Removal of arsenic from synthetic groundwater by adsorption using the combination of laterite and iron-modified activated carbon

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ABSTRACT

Groundwater is one of the most important sources for drinking water. Arsenic (As) contamination of groundwater is a serious problem worldwide, especially in Bangladesh, India and South East Asia. Adsorption using appropriate and readily available adsorbents is a promising method for the removal of arsenic with applicability in rural areas. This work aims to study the removal of arsenic from synthetic groundwater using an adsorption column by sequential combination of laterite (LA) and iron-modified activated carbon (AC-Fe) as adsorbents. The effect of ratio LA/AC-Fe, flow rate, initial arsenic concentration and pH to the breakthrough time were investigated. Adsorption equilibrium and adsorption kinetics were also studied through batch experiment. The result was found to be an efficient and feasible approach for arsenic treatment from groundwater for ready applicability in rural areas.

Keywords: adsorption, arsenic, breakthrough, iron-modified activated carbon, isotherm, laterite.

INTRODUCTION

Arsenic is known as one of the most toxic chemical elements. Long-term exposure of which can cause cancer of the skin, lungs and many others. In view of this, the World Health Organization (WHO) has set the standard for arsenic in drinking water as 0.01mg/L (WHO, 1993). Groundwater is an important source for drinking water. Recently, arsenic (As) contamination of groundwater has become a major concern on a global scale, especially in Bangladesh, India and South East Asia (Naidu et al., 2006).

Arsenic in groundwater usually exists as oxy-anion compound namely arsenite (trivalent arsenic, As[III]) and arsenate (pentavalent arsenic, As[V]) where As[III] is known as the predominant species which is more toxic and more difficult to remove than As[V] (Amin et al., 2006).

There are several techniques for the removal of arsenic from groundwater including physicochemical and biological treatments and membrane filtration. However, almost all of these techniques are either difficult to meet WHO's standard for drinking water or sophisticated and expensive which cannot be considered in application for rural areas, especially in developing and poor countries. A promising method that can be considered efficient and low cost for wide-scale application in rural areas is adsorption using appropriate, readily available and cheap adsorbents.

Laterite (LA) is a red-colored clay existing abundantly in tropical regions. It contains mainly compounds of iron, aluminum, and silica with positive surface charge under normal conditions. Therefore, LA can adsorb anionic contaminants as As[III] and As[V] (Larsson et al., 1999). A number of researchers (Larsson et al., 1999; Rupa et al., 2003; Abhijit et al., 2007; Partey et al., 2005) have studied LA and indicated that more than 90% of As can be removed at neutral pH and Langmuir isotherm can fit with equilibrium data. Sanjoy et al., (2007) investigated the performance of a fixed-bed column for the removal of As[III] using laterite soil and showed that a column with 2cm diameter and different bed heights such as 10cm, 20cm and 30cm, could produce 1.3, 7.9 and 14.88 liters of arsenic-free water, respectively, with As[III] initial concentration of 0.5mg/L with a flow rate of 7.75mL/min.

Activated carbon (AC) is always considered as a typical adsorbent, but AC has low arsenic removal capacity. The percentage removal of As[III] by AC is just 50% (Prasenjit et al., 2007). However, it can be improved by its surface modification with metals such as iron, manganese, aluminum, calcium, titanium or copper. Maximum removal of 98% for both As[V] and As[III] can be achieved in the pH ranges of 5–7 and 9–11, respectively (Prasenjit et al., 2007; Gu et al., 2005; Gu et al., 2007). Weifang et al. (2007) indicated that preloading AC with iron greatly improved its arsenic adsorption capacity. When comparing rapid small-scale column tests (RSSCT) results, iron-preloaded carbon could get over 200 times longer bed life than non-modified carbon.

Although iron-modified activated carbon (AC-Fe) is known as a good adsorbent, it is still expensive, whereas LA has been considered a cheap adsorbent. Therefore, the sequential combination of these two adsorbents in one column, where LA is as a rough adsorbent for cost reduction and AC-Fe is as an advanced adsorbent for further removal, is an efficient and feasible way to remove arsenic from groundwater for ready applicability in rural areas, especially in developing and poor countries.

This work aims to study the adsorption column for removal of arsenic from synthetic groundwater using sequential combination of LA and AC-Fe as adsorbents. The effect of LA/AC-Fe ratio, flow rate, initial arsenic concentration and pH to the breakthrough time were investigated. Adsorption equilibrium and adsorption kinetics were also studied through batch experiment.

