Proficient Removal of As(III) from Water Using Orchid Plant (*Vanda* sp.) as Biosorbent

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This paper reports the use of an orchid plant of *Vanda* species for absorptive removal of hazardous As(III) from aqueous solution. Optimum conditions were determined by analyzing various parameters such as pH, contact time, biomass dose and initial metal ion concentration and the interfering radicals. In batch studies the percentage removal of As(III) was 98 % per gram of adsorbent for As(III) concentration of 2 mg/L. The adsorption isotherm models such as Langmuir and Freundlich were studied and both were found to be valid. Characterization of biosorbent was done by FTIR and SEM-EDS analysis. FTIR analysis confirms the biosorption process involved OH, CO and NH functional group in the removal of As(III). SEM result shows structural changes on surface and EDS result reveals the presence of As(III) on the surface of biosorbent. The maximum biosorption potential, through the continuous flow study was 0.9 mg/g.

Keywords: Arsenic(III), Biosorption, Biosorbent, Water, *Vanda* species.

INTRODUCTION

Arsenic is a well known toxic element which can cause acute and chronic toxic effects in the environment and has historically been used as a poison [1]. Arsenic enters in the body of human and plant through groundwater. There are many reports which confirm the presence of arsenic as a hazardous element in high concentration in drinking water in India [2-5]. Arsenic is introduced in natural water through natural processes such as geological, biological, weathering reactions and volcanic emissions [6] as well as through anthropogenic activities such as application of pesticides, herbicides, gold mining, non-ferrous smelting, petroleum refining and combustion of fossil fuels in power plants [7,8]. Consequently, drinking water arsenic is reported to affecting millions of people all over the world. Ingested arsenic in the human body affects almost all cellular mechanism [9]. Long term exposure to arsenic significantly increases the risk of various disorders such as dermal (Blackfoot disease), cardiovascular, reproduction, immune system and cancers of skin, liver, lungs, bladder and kidney [10-14].

The inorganic trivalent arsenic compounds are more toxic than pentavalent organo arsenic [15]. Inorganic arsenic is also classified as a human carcinogen by the International Agency for Research on Cancer [16]. Based upon these health issues, the World Health Organization recommended the maximum permissible concentration of arsenic in drinking water as 10

µg/L [17]. Hence, its removal is an important area of continued research.

Various conventional methods are used for the removal of arsenic such as coprecipitation, adsorption, electrolysis, ion exchange, coagulation, reverse osmosis [18-23] and also oxidation of As(III) followed by removal of total arsenic using ferric hydroxide, ferric chloride [24]. Most of the above technologies are not easily adoptable owing to the reasons such as being technically demanding, expensive or resulting in incomplete removal, or the production of large amount of sludge. The sludge thus produced requires further treatment before being disposed. The biosorption, on the other hand, is an effective treatment method to remove arsenic from water using inexpensive biomaterial. Biosorption process is advantageous as the methods are generally inexpensive, green method, display short resolve time, reusability of the biomass, improved selectivity for specific metals of interest, removal of heavy metal from effluent irrespective of toxicity and no production of secondary compounds which might be toxic [25-28].

In our previous work we have already reported the effectiveness of *Vanda* species for the removal of toxic Pb(II) ion [29] so, its efficient adsorption capacity, low cost and ease in availability in the forest of Chhattisgarh, India encouraged us to work on further removal of toxic elements.

The objective of this study was to investigate the potential of *Vanda* sp. as biosorbent for the removal of As(III). Both the

batch mode and continuous flow method were performed to evaluate maximum efficiency of biomaterial. Various experiments were performed in batch mode study such as the effect of pH, contact time, initial metal ion concentration and biomass dose *etc.* to obtain the optimum removal. The Langmuir and Freundlich models were used to describe equilibrium isotherms. FTIR and SEM-EDS were used to find out the biosorption mechanism of As(III) by *Vanda* sp.

EXPERIMENTAL

Preparation of stock solution: All the chemicals used in this study were of AR grade Merck (India/Germany). All the solutions were prepared in double distilled water. A stock solution of As(III) (1000 mg/L) was prepared by dissolving the appropriate amount of sodium arsenite in double distilled water. pH of the solution was adjusted using 0.1 M of HCl acid solution and 0.1 M NaOH solution. The desired concentration of lead solution was prepared by diluting 1000 mg/L stock solution in double distilled water. All the glass wares were washed with 5 % HNO₃ solution and then rinsed thoroughly by tap water and distilled water, respectively.

Preparation of biosorbent: The *Vanda* sp. was collected from the forest area of Chhattisgarh, India. Whole plant (leaf, stem and root) was used as a biomaterial for the study. The collected plant material was adequately dried in sunlight. The dried plant was then grinded and sieved using the standard sieves. The biomaterial used for experiment had standard sieve size of 1.18 mm.

Before its use the biomaterial was washed using 0.01 N HCl and then repetitively with double distilled water until the filtrate pH values was near normal. The biomaterial was then dried up in a hot air oven and stored in an air tight container. This process ensured clean and primed biomaterial for the adsorptive studies.

Batch study: To find out the biosorption efficiency, batch experiments were carried out. Batch experiments were performed by taking As(III) solution (0.5-6.0 mg/L) in a flask, pH (2-8) of solution was adjusted by adding dilute HCl/NaOH and then the biosorbent dosage (0.5-4.0 g) was added to the solution. The flask was then well shaken and then was kept still for the desired contact time (5-120 min) with occasional shaking. After equilibration time, the solution was filtered by vacuum filtration to separate the biomass. Then filtrate part was analyzed by fast sequential atomic absorption spectrometer (AA240FS, Varian) with hydride generator to measure the metal concentration after sorption. The percentage removal efficiency of *Vanda* sp. for As(III) was evaluated by following equation:

$$\text{Removal of As(III) (\%)} = \left(\frac{C_i - C_f}{C_i} \right) \times 100 \quad (1)$$

where, C_i and C_f is the concentration of As(III) before and after sorption. After equilibration study, the co-ion experiments were also conducted.

Methodology of column study: The continuous flow mode study was carried out using glass column. The internal diameter and height of the glass column was 4.5 cm and 70 cm respectively. Glass beads were loaded in the bottom of the

column up to a height of 2 cm. The weighted amount of biosorbent (20 g) was filled in glass column. The biosorbent was then covered with another layer of glass beads up to 2 cm height, to prevent the biomass from floating. This filling method was adopted to ensure a uniform elute distribution through the biosorbent bed.

Arsenic(III) solution having an initial concentration of 1 mg/L was then pumped through column in an up flow mode with a flow rate of 13 mL/min using peristaltic pump. The influent was adjusted to optimum pH using 0.01 M NaOH and 0.01 M HCl. Effluent solution was collected and analyzed for metal concentration by HG-AAS fast sequential atomic absorption spectrometer (AA240FS, Varian). Column experiment was continued until the effluent showed the same concentration as that of the influent.

Characterization of biosorbent material: For confirmation of biosorption of As(III) in *Vanda* sp. biomaterial, the characterization was done before and after biosorption by FTIR analyzer using Thermo Nicolet Avatar 370 having a range of 4000-400 cm⁻¹ and resolution of 4 cm⁻¹, at the Sophisticated Analytical Instruments Facility (SAIF), Cochin, India. The surface morphology of biomaterial before and after biosorption was analyzed by SEM-EDS. A scanning electron microscope ZEISS EVO SEM 18 model equipped with INCA 250 EDS X-MAX 20 mm Detector Oxford was used for SEM-EDS analysis.

RESULTS AND DISCUSSION

An interesting outcome was noted in the beginning of the experiment that this orchid plant possesses a unique ability to tolerate the Pb(II) [29]. Based on this properly it was hypothesized that the plant material may possess a bio-sorptive capability for As(III) also. Therefore, the biosorption of As(III) was carried out using *Vanda* sp. as a biosorbent by various parameters. Different parameters are discussed below:

Effect of pH: The influence of pH showed a significant part in the biosorption of As(III). Experiments were carried out by varying 2-8 pH, having concentration of As(III) solution 1 mg/L. Fig. 1 shows that at pH value 5 the adsorption was 97 %. With the increase of pH value, the percentage removal

Fig. 1. Effect of pH on removal percentage of As(III)

of arsenic was decreased. Good adsorption was observed from 5-8 pH value but highest sorption was observed at pH value of 5. Thus, for further experimentation, the optimum pH value for Arsenic was taken at 5 pH.

Arsenic(III) exists in non-ionic (H_3AsO_3) and anionic ($\text{H}_2\text{AsO}_3^{-1}$ and HAsO_3^{-2}) types in the pH range 2.0-7.0 and 7.5-9.0 respectively [23]. Non-ionic species present at pH range 2-4, hence the removal of As(III) was not favorable on surface of biosorbent. With the increase of pH, there was gradual increase in interaction between adsorbent surface and charged adsorbate species [30]. At higher pH value the OH^- ion starts dominating and thus biosorbent surface becomes negative charge [31]. Therefore, at alkaline condition a repulsive force may arise between the negative surfaces of sorbent and H_2AsO_3^- species resulting in decrease of adsorption.

Biosorbent dose: To study the influence of biosorbent dosage on the percentage removal of As(III). