

## Removal of Arsenic from Contaminated Water by Selected Geological Natural Materials

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**Abstract:** This study investigates the efficiency of peat soil, bauxite and iron concretion to remove arsenic from contaminated water. Batch experiments were carried out to study the removal kinetics of arsenic under different pH values and in the presence of low and high concentrations of arsenic. Mini column test was carried out to investigate the removal of arsenic from contaminated water under different pore volumes. The results showed that peat soil has high specific surface area (SSA) 422 - 533 m<sup>2</sup>/g compared with bauxite and iron concretion. However, iron concretion has high cation exchange capacity (CEC) 59-65 meq/100g while, peat soil and bauxite have 33-48 meq/100g and 24-33 meq/100g respectively. Batch test results present that the iron concretion is the best materials to remove arsenic from contaminated water with adsorbed amount (q) 4999 mg/kg after 48 hours shaking time. With the same shaking time, it appeared that the maximum amount of arsenic adsorbed by peat soil and bauxite are 4996 mg/kg and 4559 mg/kg, respectively. However, the mini column test results showed that iron concretion has low adsorption arsenic removal compared with bauxite and peat soil. The results showed that bauxite have maximum adsorption values with a relative concentration ( $C_e/C_o$ ) of  $1.4 \times 10^{-2}$  after 3 PV of infiltration. Iron concretion material had minimum adsorption with low  $C_e/C_o$  value of 0.99. In general, efficient removal of arsenic was observed resulting at concentrations below the limit of 10 µg/L in water.

**Key words:** Arsenic, batch test, mini column test, remediation

### INTRODUCTION

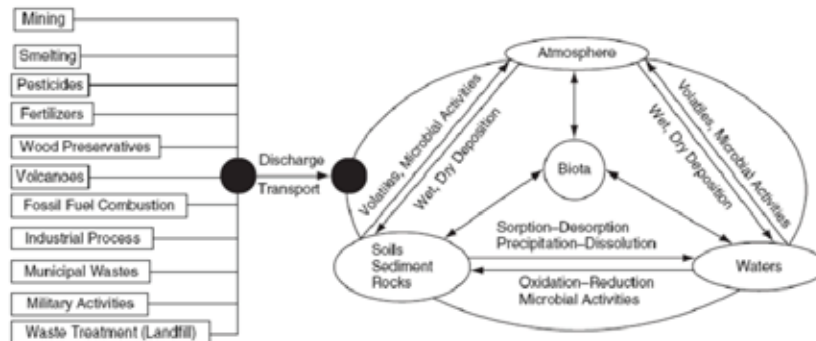
Arsenic is released into the environment from two principal pathways; natural processes and industrial activities through natural processes such as weathering of rocks and sediments, hydrothermal ore deposits, volcanic eruptions, geothermal activities, forest fire, wind-blown dust, and seasalt spray. Besides, it can be transported over long distance as suspended particulates or gaseous forms through water or air (Nriagu *et al.* 2007). Its flow in different compartments of the ecosystem is as shown in Figure 1.

The detrimental health effects of arsenic prompted the World Health Organization (WHO) and the United States Environmental Protection Agency (EPA) to reduce the drinking water arsenic standard from 0.05 to 0.01 mg/L (WHO 1993). Effectively, removing arsenic from waters with concentrations >0.01 mg/L is costly, commonly requiring expensive man-made arsenic sorbents. Consequently, there is a real need in the developing countries for low-cost materials and methods to remove arsenic from drinking water. One of the promising methods appears to be the sorption of inorganic arsenic compounds from solution using natural sorbents. Low-cost natural materials, e.g. activated red mud (Altundogan *et al.* 2002) and natural zeolites (Elizalde-González *et al.* 2001), were tested as potential sorbents.

Sorption is one of the most important factors that affect the fate of pesticides in the soil and consequently determine their distribution in the soil/water environment. Sorption is used to describe the process of pesticide partitioning between the water solution and soil. The processes of pesticide movement such as volatilization to the atmosphere, leaching into ground water and runoff to the surface water are explained by the sorption coefficient (Farahani *et al.* 2007).