MATERIALS AND METHODS
Synthetic groundwater
Typical groundwater with average concentration of the major components was simulated and used in this study. The major components of the synthetic groundwater are shown in *Table 1* as referred from previous work (Lien et al., 2005).

Arsenic stock solution
The stock solutions of As(III) were prepared from As(III) standard solution of 1003mg/L (Wako Pure Chemicals Ltd, Japan). The stock solutions of As(V) were prepared by dissolving 4.1646mg of $Na_2HAsO_4.7H_2O$ (Wako Pure Chemicals Ltd, Japan) into distilled water with a total volume of 1000mL. This stock solution has an arsenic concentration of 1000mg/L.

Table 1. Composition of synthetic groundwater

Composition	Concentration, mg/L
$CaCl_2 \cdot 2H_2O$[*]	230
Na_2SO_4[*]	1200
$NaHCO_3$[*]	370
$MgCl_2 \cdot 6H_2O$[*]	135

(*Wako Pure Chemicals Ltd, Japan)

Preparation of Adsorbents
Preparation of LA
In this study, laterite was collected from Ogasawa Island (Tokyo, Japan). It was sieved to a particle size of 0.45-0.71mm. Then, LA was heated at 150°C for two hours under anaerobic condition without washing, acidic or alkali treatment. The BET (Brunauer, Emmett and Teller) surface area is 70.19m²/g, pore volume of 0.00062cm³/g and pore size of 126Å.

Preparation of AC-Fe
AC from coconut shell was taken from MAPECOH, Inc (Philippines) with a particle size of 0.45-0.71mm after sieving. Then, it was washed with distilled water, dried at 105°C for 24 hours before being modified with iron. The preparation of iron-modified activated carbon (AC-Fe) was conducted by impregnation with 5% of iron onto AC (theoretical calculation based on weight). The impregnation method for preparation of AC-Fe was similar to that of Prasenjit et al. (2007). The BET (Brunauer, Emmett and Teller) surface area was 508m²/g, pore volume was 0.2426 cm³/g, and pore size was 43.23Å.

Figure 1a and *1b* showed SEM (Scan Electron Microscopy) photographs of original AC (non-modified) and iron-modified AC, respectively. It can be seen that while the surface of the original AC seems to be clean (*Figure 1a* on left side), there are some patches of iron particles stuck on the surface of AC-Fe *(Figure 1b* on right side*).* These may be the active sites for arsenic adsorption.
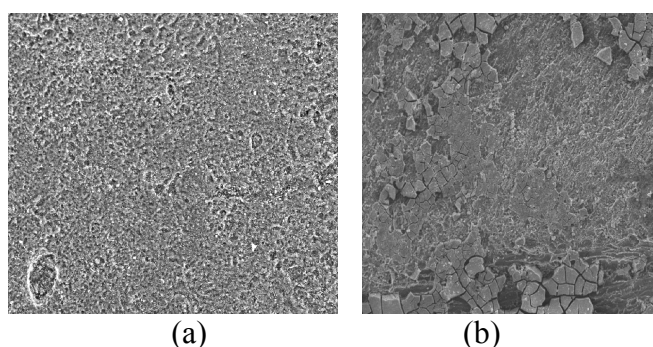

(a)   (b)

Fig.1   SEM (200x magnification) for original AC and iron-modified AC (with 5% of iron onto AC)

Experimental procedure
Batch experiment
In this study, batch experiments for equilibrium and kinetic studies were carried out in a series of 1000mL flasks. Each of them contained 5g of adsorbents (adsorbent dose) and

1000mL synthetic groundwater with various levels of total arsenic content (initial arsenic concentration) at pH of synthetic groundwater (pH 8.18). The sample flasks were stirred at 300rpm by speed-controlled stirrers in a temperature-controlled bath for 168h for LA and 120h for AC-Fe. These were performed under room temperature (25°C) and atmospheric condition. The samples were taken then acidified and analyzed for residual arsenic concentration. A mixture of 70% As[III] and 30% As[V] was used in all of the experiments and the total arsenic content was analyzed by ICP-MS (Seiko SII).

Column experiment
Column experiments for the determination of breakthrough time were carried out in a column of 0.025m diameter and 0.5m length. The inlet flow rate was controlled by adjustable flow meters in up-flow mode operation. The column was divided into 2 sections: one for LA and another for AC-Fe. The parked-bed densities are 0.55m^3/m^3 for LA section and 0.54m^3/m^3 for AC-Fe section.

