiency with column packing during preliminary studies, and as we planned to use the result obtained from the present study to design a column-packed arsenic removal module, laterite with particle size 355-710 μm was used throughout the study. The definite particle sized laterite was obtained by grinding the raw laterite sample followed by sieving, which was then heat treated in air atmosphere at 500°C to remove the organic material remaining in the raw laterite. The heat treatment condition was also predetermined through series of preliminary studies.

Water Sample Preparation

Measured amount of standard arsenic solution, supplied by Kanto Chemical Co., INC., Tokyo, Japan, is added with distilled water in the 500 ml/1000 ml volumetric flask and made up to the mark to prepare the sample solution. The artificially prepared arsenic-contaminated water is used throughout the study.

Experimental Setup

Batch experiments were conducted in the closed-vessel type assembly. A 300 ml flask was used as the vessel. Rotating impeller, inserted into the flask through the top-inlet, was used to ensure vigorous stirring. Impeller rotation speed was measured by a tachometer, and fixed at certain values. Temperature-controlled water bath was used to ensure the constant temperature environment.

Experimental Parameters

Laterite amounts, agitation speeds and initial arsenic concentrations were the variables studied, selected on the basis of their probable effect on the arsenic removal efficiency of laterite. Series of experiments were conducted to determine the most suitable laterite dose and steady removal tendency was observed with the laterite dose of 5 g L^{-1} which was considered as standard for the next set of experiments. The effect of agitation speed on the q of laterite was studied through series of experiment and it was observed that arsenic-spiked water and laterite contact was better ensured at the 400 rpm speed, and was kept fixed for the next set of experiments. Experiments were conducted at four different initial arsenic concentrations (0.3163 , 0.5654 , 0.8484 , 1.0086 mg L^{-1}) at 308K with pre-determined laterite dose (5 g L^{-1}) and agitation speed, $400 (\pm 5) \text{ rpm}$. Equilibrium study, adsorption rate and other calculations were done with that set of experimental data. Three different temperatures: 298 K , 308 K , 318 K were considered for the calculations of adsorption isotherms and thermodynamic parameters.

Arsenic Analysis

Arsenic concentrations in the initial and laterite-treated water samples were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS), a standard method for the determination of trace arsenic in water with estimated detection limit of 25 pg g^{-1} (APHA, 1989).

Data Optimization and Illustration

MS Excel 2003 SP1 was used for data reduction and optimization. Sma4 for windows95 ver. 1.48E was used for data illustration.

RESULTS AND DISCUSSION

Equilibrium Study: Contact Time and Initial Arsenic Concentration

A time-limited study with varying initial arsenic concentration was conducted to find out the time required to attain the equilibrium during laterite-arsenic contact. Variation in the residual arsenic

concentration with contact time at different initial concentrations is shown in Figure 1 and it is evident that the decrease-rate of arsenic from bulk solution was dependent on the initial arsenic concentrations. The figure shows a high initial rate of removal followed by a slower subsequent removal rate that gradually approached an equilibrium condition. The high concentration difference between the bulk solution and adsorption sites initiate the rapid removal at the initial hours while the rate tend to be slowed down with the saturation of the adsorbent sites at the late hours. It was observed in all the concentrations studied that the concentration of arsenic decreased with time up to 72 hours (approximately) and then the curves seem to be flattened i.e. approaching to the equilibrium. Based on these findings, the contact time of 96 hours was used in all further experiments.

In Figure 2, arsenic removal rate (%) as a function of time is shown and it is observed that approximately 70% arsenic from the bulk solution was adsorbed by laterite within the initial 24 hours. Though the increase in initial arsenic concentration decreases the rate slightly, the ultimate result can be assumed almost equal for all the tested conditions i.e. the arsenic removal capability of laterite for first 24 hours can be considered independent of initial arsenic concentration. The approximation is a qualitative one, though.