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**Fig. 1:** Sources and distribution pathways for arsenic in the environment (reproduced from (Wang & Mulligan 2006).

Peat soils have often been used to trace atmospheric arsenic pollution but relatively rarely been investigated with respect to arsenic biogeochemistry, although it has become evident that organic-rich soils are often highly enriched with arsenic and that pore water concentrations in these systems can be very high (Gonzalez *et al.* 2006). In particular, little is yet known about the mechanisms causing a phase transfer of arsenic from dissolved to solid state in organic-rich soils and the geochemical conditions and time scale involved. In less organic-rich aquifers, arsenic dynamics have been linked primarily to the redox processes of iron and sulfur (Blodau *et al.* 2008). Of importance for the distribution of arsenic between dissolved and solid phase associated state are further the competition of arsenic with phosphate and dissolved organic matter (DOM) for sorption sites (Bauer & Blodau 2006). Heavy rainfall leaches out soluble weathering products in lateritic soils leaving behind the clay minerals like kaolinite ( $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ), rutile ( $\text{TiO}_2$ ), gibbsite ( $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$ ), goethite ( $\text{HFeO}_2$ ), lepidocrosite ( $\text{FeOOH}$ ), and hematite ( $\text{Fe}_2\text{O}_3$ ). Commonly, iron mineral concretions incorporate other soil constituents and grow in size until they occupy most of the soil horizon forming hardpan or ferricrete. In the extreme, ferricrete (called canga in South America) can be up to 5 m thick. Most commonly, lateritic soils have iron concretions that vary from pebble-size to cobble-size (Partey *et al.* 2008).

## MATERIALS AND METHODS

Peat soil, bauxite mineral and iron concretion were used in this study as a removal material for arsenic contamination. The samples were collected from Johor baru south part of Malay Peninsula and Bandar Baru Bangi. Materials sample was subjected to two main tests; i.e. (i) physical-chemical tests; and (ii) sorption tests via batch equilibrium test and mini column infiltration test. The physical tests were compaction and water content. While the chemical tests were cation exchange capacity (CEC) which uses ammonium acetate, specific surface area (SSA) which uses EGME method, and pH measurement.

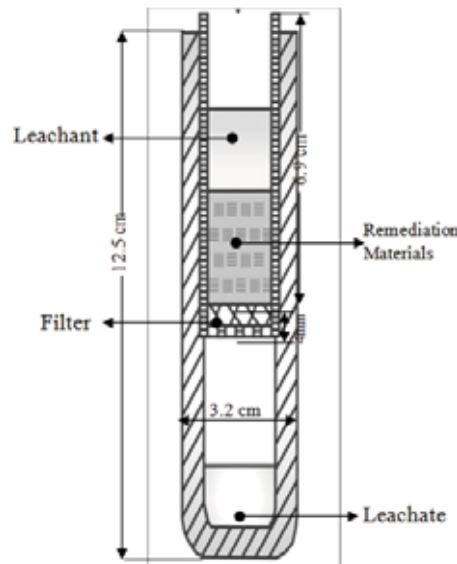
The batch sorption experiment was used to study the effect of three factors initial concentrations, shaking time and different pH of solutions. The same procedure was also reported by Wan Zuhairi (2003a, 2003b) and Wan Zuhairi *et al.* (2004). Four grams of soils were weighed into the centrifuge tubes with 40 ml of arsenic contaminant mixed for 24 hours for different stock solutions at different concentrations, i.e. 5mg/l, 10 mg/l, 20 mg/l, 50 mg/l, 100 mg/l, 200 mg/l, 300 mg/l, 400 mg/l, and 500 mg/l. The test was also performed using different pH in the initial solutions, i.e. 2, 3, 4, 5, 6, 7, 8, 9 and 10. Kinetic test was also conducted to investigate the effect of time on arsenic sorption by remediation materials. The test was conducted at different time intervals such as 1, 5, 10, 20, 40, 60, 90, 120, 360, 720, 1440 and 2880 minutes. After it reached equilibrium stage, the tube was centrifuged at 1000 - 2000 revolutions per minute (RPM). The solutions yielded were analysed using inductively coupled plasma mass spectrometry (ICP-MS). The arsenic left in the solution was used to calculate the amounts absorbed by the remediation materials. The sorption isotherms for all soil samples were plotted between the amounts of arsenic species removed from nitrate solution per gram of soil sample,  $C_{\text{ads}}$  in (mg/kg) and as solute,  $C_{\text{aq}}$  in (mg/L). The values of  $C_{\text{ads}}$  were calculated using the equation below:

$$C_{ads} = \frac{V_w (C_o - C_{aq})}{M_{sed}}$$

Where  $C_{ads}$  is the amount of the metal absorbed per mass unit of soil (mg/kg),  $C_o$  and  $C_{aq}$  are the arsenic concentration in the initial solution and after the reaction time in the filtrate, respectively (mg/l).  $V_w$  is the volume of solution added (ml), and  $M_{sed}$  is the air-dried mass of soil (g).

Mini column centrifuge test was conducted to investigate the sorption and transport of arsenic through the remediation materials. The experiments studied the metals both without competition, and in pairs, so that the effect of competition could be assessed. In the single-element mini column test, a solution of metal as its sodium arsenate ( $Na_2HAsO_4 \cdot 7H_2O$ ) was infiltrated at a concentration of 100 mg/l. This concentration was chosen to permit good delineation of the breakthrough curves without significant analytical difficulties.

Materials samples weighing 4 g were placed into the infiltration centrifuge cell. The cell had an upper section with a porous base, which retained the clay, and a lower section in which the leachate was collected (Figure 2). In use, the cell resembled a falling head permeameter. The centrifuge equipment permitted sixteen infiltration cells to be used at once. Dried samples were slurried with deionised water, left overnight so that it may be saturated, and then placed in the infiltration cell. Initially the cells were run until one pore volume of water was collected. The centrifuge was then stopped, the supernatant fluid replaced with leachant solutions, and infiltration tests were performed at a constant temperature of 218°C, at 2600 gravities. Periodically, the centrifuge was stopped and fresh leachant was added on the top of the cell to replace fluid passing through the clay; the collected leachate was removed at the same time (Antoniadis *et al.* 2007).



**Fig. 2:** Mini column centrifuge cell

This leachate was acidified with 5%  $HNO_3$  and then stored in a refrigerator (at 48°C), until the concentrations of heavy metals were determined by inductively coupled plasma mass spectroscopy (ICP-MS).

## RESULTS AND DISCUSSION

The physico-chemical properties for all samples are summarized in Table 1. Peat soil had a very high water holding capacity because soil had very high organic matter more than 94% (Ochsner *et al.* 2006). While the bauxite and iron concretion were ranging between 24.4 to 26 and 15 to 28; respectively. The pH reading for peat soil and bauxite were ranging at acidic side where it were ranging between 4 to 4.2 for peat and 5.1 to 5.4 for bauxite which confirm the finding by (Yonebayashi *et al.* 1994) that peat soil is generally acid. On other hand, the pH reading for iron concretion 5.9 to 6.1. SSA results show that peat soil had high reading which was ranging between 422 to 533  $m^2/g$  when, SSA reading was lower for bauxite and iron concretion

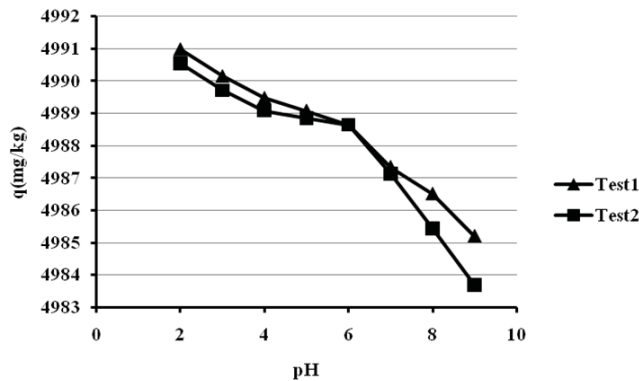
**Table 1:** The physico-chemical properties