The LA/AC-Fe ratio was investigated first to determine an optimum ratio, and this optimum ratio will be fixed to investigate the effect of other parameters such as pH, initial arsenic concentration and inlet flow rate. The samples taken were acidified and analyzed for residual arsenic concentration. A mixture of 70% As[III] and 30% As[V] was used in all experiments and the total arsenic content was analyzed by ICP-MS (Seiko SII). Experimental set-up is shown in *Figure 2*.
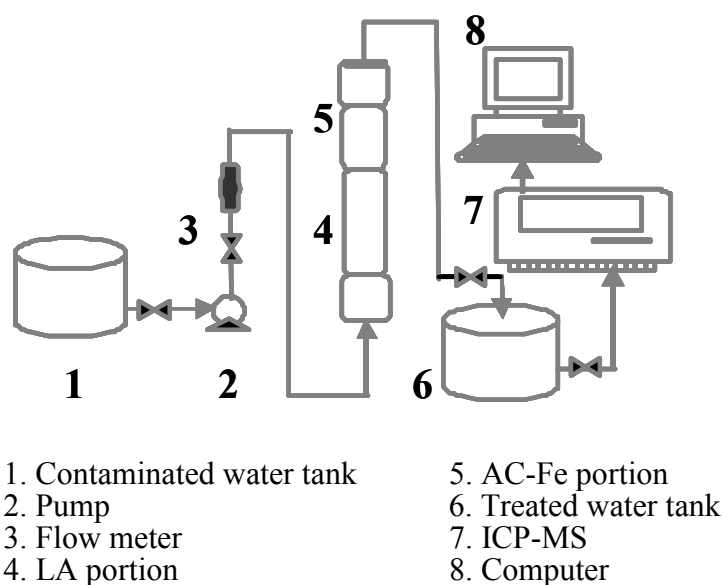

1. Contaminated water tank   5. AC-Fe portion
2. Pump   6. Treated water tank
3. Flow meter   7. ICP-MS
4. LA portion   8. Computer

Figure 2 Experimental set-up

RESULTS AND DISCUSSION
Adsorption isotherm
Assuming that the adsorption/desorption process occurs in a closed batch system with complete mixing and no mass transfer resistance, the equation is as follows:

$$nA + S \leftrightarrow A_n S \tag{1}$$

This equation is written based on the adsorption site *S*, where *A* is arsenic molecule and *n* is the number of arsenic molecule (*n*: greater than zero). The overall adsorption/desorption rate is:

$$\frac{d\theta}{dt} = k_a C_A^n (1-\theta) - k_d \tag{2}$$

in terms of adsorbate concentration *q* is:

$$\frac{dq}{dt} = k_a C_A^n (q_0 - q) - k_d q \tag{3}$$

where θ is surface coverage, k_a is the adsorption rate constant, *L/mgs*; k_d is the desorption rate constant, 1/s; *q* is adsorbate concentration *mg/g*, q_0 is arsenic adsorption capacity of adsorbent, (*mg/g*); and C_A is concentration of arsenic molecule in the solution, *mg/L*. At equilibrium, from equation (3) *dq/dt = 0*; $q = q_e$ and $C_A = C_{A,e}$, and by derivation:

$$q_e = \frac{q_0 K C_{A,e}^n}{1 + K C_{A,e}^n} \tag{4}$$

where q_e is adsorbate concentration at equilibrium (*mg/g*); $C_{A,e}$ is equilibrium concentration of arsenic in solution (*mg/L*); and $K = k_a / k_d$ is isotherm constant, (*L/mg*). Equation (4) is well known as Langmuir-Freundlich isotherm. To estimate the isotherm coefficients *n*, *K* and q_0, non-linear regression method must be employed. Several computer programs are available in dealing with nonlinear regression. In this study, Microsoft Excel was used. The isotherm coefficients for both AC-Fe and LA are shown in *Table 2*.

