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Removal of Arsenic from Contaminated Water by Iron Based Titanium-Dioxide from Beach Sand

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Abstract: Iron based Titanium dioxide composites were prepared from beach sand of Cox's Bazaar, Bangladesh, to monitor their efficiency in removing arsenic from contaminated water by column adsorption filtration method in laboratory condition. It is observed that the prepared Iron based Titanium dioxide composite (ITDO-1 and ITDO-2) can do remove 100% both states of arsenic from the prepared 10 mg L^{-1} arsenic solution up to the breakthrough volume (ITDO-1 at 3.1 L and ITDO-2 at 1.9 L, for 4 g of column material) in each case. Adsorbed arsenic is amounted to 4871 mg kg^{-1} of composite ITDO-1 and removal efficiency is found 72.46% up to the saturation volume (7.35 L) while the corresponding figure for composite ITDO-2 is 6708 mg kg^{-1} of material with an efficiency of 62.92% up to saturation volume (7.5 L). This study also revealed that the residue (ITDO-3) left after the prepared ITDO-1 and ITDO-2, exhibited least performance on removing arsenic.

Key words: Adsorption filtration, adsorbing materials, arsenic pollution, saturation volume

INTRODUCTION

Alluvial Ganges aquifers used for public water supply are polluted with naturally occurring arsenic which adversely affects the health of millions of people in Bangladesh and India (West Bengal) (Nickson *et al*., 1998). This arsenic contaminated water causes widespread death and disease in arsenic affected area. To overcome this problem some procedure and system have been investigated, but for complete solution more research is needed on mitigation and suitable technique that can help millions of people to secure arsenic free water.

Recently, high-capacity arsenic-selective adsorbents including activated alumina (AA), modified activated alumina (MAA), granular ferric hydroxide (GFH), granular ferric oxide (GFO) and granular titanium dioxide (TiO_2) have been developed and implemented (Driehaus *et al*., 1998; Bang *et al*., 2005; Westerhoff *et al*., 2006). Furthermore, water treatment methods based upon the process of aeration, coagulation and sand-filtration can remove a substantial amount of the arsenic by co-precipitation with iron have been described by Joshi and Chaudhuri (1996), Bhattacharaya *et al*. (1997) and Raven *et al*. (1998) and show promise to local use.

Ferric oxide and titanium dioxide can be used as a potential adsorbing material for arsenic removal. Some well documented research also revealed that sedimentary iron-oxyhydroxide are known to scavenge arsenic (Joshi and Chaudhuri, 1996) and arsenite and arsenate have strong sorption affinity for iron hydroxide and oxyhydroxide minerals such as ferrihydrite and goethite (O'Day, 2006). In

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addition, Titanium dioxide, a non-toxic semiconductor, is stable over a pH range of 2-14 (Esumi *et al*., 1998) and possess strong attraction toward arsenic though it has long been used as a light-inducing catalyst (Balaji *et al*., 2002). Another study suggests that because of the high surface area and the presence of high affinity surface hydroxyl groups in TiO_2, it can be used as an effective adsorbent for arsenic removal (Pena *et al*., 2006).

In the present research, ilmenite collected from Cox's bazaar beach sand was used as a raw material for iron based titanium dioxide composite. Ilmenite is a weakly magnetic titanium-iron oxide mineral which is iron-black or steel-gray. It is a crystalline iron titanium oxide ($Fe^{++}TiO_3$), a common accessory mineral in igneous and metamorphic rocks and commonly concentrated in placers as black sand deposits. Although, there is apparent evidence of the complete range of mineral chemistries in the (Fe, Mg, Mn, Ti)O_3 system naturally occurring on Earth, the vast bulk of ilmenites are restricted to close to the ideal $FeTiO_3$ composition, with minor mole percentages of Mn and Mg.

Adsorption filtration method is preferred for arsenic removal in small-scale treatment system because of its simplicity, ease of operation and handling, regeneration capacity and sludge free operation (Thirunavukkarasu *et al*., 2003; Nurul *et at*., 2006). The present study investigates the possibility of the use of Iron based Titanium dioxide as an adsorbing material from available natural source (beach sand from Cox's Bazaar) since it can reduce the cost as well as provide better performance in removing arsenic.

MATERIALS AND METHODS

Preparation of ITDO Composites

Freshly prepared 200 mL Aqua Regia was added to the 200 g beach sand (supplied by Carbon Mining Company Ltd., Dhaka) from Cox's Bazaar, Bangladesh and then heated at 200°C with sand bath until dried; 200 mL water was added and stirred slowly. The yellow slurry formed was filtered and 200 mL 5% ammonia solution was mixed into the filtrate. After five minutes reddish-brown precipitate was settled. The precipitate thus obtained was dried at 100°C in a temperature controlled oven for 6 h to obtain a reddish-brown solid and named it ITDO-1. The residue from the preparation of composite ITDO-1 was washed with deionized water and filtered by filter paper and then 200 mL 5% ammonia solution was dropped into filtrate. After few minutes reddish-brown precipitate was deposited which was separated by using filter paper. The obtained precipitate was dried at 100°C in a controlled oven for 6 h and again a reddish-brown solid which named ITDO-2 was obtained. The residue left from the preparation of composite ITDO-2 was used as an adsorbing materials ITDO-3.

Column Preparation

Three columns were made with 4 g uniformly grained ITDO-1, ITDO-2 and ITDO-3 (particle size 0.1 cm) and the prepared 10 mg L^{-1} As^{3+}/As^{5+} solution was passed through the columns until the break through volume as well as saturation volume of the ITDO-1, 2, 3 was reached. Water samples that passed through the columns were collected in the sample bottles after several time intervals. The flow rates of the column ITDO-1, 2 and 3 were measured and it was 3, 3.2 and 3.5 mL min^{-1}, respectively.

Arsenic Detection Method

The silver diethyldithiocarbamate (SDDC) colorimetric method was employed by Eaton *et al*. (2005) that based on the evolution of arsine gas in which inorganic arsenic is reduced to arsine, AsH_3, by zinc in acid milieu, the arsine is bubbled through a solution of silver diethyldithiocarbamate, $AgS.CS.N(C_2H_5)_2$, in pyridine or chloroform; a red soluble complex is formed that can be measured photometrically at a specific wavelength of 535 nm.

Arsine Generation

Collected water sample (10 mL) was taken into the generator flask followed by the addition of 5 mL concentrated hydrochloric acid, 2 mL 15% potassium iodide and 10 drops of stannous chloride solution. It was allowed to stand, with random agitation for about 15 min to ensure complete reduction of As (V) to As (III). The absorption tube was charged with 4.00 mL of SDDC solution. Cotton wool impregnated with lead acetate solution was placed in the scrubber to absorb any hydrogen sulphide, which may be subsequently evolved. After adding three pieces of pure granulated zinc to the solution in the generating flask, the scrubber-absorber was connected immediately. The evolution of arsine was completed 99% in 30 min and virtually finished in about 45 min. The volume of the solution was readjusted to the original volume and then poured into a 1 cm cell and the absorbance was recorded at 535 nm using the reagent (SDDC solution) as the reference.

Preparation of Standard Curve for Arsenic Measurement

For measuring the arsenic content in the collected water sample it was essential to prepare standard curves. A mother solution of 10 mg L^{-1} (As^{3+}/As^{5+} at 1:1 ratio) was prepared and it was diluted to many other intermediate solutions of different concentrations. After preparing all these solutions, their absorbance were measured in spectrophotometer (M-390) and standard curve was generated for total arsenic (As) in various concentrations.

RESULTS AND DISCUSSION

In the present investigation, it is observed that iron based titanium dioxide can successfully remove both states of (As^{3+} and As^{5+}) arsenic. The amount of arsenic passed through the ITDO-1 composite is 17695.13 mg kg^{-1} and the amount of arsenic absorbed 4871.75 mg kg^{-1} in Table 1. So, the average arsenic adsorbing capacity of ITDO-1 is 72.46% (4 g of the absorbent). Similarly, the amount of arsenic passed through the ITDO-2 is 18120 mg kg^{-1} and the amount of arsenic absorbed 6708 mg kg^{-1} in Table 2. Therefore, the mean arsenic adsorbing capacity of ITDO-2 is 62.98% (4 g of the absorbent). This study also supports the previous finding performed by Bissen *et al*. (2001), Manna *et al*. (2004), Dutta *et al*. (2004) and Pena *et al*. (2006).

Table 1: Arsenic (As^{3+}/As^{5+} at 1:1 ratio) removing performance by ITDO-1

Amount of adsorbent (g)	Initial As concentration (mg L^{-1})	Volume passed (L)	As concentration (mg L^{-1})	Amount of As adsorbed (mg)
4	10	3.1	0.162	0.491
		3.3	0.77	2.337
		3.5	2.3	3.877
		3.7	2.53	5.371
		3.9	3.4	6.691
		4.1	4.1	7.871
		4.3	4.53	8.965
		4.5	3.25	10.315
		4.7	3.8	11.555
		4.9	4.6	12.635
		5.1	5.18	13.599
		5.3	5.25	14.549
		5.5	5.8	15.389
		5.7	6.2	16.149
		5.9	6.46	16.857
		6.1	5.58	17.741
		6.3	6.8	18.381
		6.5	7.54	18.873
		6.7	8.62	19.149
		6.9	9.23	19.303
		7.1	9.45	19.413
		7.3	9.63	19.487

Table 2: Arsenic (As^{3+}/As^{5+} at 1:1 ratio) removing performance by ITDO-2

Amount of adsorbent (g)	Initial arsenic concentration (mg L^{-1})	Volume passed (L)	As concentration (mg L^{-1})	Amount of arsenic adsorbed (mg)
4	10	1.9	0.157	0.492
		2.3	1.20	4.012
		2.7	2.60	6.972
		3.1	3.20	9.692
		3.5	3.90	12.132
		3.9	4.20	14.452
		4.3	3.80	16.932
		4.7	4.70	19.052
		5.1	5.60	20.812
		5.5	6.20	22.332
		5.9	6.70	23.652
		6.3	6.85	24.912
		6.7	7.40	25.952
		7.1	8.20	26.672
		7.5	9.60	26.832

Table 3: Arsenic (As^{3+}/As^{5+} at 1:1 ratio) removing performance by ITDO-3

Amount of adsorbent (g)	Initial arsenic concentration (mg L^{-1})	Volume passed (L)	As concentration (mg L^{-1})	Amount of As adsorbed (mg)
4	10	0.15	0.935	1.35
		0.35	3.00	2.75
		0.55	5.20	3.71
		0.75	7.45	4.22
		0.95	8.40	4.54
		1.15	9.40	4.66

Bissen *et al*. (2001) reported that As (V) adsorbed faster than As (III) by nanocrystalline TiO_2. Manna *et al*. (2004) investigated the removal of As (III) using a synthesized crystalline hydrous titanium dioxide and the study revealed that 70% of As (III) adsorption occurred within the first 30 min of contact time. Dutta *et al*. (2004) investigated the adsorption of arsenate and arsenite on suspensions of titanium dioxide. Commercially available Hombikat UV100 and Degussa P25 were used to investigate adsorption as a function of pH and adsorbate concentration. It was found that adsorption of arsenate was much higher at pH 4 than the adsorption of arsenite. In contrast, arsenite adsorption was higher than arsenate adsorption at pH 9. Similarly, (Pena *et al*., 2006) determined that adsorption of As (V) was effective below pH 8 and that maximum adsorption of As (III) occurred at a pH of approximately 7.5 in test waters with nanocrystalline titanium dioxide. The findings provided by the above authors suggest that TiO_2 is an effective adsorbent for arsenic removal due to its high surface area and the presence of high affinity surface hydroxyl groups. In addition, field filtration results demonstrated that the granular TiO_2 adsorbent was very effective for the removal of arsenic in groundwater (Bang *et al*., 2005).

However, the present study focused on the both states of arsenic removing performance of iron based titanium dioxide that prepared from available natural source of ilmenite in Cox's bazaar's beach sand. Ilmenite is the potential source of TiO_2 and FeO. It contains 52.65% TiO_2 and 47.35% FeO as well as 31.56% titanium, 36.81% iron and 31.63% oxygen. Therefore, the aqua regia treatment on ilmenite allows iron and titanium metal in solution as digestion dissolves a fraction of metals that can be put into solution under relatively extreme conditions (Manaham, 1999).

The prepared composite ITDO-2 showed better performance than ITDO-1 in adsorbing arsenic from contaminated water. Interestingly, composite ITDO-2 can adsorb 1836.25 mg kg^{-1} more arsenic than that of ITDO-1. Though, iron play a significant role to remove arsenic, titanium dioxide also helps to bind arsenic with it. The extended X-ray absorption fine structure (EXAFS) spectroscopy study indicated that As (V) and As (III) formed bidentate binuclear surface complexes as evidenced by an average Ti-As (V) bond distance of 3.30Å and Ti-As (III) bond distance of 3.35Å (Pena *et al*., 2006).

However, the residue ITDO-3 left after preparing the two composite ITDO-1 and ITDO-2 showed least performance on removal of arsenic and the reason is that it contains less amount of iron and titanium oxide (Table 3).

Overall, from the results it seems that arsenic removal is possible by iron based titanium dioxide (from Cox's Bazaar's beach sand that contain ilmenite) a low-cost adsorbent which exhibit superior adsorption capacities and local availability.

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