The arsenic concentration in the adsorbent phase, q_t , in $\text{mg-As (g-adsorbent)}^{-1}$ is calculated using the following equation (Onyango et al., 2003):

$$q_t = (C_i - C_t) \frac{V_f}{m} \quad (1)$$

where, C_i and C_t are the initial concentration and concentration of adsorbate in solution at any time [mg L^{-1}], V_f is the volume of the solution [L] and m is the mass of the adsorbent [mg]. Figure 3 shows the adsorbent phase concentration against time. An increase in the amount of adsorbed arsenic was observed with an increase in initial concentrations.

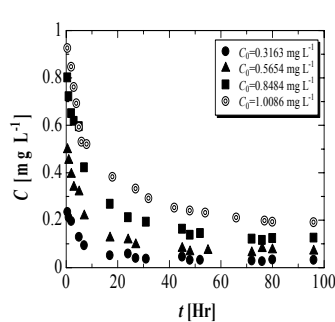


Figure 1

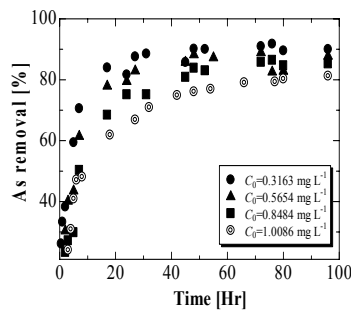


Figure 2

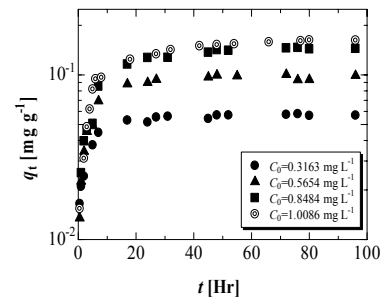


Figure 3

Adsorption Dynamics

The adsorbent phase concentrations of arsenic against time data were further processed to derive the rate equations. Lagergren's first order rate equation and Ritchie's pseudo-second order rate equation were used to describe the kinetics of arsenic uptake by laterite. Lagergren's first order rate equation (Lagergren and Bil, 1898) is in the form:

$$\ln(1 - q_t/q_e) = -k_1 t \quad (2)$$

where, k_1 is the rate constant of a first order adsorption reaction [min^{-1}], q_t is the amount of arsenic adsorbed at any time [$\text{mg-As (g-adsorbent)}^{-1}$] and q_e is the amount of arsenic adsorbed at equilibrium [$\text{mg-As (g-adsorbent)}^{-1}$]. According to the stated reaction scheme, one adsorbate species reacts with one site to form reaction products. The fitting validity of this model can be checked by linear plot of $\ln(1 - q_t/q_e)$ against t .

The pseudo second order model, also known as Ritchie's model, has the following linearized form ((Onyango et al., 2003, Ho and Mckay, 2000, Cheung et al., 2001):

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e} t \quad (3)$$

where, q_t is the amount of arsenic adsorbed at any time [$\text{mg-As (g-adsorbent)}^{-1}$], q_e is the amount of arsenic adsorbed at equilibrium [$\text{mg-As (g-adsorbent)}^{-1}$] and k is the rate constant expressed in $\text{g-adsorbent (mg-As min)}^{-1}$. One advantage of this model is that there is no need to know the value of equilibrium uptake q_e beforehand. The batch kinetic data for the adsorbent phase was fitted to both models. Figure 4 shows the plots for Lagergren's first order rate model based on Equation 2. Plots for Ritchie's model based on Equation 3 are shown in Figure 5. All the plots obtained for Ritchie's model were linear over the whole range of experimental time while for Lagergren's model, deviation of the experimental data from the theoretical ones were observed. The comparison of experimental equilibrium sorption capacities (q_e) and the theoretical values were estimated from the both models and are presented in Table 1. Considering the pattern of the plots and comparing the q_e values qualitatively, it can be said that the pseudo-second order model better describes the behavior of arsenic adsorption by laterite.