Parameters	Peat Soil	Bauxite	Iron concretion
W%	200 - 405	24.4-26	15-28
Porosity %	51 - 54	34-40	29-35
Clay %	ND	5-15	3-4
pH	4 - 4.2	5.1-5.4	5.9-6.1
SSA (m <sup>2</sup> /g)	422 - 533	19- 22	23-27
CEC (meq/100g)	33- 48	24 - 33	59-65
O.M%	94 - 98	4 - 5	2-3
C.C%	5.7 - 6.4	2 -2.5	0-1.5
Mineralogy	ND	Gb > H>K	Go>H>K

Gb =Gibbsite, K= Kaolinite, Go=Goethite, H= Hematite, W%= water content, Gs= specific gravity, SSA= specific surface area, CEC= cation exchange capacity, O.M= organic matter, C.C= Carbonate content

where it were ranging between 19 to 22 m<sup>2</sup>/g for bauxite and 23-27 m<sup>2</sup>/g for iron concretion. The porosity is 51–54 %, 34-40 % and 29-35 % for peat soil, bauxite and iron concretion respectively. Peat soil has more than 90% of organic matter which gave it high reading of CEC and SSA more than bauxite and iron concretion. The percentage of organic matter in bauxite ranging between 4–5 % and 2-3 % for iron concretion. Cation exchange capacity (CEC) result showed that iron concretion has high adsorption capacity with 59-65 (meq/100g) compared to peat soil 33-48 (meq/100g) and bauxite 24–33 (meq/100g). The sorption capacity of the mineral ? increases with the increasing value of cation exchange (Sharma & Reddy 2004). These results strengthen the capacity of the mineral adsorption of contamination. The sorption capability of the bauxite increases with the increasing values of CEC and SSA (Yong 2001; Wan Zuhairi 2003a, 2003b). Iron concretion sample has low carbonate content compared to other materials. According to Yong (2001), carbonate content in soil will lower down the values of SSA. The discrepancy in SSA and CEC values is due mainly to different types of mineralogy in these soils. Bauxite soil contains gibbsite, hematite and kaolinite and iron concretion contains goethite, hematite and kaolinite. It is an active clay mineral that increases the CEC and SSA values. However, it is absent in peat soils resulting in small values of CEC.

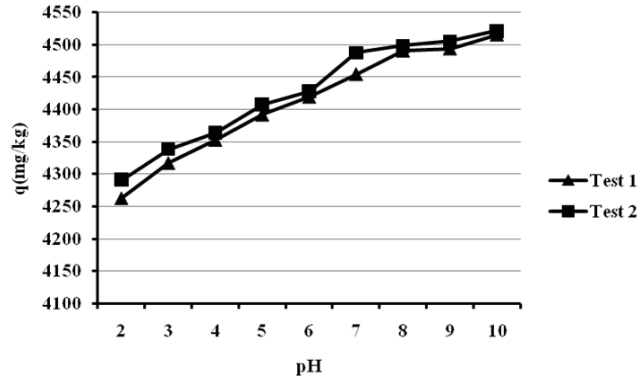
The effect of arsenic pH on bauxite sorption is shown in Figure 3. The pH is an important factor affecting the removal of arsenic from aqueous solution. The results show that arsenic adsorption capacity was higher when the pH was acidic compared with the alkaline pH . Where the highest adsorption value of arsenic was 4991 mg/kg in the first test and 4990 mg/kg in second test at pH 2 which refers to more than 99.8% of arsenic was removed. However, above pH 3 there was a slight decreased in uptake of arsenic to 4988 mg/kg at pH 6 and further reduced to 4554 mg/kg at pH 10. As it is clear in Figure 3, the removal of arsenic is consistent in a wide pH range of 2 to 6 indicating that arsenic is effectively adsorbed by bauxite. Xu *et al.* (2002) reported that at pH range between 3 to 6 arsenic ion occurs mainly in the form of H<sub>2</sub>AsO<sub>4</sub><sup>-</sup> whereas, the divalent anion HAsO<sub>4</sub><sup>2-</sup> dominates at higher pH of 8–10. Also in the intermediate pH range of 6 to 8, both species co-exist.



**Fig. 3:** Effect of pH on arsenic sorption using bauxite

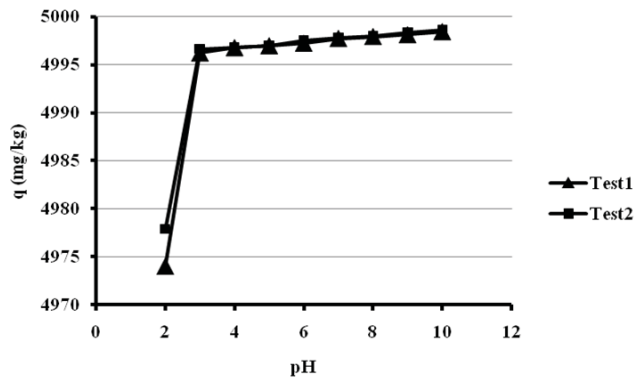
The adsorption mechanism at pH range 3 to 6 may be due to the binding of the arsenic species to the available partially positive surfaces (Bhakat *et al.* 2006; Mohapatra *et al.* 2008). Whereas, the decrease in the adsorption yields above pH 6 may be attributed to the increasing electrostatic repulsion between the negative surface sites and the negative arsenic species. Bahakat *et al.* (2006) found that consistency over a wide pH range gives bauxite an edge for its practical application in drinking water treatment.

Figure 4 shows the effect of pH on the sorption of arsenic by peat soil. We found that when the pH was at 2, the amount of adsorption was 4263 mg/kg. When the pH increases to 10, it was found that the amount of adsorption increased to 4516 mg/kg. Therefore, when pH was high, (alkaline), the soil was able to remove the largest amount of arsenic compared to when pH was low (acid).



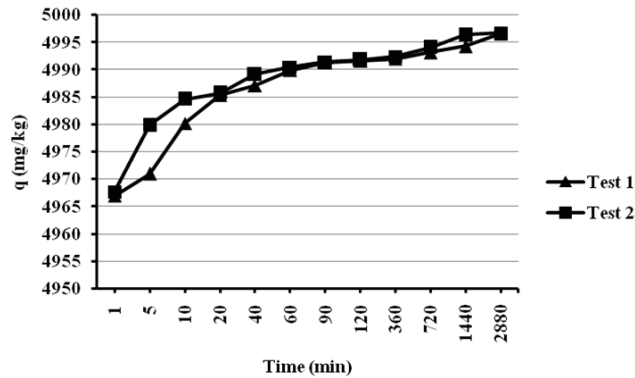
**Fig. 4:** Effect of pH on arsenic sorption using peat soil

The pH effect on the sorption of arsenic by iron concretion is presented in Figure 5. Arsenic shows a decrease in sorption as pH increases over the range 4 to 10. The variation of arsenic sorption onto iron concretion with pH is similar in some respects to arsenic sorption reported onto iron hydroxides (Aggett *et al.* 1981). A possible explanation for this behavior is that sorption of arsenic is affected by an increased repulsion between negatively-charged arsenate species and negatively-charged surface sites thus increasing competition with OH<sup>-</sup> for sorption sites. Because of the complex mineralogy and multiple oxides in iron concretion, it is not clear if the behavior we observe is the result of several oxides acting independently or an inherent synergistic property of oxides with several constituents (Partey *et al.* 2008). What is clear is that iron concretion arsenic sorption is not the result of the action of ferric and aluminum oxides alone. Goldberg and Johnston (2001) show a similar trend for arsenic sorption onto iron and aluminum oxide at pH 4–10.