Results of the study showed that *n* is approximately one unit (*Table 2*). Hence, *n=1* is assumed. This means that one site of LA or AC-Fe adsorbent may adsorb one arsenic molecule (Langmuir isotherm). Using non-linear regression method to examine Langmuir isotherm, the value is shown in *Table 3*. It can be seen that Langmuir isotherm also fits with equilibrium data for both LA and AC-Fe as shown in the values of the regression coefficient r^2. Therefore, adsorption process of arsenic onto LA and AC-Fe may be described by Langmuir isotherm and the adsorption capacities were 0.48mg/g and 1.18mg/g, respectively, as shown in the value of q_0 of Langmuir isotherm. *Figure 3* and *Figure 4* are non-linear plots of the most common used isotherms: Langmuir, Langmuir-Freundlich and Freundlich for AC-Fe and LA. Apparently, Langmuir-Freundlich and Langmuir isotherms fit better with experimental equilibrium data than Freundlich isotherm.

Table 2. Values of Langmuir-Freundlich isotherm coefficients

Adsorbents	q_0, mg/g	K, L/mg	n	r^2
AC-Fe	1.28	1.82	0.86	0.991
LA	0.45	0.82	1.08	0.968

Table 3. Values of Langmuir isotherm coefficients

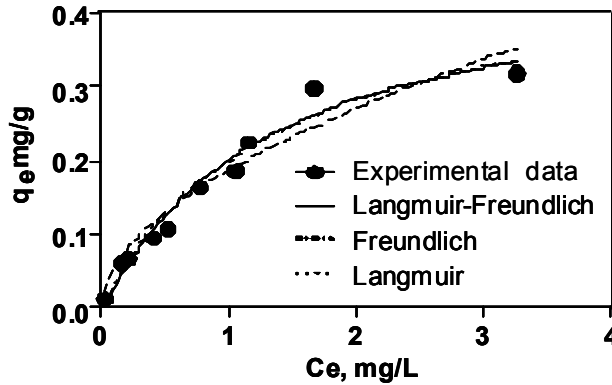| Adsorbents	q_0, mg/g	K, L/mg	r^2
AC-Fe	1.18	2.47	0.988
LA	0.48	0.72	0.968

Figure 3. Non-linear plot for adsorption isotherms for LA, particle size 0.45-0.71mm, $C_{A,0}$ 0.09 - 4.84mg/L, dose 5g/L, pH of synthetic groundwater (pH8.18).
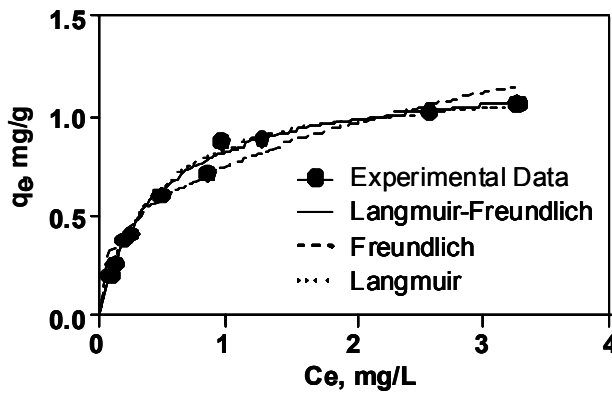

Figure 4. Non-linear plot for adsorption isotherms for AC-Fe, particle size 0.45-0.71mm, $C_{A,0}$ 1.11 - 8.60mg/L, dose 5g/L, pH of synthetic groundwater (pH8.18).

Adsorption kinetics

Two most popular adsorption kinetic models, pseudo-first order (Lagergren model) and pseudo-second order, have been used by some researchers (Ho et al., 2000) to describe the process kinetics of adsorption. From equation (1), the use of $n=1$ from calculation and the assumption in section 3.1 (adsorption equilibrium), gives the overall rate of adsorption/de-sorption as:

$$\frac{d\theta}{dt} = k_a C_A (1-\theta) - k_d \theta \tag{5}$$

From equation (5), an integrated form of the well-known pseudo-second order kinetics was derived by Saeid (2004), as follows:

$$\frac{t}{q} = \frac{1}{k_2 q_e^{\,2}} + \frac{1}{q_e} t \qquad\qquad (6)$$

where q is the amount of arsenic adsorbed (adsorbate concentration) at time t, mg/g; q_e is the amount of arsenic adsorbed at equilibrium mg/g; and k_2 represents the observed adsorption rate coefficients. A plot between t/q versus t gives the value of the constant k_2 and q_e (mg/g) can also be calculated (*Figures 5* and *Figure 6*).
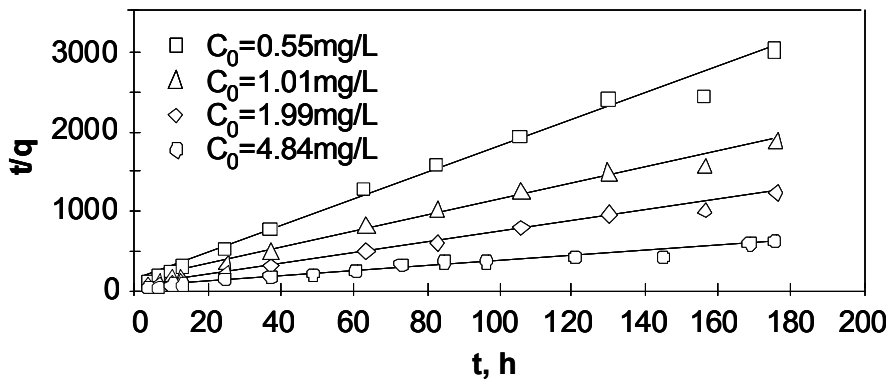
Figure 5. Linear plot of pseudo-second order kinetics for LA, particle size 0.45-0.71mm, $C_{A,0}$ 0.55 - 4.84mg/L, dose 5g/L, pH of synthetic groundwater (pH8.18).
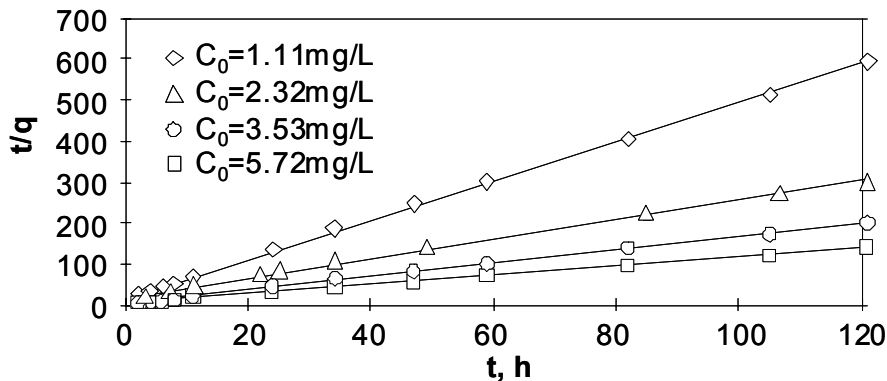

Figure 6. Linear plot of pseudo-second order kinetics for AC-Fe, particle size 0.45-0.71mm, $C_{A,0}$ 1.11 - 5.72mg/L, dose 5g/L, pH of synthetic groundwater (pH8.18).

The values of the observed rate coefficients k_2 and the corresponding linear regression correlation coefficient r^2 were calculated and shown in *Table 4*. It can be seen that pseudo-second order kinetics fits with the experimental data as shown in the value of r^2 for both LA and AC-Fe. In other words, this shows the applicability of the pseudo-second order kinetic model in predicting arsenic adsorption process onto LA and AC-Fe. Indeed, Saeid (2004) also indicated that the sorption process obeys pseudo-second order kinetics model at lower initial concentration of solute. In addition, the present pseudo-second order kinetic expression was tested for predicting the amount of arsenic adsorbed for the overall adsorption time. The q_e was predicted by applying the

calculated kinetic coefficients to their corresponding kinetic expressions. These predicted values of q_e can be compared with q_e from the experimental data. It means that the experimental data fit onto the model *(Table 5)*.

Table 4. Rate coefficients of pseudo-second order kinetics

Adsorbents	$C_{A,0}$ (*mg/L*)	k_2 (*g/mg.s*)	r^2
AC-Fe	1.11	1.69	0.999
	2.32	0.72	0.998
	3.53	0.35	0.999
	5.72	0.30	0.999
LA	0.55	2.23	0.994
	1.01	0.93	0.987
	1.99	0.78	0.993
	4.84	0.23	0.970

Table 5. Comparison of q_e for model and experiment

Adsorbents	$C_{A,0}$ (*mg/L*)	q_e, (*mg/g*) Experiment	q_e, (*mg/g*) Model
AC-Fe	1.11	0.21	0.21
	2.32	0.42	0.44
	3.53	0.61	0.62
	5.72	0.89	0.89
LA	0.55	0.06	0.06
	1.01	0.10	0.10
	1.99	0.15	0.15
	4.84	0.32	0.30

Adsorption column
The appropriate LA/AC-Fe ratio
It can be understood that the longer portion of AC-Fe in the column, the longer the breakthrough time, because according to the equilibrium and kinetics in this study, AC-Fe is a much more effective adsorbent than LA (Langmuir adsorption capacity for LA and AC-Fe are 0.48mg/g and 1.18mg/g, respectively). However, it is also possible to realize that the longer the portion of AC-Fe, the higher the adsorbent cost. Therefore, optimizing the AC-Fe portion in the column is one of the key factors for a feasible arsenic treatment system.