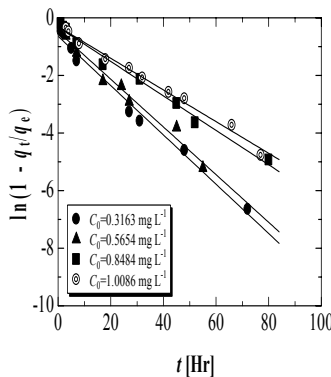


Figure 4

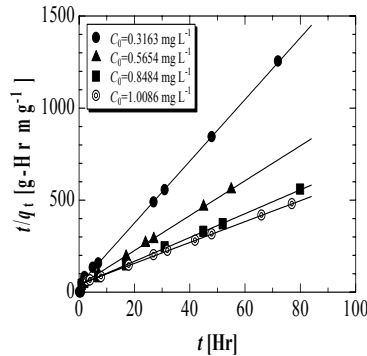


Figure 5

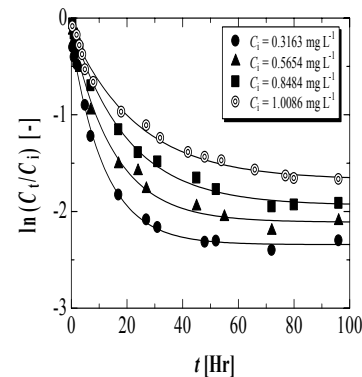


Figure 6

Table 1. Comparative data for equilibrium sorption capacities.

Initial As conc. [mg L^{-1}]	q_e (exp.) [mg g^{-1}]	q_e (theo., Lagergren's model) [mg g^{-1}]	q_e (theo., Ritchie's model) [mg g^{-1}]
0.3163	0.0575	0.0536	0.0596
0.5654	0.0990	0.0932	0.1054
0.8484	0.1444	0.1324	0.1545
1.0086	0.1621	0.1487	0.1762

The pseudo-first order rate constants, k_1 , determined from the plots of Figure 4 were 5.64×10^{-03} , 3.67×10^{-03} , 2.80×10^{-03} and $2.15 \times 10^{-03} \text{ min}^{-1}$ for initial concentrations 0.3163, 0.5654, 0.8484, 1.0086 mg L^{-1} , respectively. The rate constants were relatively lower than those obtained by Altundogan et al. (2002) in the study of arsenic uptake by activated red mud. The difference is

evident because Altundogan et al. (2002) operated the system at a dosage of 20 g L^{-1} , which was 4 times higher than the amount used in the present study.

The pseudo-second order rate constants calculated from Figure 5 were 0.0363, 0.0251, 0.0146 and $0.0071 \text{ g (mg-min)}^{-1}$ for initial concentrations 0.3163, 0.5654, 0.8484, 1.0086 mg L^{-1} respectively.

The data shows a decreasing trend with an increase in initial arsenic concentration. Observed trend is similar to that for arsenic uptake by iron-conditioned zeolite reported by Onyango et al. (2003) Ho and McKay (2000) also observed similar trend in the kinetics of sorption of divalent metal ions onto sphagnum moss peat that followed a pseudo-second order mechanism.

Role of Transport in Arsenic Uptake

An adsorption process may be considered to consist of transport of reactants from the bulk phase to the laminar liquid film at the surface, transport across the film and into the interior of the adsorption sites where chemical transformation (chemisorption) takes place. The adsorption mechanism can be controlled by reaction kinetics and/or transport processes such as film and pore diffusion. Figure 6 shows the plot of $\ln(C_t/C_i)$ versus time, obtained from kinetic experiments performed to study the effect of initial concentration on arsenic removal based on the following equation (Van Lier, 1989):

$$\ln \frac{C_t}{C_i} = -\frac{K_f W S_w}{V_f} t \quad (4)$$

where, K_f is the mass transfer coefficient for film diffusion, S_w is the specific external surface of adsorbent on weight basis, W is the adsorbent dosage, V_f is the volume of the fluid, C_i is the initial adsorbate concentration and C_t is the concentration of the adsorbate at any time. Figure 6 was further used to evaluate whether the film resistance to mass transfer based on Equation 4 controlled the arsenic uptake in the batch adsorption process. From Equation 4 a plot of $\ln(C_t/C_i)$ versus time should be linear if the adsorption mechanism is controlled by film diffusion (Van Lier, 1989). Apparently, all the curves in the Figure 6 are non-linear, suggesting that film diffusion did not control the arsenic uptake. Considering the high speed of agitation of 400 rpm used during the kinetic experiments, it is therefore not surprising that film resistance did not control the uptake of arsenic. At