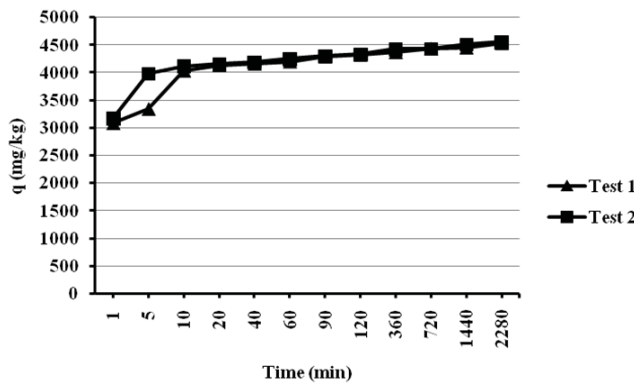
**Fig. 5:** Effect of pH on arsenic sorption using iron concretion

Figure 6 represents the effect of contact time (1 to 2880 min) of arsenic adsorption on bauxite. The kinetic test was performed to investigate the effect of time on adsorption of arsenic. Increasing of the shaking time helps mineral bauxite to remove the largest amount of arsenic. The highest value of the adsorbed arsenic was 4996 mg/kg after 48 hours of shaking which means that 99.9% of the arsenic was removed. The lowest value was 4966 mg/kg, i.e. 99.3% of arsenic removed after 1 minute shaking. It is noted that the removal of arsenic increases with time. In addition, kinetic test showed that bauxite have high capacity to adsorption of arsenic and the adsorption increase with the increasing of shaking time.



**Fig. 6:** Effect of kinetic sorption on arsenic sorption using bauxite

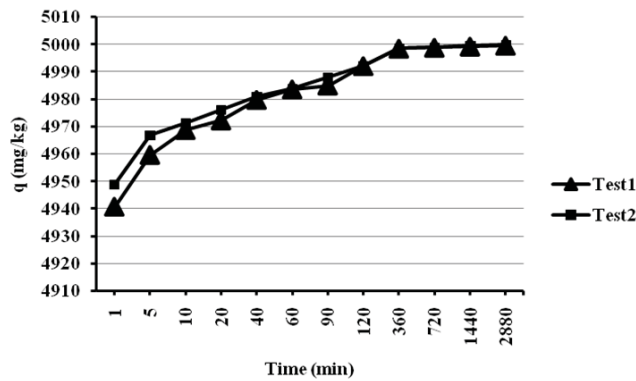
The kinetic test was performed to investigate the effect of time on adsorption of arsenic on peat soil. Increasing of the shaking time helps soil to remove the largest amount of arsenic. The highest value of the absorbed arsenic was 4559 mg/kg after 48 hours of shaking which means that 91.2% of the arsenic was removed. The lowest value was 3088 mg/kg, i.e. 61.7% of arsenic removed after 1 minute shaking. It is noted that the removal of arsenic increases with time. The results of removal are shown on Figure 7.



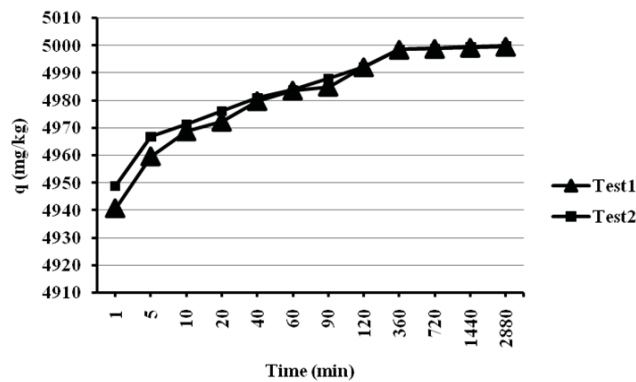
**Fig. 7:** Effect of kinetic sorption on arsenic sorption using peat soil

The effect of kinetics test on iron concretion is shown in Figure 8. It is noted that iron concretion show the increasing of capability of arsenic adsorption from the first minute of the shaking time where the amount of arsenic adsorbed is 4940 mg/kg. This totally represents arsenic removal percentage of 98.8 % by iron concretion samples after 1 minute of shaking. The amount of arsenic adsorbed increases with the shaking time until it reaches 4999 mg/kg after 48 hours.

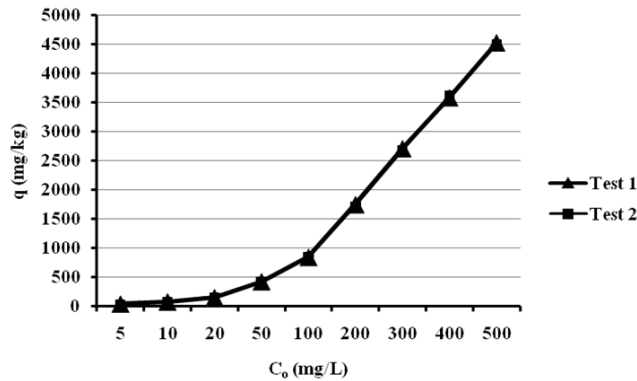
The behavior of arsenic sorption by that's materials at different initial concentrations showed that the sorption of arsenic increases with the increase of initial concentrations. The removal amount of arsenic was 75.5%, 99.28%, 99.74% for peat soil ,bauxite and iron concretion respectively at initial concentration of 10 ppm, while 90.6%, 99.78%, 99.94% of arsenic was removed from the initial concentration of 500ppm for peat soil ,bauxite and iron concretion respectively. Brief results are shown in Figures 9, 10 and 11. This result gives the impression that the adsorption of arsenic by used materials is not affected by the arsenic concentration.



**Fig. 8:** Effect of kinetic sorption on arsenic sorption using iron concretion



**Fig. 9:** Effect of initial concentration on arsenic sorption using peat soil

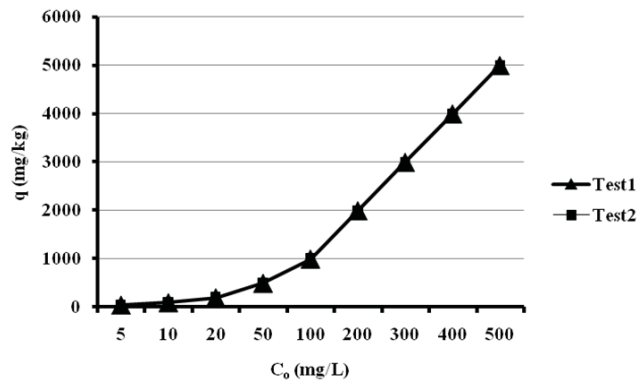


**Fig. 10:** Effect of initial concentration on arsenic sorption using bauxite

Figures 12, 13 and 14 show the results of arsenic concentration in the effluents ( $C_e$ ) relative to the influent concentration ( $C_0$ ) in the in mini column influent and pore volume (PV) for peat soil, bauxite and iron concretion, respectively.

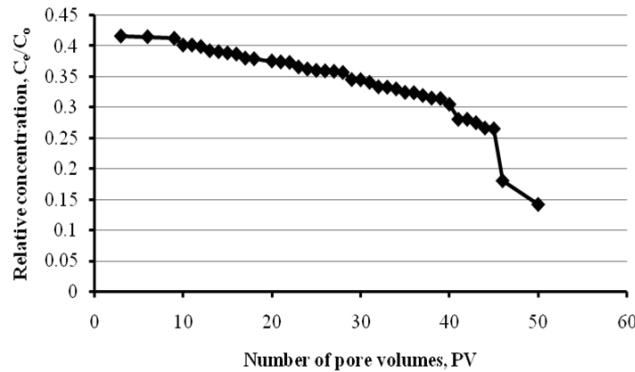
Concerning the breakthrough curve obtained with peat soil, it is presented in Figure 12. The pore volumes (PV) treated by peat soil were about 50 until total arsenic and the effluent was up to 18 mg/L where the initial concentration was 500 mg/L and about 85.7 %. Meanwhile, the high concentration of arsenic on 3 PV was 53.9 mg/L; that was 58.3 % of arsenic removal by peat soil. This result shows that the arsenic removal by peat soil has increased with the increasing of PV. Moreover, it is a clear pattern that has been the





**Fig. 11:** Effect of initial concentration on arsenic sorption using iron concretion

process of arsenic adsorption by mini column test where the removal curve was increasing. The relative concentration value ( $C_t/C_0$ ) in the mini column influent varied from 0.26 to 0.41 during the early operational periods, which were beyond the optimum value for the PV from 1 to 44, and then reached 0.14 after 50 PV. That is because the peat soil has very high surface area 422-533  $m^2/g$  and high porosity 48 % to 54 % which helps to increase the possibility of arsenic adsorption. According to Garelick *et al.* (2005), the porosity properties and chemical characteristics enable the increase in the arsenic adsorption capacities.

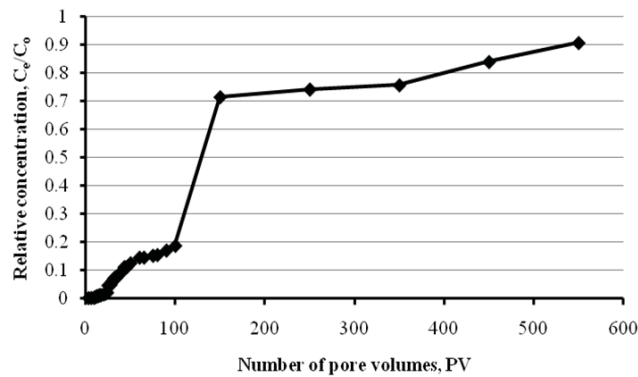


**Fig. 12:** Breakthrough curves of arsenic removal by peat soil

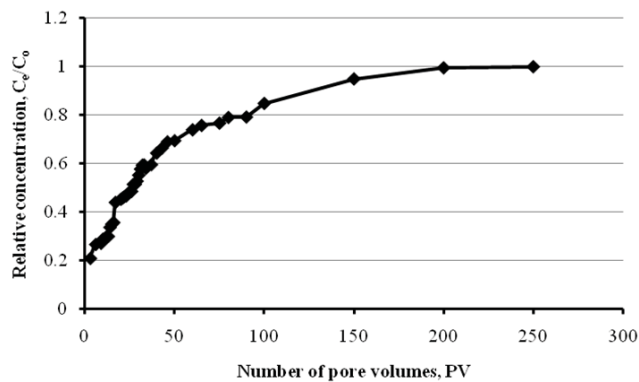
For a practical application of bauxite to the continuous removal of arsenic from contaminated water, the mini column test was conducted to compare the measured adsorption capacity for the batch and mini column systems. The result of mini column test in Figure 13 shows that bauxite has a high capacity to adsorb of arsenic. From 3 PV until 100 PV more than 93 % of arsenic was adsorbed by bauxite and the  $C_t/C_0$  values were 0.0014 and 0.187, respectively. However, after 100 PV to 550 PV, the maximum PV value, the percentage of removal ranged between 70 % to 79 % with 0.72 and 0.91 of  $C_t/C_0$  value. Bhakat *et al.* (2006) found that the maximum removal of arsenic was 90 % using bauxite in mini column test.

Figure 14 shows the ratio of  $C_t/C_0$  which presented the arsenic adsorption by iron concretion. The mini column test was conducted up to 550 PV. The result of mini column test showed that iron concretion can be used to remove arsenic with the ratio of  $C_t/C_0$  0.21 after 3 PV. It was a slow process so the adsorption decreases when the pore volume increases (Figure 13). On the other hand, the lowest adsorption was obtained through the experiment at 550 PV and removal percentage 0.032 % only. According to Daus *et al.* (2004), preferential flow paths may have a significant influence on the early break-through using such small-scale experiments. However, the conditions were the same for all materials and therefore comparable.





**Fig. 13:** Breakthrough curves of arsenic removal by bauxite



**Fig. 14:** Breakthrough curves of arsenic removal by iron concretion

**Conclusion:**

The results revealed that peat soil, bauxite and iron concretion are a potential adsorbent for arsenic removal from the aqueous environment. Iron concretion has more suitable properties such as high values of CEC compared with peat soil and bauxite resulting, high adsorption capacity that make iron concretion as a potential natural material to remove arsenic from contaminated water. In general, the use of these materials is a positive view of the economic cost and large capacity and high effective of remediate contaminated water to make it more useful for human.

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