To determine an appropriate LA/AC-Fe ratio, the value of V/W_{ACFe} (V, the volume of drinkable water produced from the start using adsorbents until breakthrough / W_{ACFe}, the weight of AC-Fe in the column) was calculated and found that the appropriate ratio of LA/AC-Fe is 0.3m/0.2m. This is because at this ratio, the volume of arsenic-free water per gram of AC-Fe is highest (0.8L/g-AC-Fe) compared with other tested LA/AC-Fe ratios of 0.4m/0.1m, 0.25m/0.25m, 0.2m/0.3m (with corresponding V/W_{AC-Fe} values of 0.71, 0.78 and 0.76L/g-AC-Fe, respectively) at the same experimental conditions *(Table 6)*. *Figure 7* shows the breakthrough curves for different ratios of LA and AC-Fe portion. *Figure 8* shows the breakthrough curve for three single adsorbents: AC, LA, and AC-Fe and *Figure 9* illustrates the comparison of LA-AC and LA-AC-Fe. These figures indicated that the combination of LA and AC-Fe is the most feasible.

Table 6. Appropriate LA/AC-Fe ratio

LA/AC-Fe, m/m	V/W_{AC-Fe} , L/g
0.4/0.1	0.71
0.3/0.2	**0.80**
0.25/0.25	0.78
0.2/0.3	0.76

Experimental conditions: L=0.5m, D=0.025m, pH of synthetic groundwater (pH 8.18), Co=0.61mg/L (70%AsIII&30%AsV), F=6ml/min.
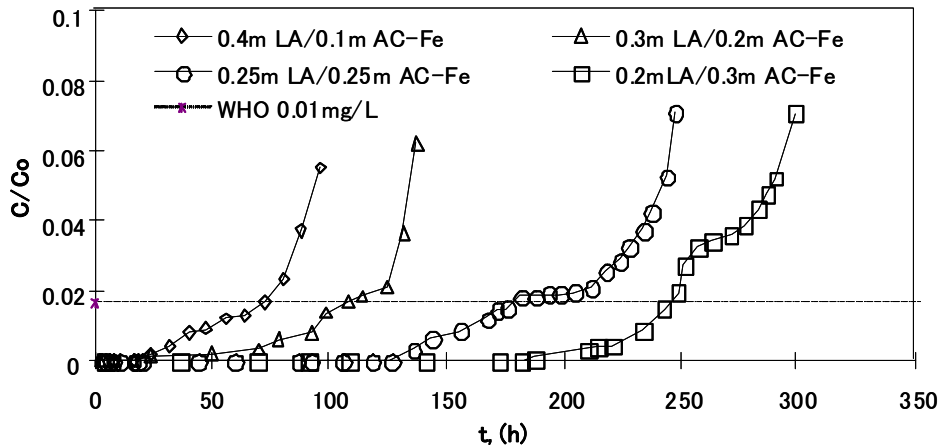

Figure 7. Breakthrough curve for different ratios of LA and AC-Fe in the column: L=0.5m, D=0.025m, pH of synthetic groundwater (pH 8.18), Co=0.61mg/L (70%AsIII&30%AsV), F=6ml/min.
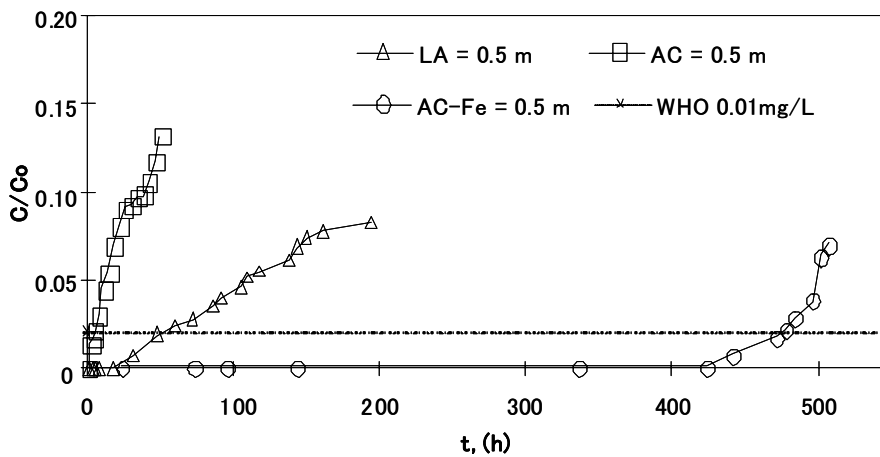

Figure 8. Breakthrough curve for comparison of single adsorbents: AC, LA and AC-Fe in the column: L=0.5m, D=0.025m, Co=0.47mg/L (70%AsIII&30%AsV), F=5mL/min, pH of synthetic groundwater (pH 8.18).
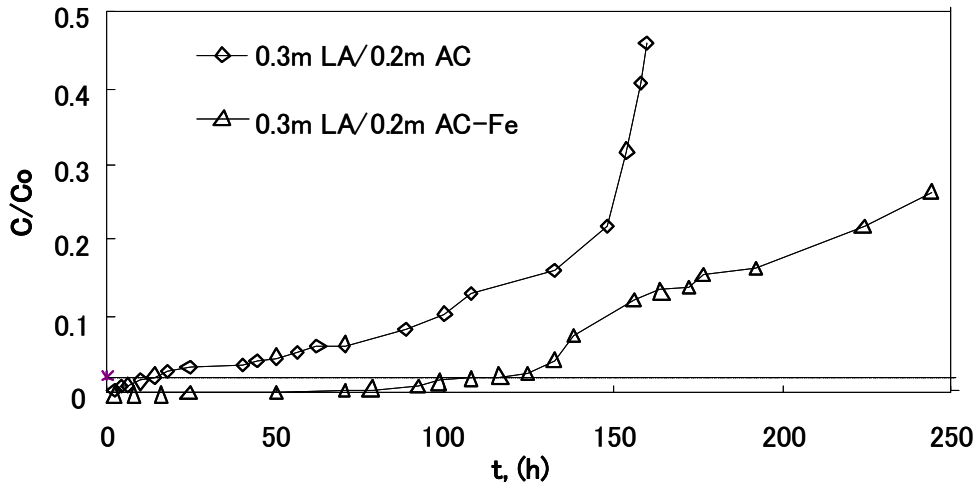
Figure 9. Breakthrough curve for the comparison of LA/AC and LA/AC-Fe combination in the column: L=0.5m, D=0.025m, Co=0.46mg/L (70%AsIII&30%AsV), F=5 ml/min, pH of synthetic groundwater (pH 8.18).

Effect of inlet flow rate

It was observed that flow rate is one of the most important parameters to look at in column study for arsenic removal by sequential combination of LA and AC-Fe. In *Figure 10,* at flow rate of 5mL/min or space velocity of 1.22m3/m3 h, the breakthrough time is very long (116h). However, at higher flow rates of 15 and 30mL/min (or space velocity of $3.67m^3/m^3$ h and $7.34m^3/m^3$ h, respectively), the breakthrough time occurred immediately. In other words, arsenic concentration of the treated water is immediately much higher than 0.01mg/L (WHO standard for drinking water) in the first hours. This is because the residence time of solute (arsenic anions) in the bed is decreased resulting to shorter contact between the feed and the adsorbent. In addition, the higher the flow rate, the smaller the film resistance, implying that arsenic adsorption by LA or/and AC-Fe may be controlled by intra-particle diffusion.
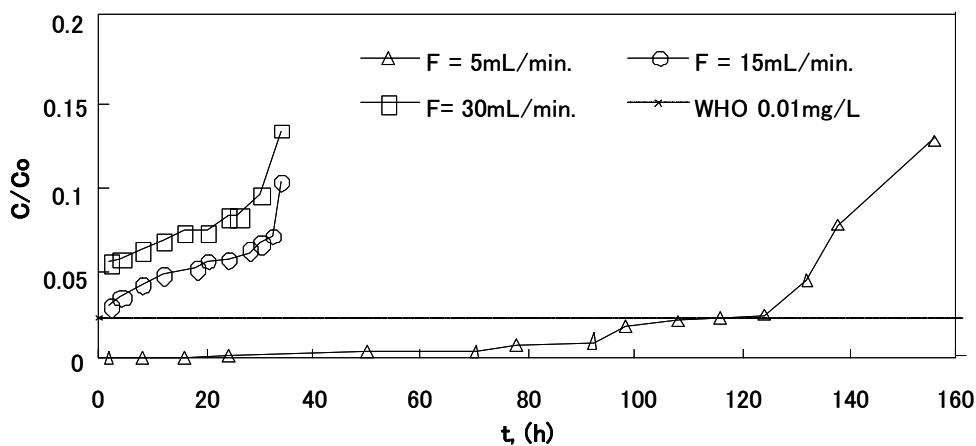

Figure 10. Effect of flow rate to the breakthrough time in the column: D=0.025m, L=0.5m with 0.3mLA/0.2AC-Fe, pH of synthetic groundwater (pH8.18), Co=0.45mg/L (70%AsIII&30%AsV).

Effect of pH

The effect of pH to breakthrough time is quite complicated, because of two different adsorbents in the column. Breakthrough times are 21, 92 and 68h for pH of 10.0, 8.18 (no adjustment, pH of synthetic groundwater) and 5.0, respectively (*Figure 11*). Although breakthrough time corresponding with WHO standard for drinking water is just 68h at pH5, the breakthrough time corresponding with the arsenic concentration of the treated water of 0.011-0.032mg/L (just a little higher than 0.01mg/L) is longer (112h) than that at pH8.18. In other words, a little decrease of the initial arsenic concentration or flow rate may strongly increase the breakthrough time at pH 5.
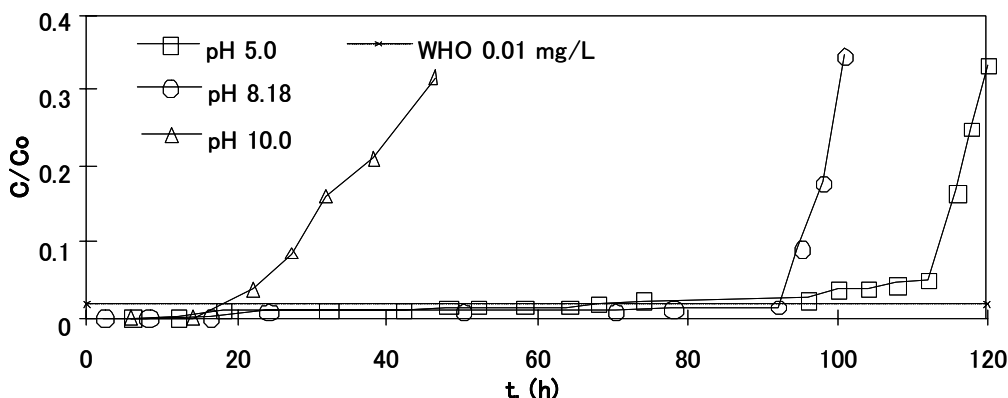

Figure 11. Effect of pH to the breakthrough time in the column: D=0.025m, L=0.5m with 0.3mLA/0.2mAC-Fe, F=5mL/min, Co=0.6mg/L (70%AsIII&30%AsV).

CONCLUSION

The removal of arsenic from synthetic groundwater by adsorption using LA and AC-Fe was investigated in this study. Langmuir, Langmuir-Freundlich isotherms and pseudo-second order model described well the equilibrium and kinetics. The Langmuir adsorption capacities are 0.48mg/g and 1.18mg/g, respectively.

A sequential combination of LA and AC-Fe in a column of 0.025m diameter and 0.5m length for arsenic from synthetic groundwater was studied. The appropriate parameters are: LA/AC-Fe ratio of 0.3m/0.2m, flow rate of 4 to 6mL/min (or space velocity of 0.98-1.47m^3/m^{3}h), pH of 7-8, and initial arsenic concentration of 0.1-0.5mg/L, to obtain a breakthrough time of more than 100 hours. The operation of the column is very sensitive with flow rate, implying that the adsorption of arsenic may be controlled by intra-particle diffusion.

In conclusion, while AC-Fe can be an effective adsorbent for the arsenic removal at high initial concentration of arsenic (1.11 - 5.72mg/L), low adsorbent dose (2-5g/L) and around pH of groundwater (pH8-9), the combination of LA and AC-Fe in a column, could effectively remove arsenic from groundwater. LA which is a rough adsorbent could be used for reducing cost and AC-Fe being an advanced adsorbent could be used for further removal. This approach may be an efficient and feasible way to remove arsenic from groundwater for ready application in the rural areas, especially in developing and poor countries.

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