this speed, the shear on the particle surface was considerably high. Therefore, the thickness of the boundary layer surrounding the particles was minimal and boundary layer resistance or film diffusion should not be a rate-limiting step. In the research by Onyango et al. (2003) on removal of arsenic from water using iron-conditioned zeolite and by Cheung et al. (2001) on removal of cadmium ions from effluents using bone char, the agitation speed was set at 200 rpm and 400 rpm respectively, at which speeds boundary layer surrounding the particles did not control the sorption mechanism, similar to the results of this study.

The existence of transport of adsorbent ions from the solution to adsorption sites i.e. intraparticle diffusion can be tested in terms of graphical relationship between amount of adsorbed arsenic and square root of time (Figure 7) as shown by Weber and Morris (1962):

$$q = k_{id} t^{0.5} \quad (5)$$

where, q is the amount of arsenic adsorbed (mg g^{-1}) at time t (Hr) and k_{id} is the intraparticle diffusion rate constant ($\text{mg g}^{-1} \text{Hr}^{0.5}$). All the plots have the same general features: a linear portion that attributed to the intraparticle diffusion effect and the plateau to the equilibrium. k_{id} values were obtained from the slope of the linear portion of the curve at each solute concentration. The values are 0.0143, 0.0289, 0.0376 and 0.0412 $\text{mg g}^{-1} \text{Hr}^{0.5}$ for initial concentrations 0.3163, 0.5654, 0.8484, 1.0086 mg L^{-1} , respectively. The values indicate that with an increase in the initial concentration arsenic uptake rate also increased. In Figure 7, the linear portions of the curves do not pass through the origin which indicates that mechanism of arsenic removal by laterite is complex, and that, though, intraparticle diffusion is the major rate determining step, the contribution due to the surface adsorption cannot be ignored (Singh and Pant, 2004, Namasivayam and Yamuna, 1995).

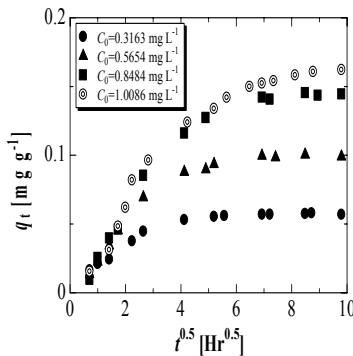


Figure 7

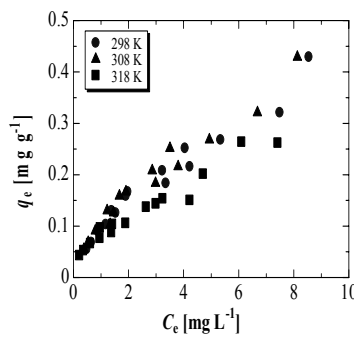


Fig 8(a)

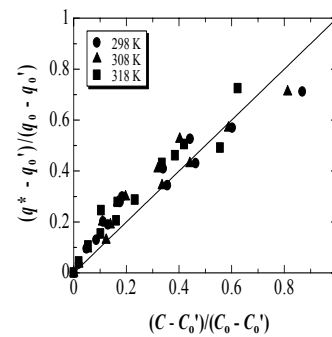


Figure 8(b)

Adsorption Isotherms

The general nature of the mass transfer zone for the laterite-arsenic interaction can be determined by the equilibrium isotherm. Depending on whether the equilibrium relation is linear, favorable, or unfavorable over the concentration range, three general cases can be distinguished. In the Figs. 8a and 8b, possible general forms along with a simple non-dimensional representation of the equilibrium relationship, expressed in terms of the reduced variables, are shown. The diagonal line shown in the non-dimensional representation of the equilibrium relationship is used to distinguish the nature. Data points above the diagonal line represent the favorable condition, and consequently the points below the diagonal line correspond with the unfavorable nature. For the laterite-arsenic interaction, within the typical arsenic concentration range, all the data points are in the favorable region i.e. laterite can be used to remove arsenic from groundwater.

The distribution of arsenic between the liquid phase and adsorbent (laterite) phase at equilibrium can also be described by two typical adsorption isotherms, Freundlich and Langmuir:

$$\ln q_e = \ln K + n \ln C_e \quad (6)$$

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m} \quad (7)$$

where, K and n are temperature-dependent constants corresponding to adsorption capacity and adsorption intensity, respectively. C_e is equilibrium concentration (mg L^{-1}), q_e is amount adsorbed at equilibrium (mg g^{-1}). q_m and b are Langmuir constants related to the adsorption capacity and energy of adsorption, respectively. The equilibrium data are fitted with both Freundlich and Langmuir isotherms and values of the adsorption constants K , n , q_m and b are calculated using non-linear optimization by Marquardt method as listed in Table 2.

Table 2. Adsorption isotherm constants and RMSD values.

Temperature [K]	Freundlich		Langmuir			
	K	n	$RMSD$	q_m	b	$RMSD$
	[mg g^{-1}]			[mg g^{-1}]	[L mg^{-1}]	
298	0.0949	0.6432	0.0721	0.5321	0.2211	0.0771
308	0.1017	0.6311	0.0740	0.4986	0.2742	0.0989
318	0.0847	0.505353	0.0804	0.2498	0.6824	0.1878

Langmuir isotherm indicates that the reaction is a reversible phenomenon (Gupta and Chen, 1978, Altundogan et al., 2002) and the coverage is monolayer (Gupta and Chen, 1978, Altundogan et al., 2002, Singh et al., 1988). As observed for the calculated values of Langmuir constants, q_m decreases with the increase of temperature but b shows increasing trend. Langmuir constant, q_m is measure of the maximum adsorption capacity of the adsorbent (Singh et al., 1988) and its decreasing trend with the increase of temperature indicates lowering of capacity as temperature rises. Langmuir constant, b is measure of the energy of adsorption (Singh et al., 1988) and its increase with temperature indicates endothermic nature of the process.

Experimental and theoretically predicted data for Freundlich and Langmuir isotherms at different temperatures were plotted for comparison (Figs. 9a, 9b, 9c) and root mean square deviation (RMSD) were calculated. The RMSD data (Table 2) shows that Freundlich isotherm can better correspond with the experimental data.

Thermodynamic Parameters

Thermodynamic parameters: standard Gibbs free energy (ΔG°), enthalpy change (ΔH°) and entropy

change (ΔS°) were estimated for arsenic adsorption by laterite using the following equations, respectively (Altundogan et al., 2002):

$$\ln \frac{1}{b} = \frac{\Delta G^\circ}{RT} \quad (8)$$

$$\ln b = \ln b_0 - \frac{\Delta H^\circ}{RT} \quad (9)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (10)$$

where b is Langmuir constant which is related to the energy of adsorption, b_0 is a constant, R is the Universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$) and T is temperature (K). Calculated values of thermodynamic parameters are given in Table 3. For the present system, ΔH° has positive value and it indicates endothermic nature of adsorption. ΔH° value also supports the above mentioned nature of reaction based on the tendency of Langmuir constant, b with temperature (Table 2). Hence, it can be concluded that the nature of arsenic adsorption on laterite is chemical type (Altundogan et al., 2002). Change in Gibbs free energy is defined as the driving force for a system to reach a chemical equilibrium. The decrease in free energy (ΔG°) with the rise in temperature shows an

increase in feasibility of adsorption with increasing temperatures (Panday et al., 1985). ΔS° is the driving force for physical and chemical changes in reactions. For the adsorption of arsenic on laterite, positive values of ΔS° were obtained which indicates increased randomness at the solid-

solution interface. It also indicates the affinity of the adsorbent for arsenic (Kuriakose et al., 2004). Some structural changes in adsorbate and adsorbent can also be assumed from the trends of ΔS° (Altundogan et al., 2002) but the characteristics of the change are yet to be explored.

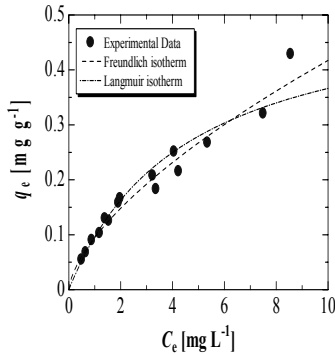


Figure 9(a)

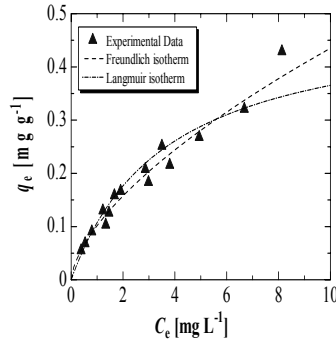


Figure 9(b)

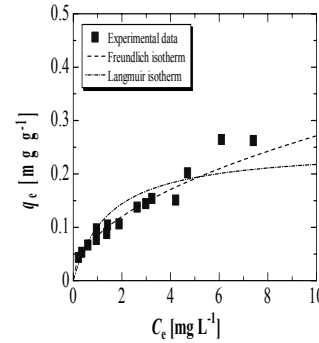


Figure 9(c)

FIGURE CAPTIONS

Figure 1. Time course for residual arsenic concentrations at different initial dose.

Figure 2. Percent arsenic removal with time at different initial concentrations.

Figure 3. Time course for q_t (change is sorption capacity) of laterite.

Figure 4. Lagergren's first order kinetic plots.

Figure 5. Kinetic plots for Ritchie's pseudo-second order model.

Figure 6. Test for film diffusion as a rate controlling step in arsenic uptake.

Figure 7. Intraparticle mass transfer curve for adsorption of arsenic on Laterite.

Figure 8. Equilibrium isotherms. (a) General form (b) Non-dimensional representation.

Figure 9. Experimental and theoretically predicted value comparison for Freundlich and Langmuir isotherms at different temperatures: (a) 298 K (b) 308 K (c) 318 K.

Table 3. Thermodynamic parameters.

Temperature	ΔH°	ΔS°	ΔG°
[K]	[kJ mol ⁻¹]	[kJ mol ⁻¹ K ⁻¹]	[kJ mol ⁻¹]
298		0.0799	1.8832
308	25.68	0.0779	1.6688
318		0.0791	0.5089

Adsorber Life-Time Calculation

A laterite-loaded packed-bed type adsorber ($L=50$ cm, $D=10$ cm), to meet the arsenic-free water requirement of a 4-member family (10 L day⁻¹) having contaminated water supply with 0.3163 mg L⁻¹ arsenic in water, found to last for maximum 74 days while to achieve WHO safe limit (0.01 mg L⁻¹), as calculated with simplest assumptions. Column experiments to explore the most-suitable design of the removal module are yet to be conducted, though.

Conclusion: Arsenic removal by heat treated laterite was experimentally investigated and it was explored that laterite can be effectively used for the removal of arsenic from contaminated groundwater. The removal rate was found dependent on the initial arsenic concentrations and adsorption

equilibrium data indicates favorable adsorption of arsenic onto laterite. It can also be concluded that change in natural weather temperature may not have any significant effect on the arsenic adsorption efficiency of the laterite. Laterite is a natural-substance, so, use of laterite for removal of arsenic from contaminated water is expected to be economical and feasible. To explain the behavior and action of laterite, in addition to the present work, further study is suggested e.g. effect of coexisting ions, chemical reaction involved with arsenic adsorption on laterite etc. Destination of spent laterite and methodology of spent laterite treatment to produce safe-sludge are some other points to consider.

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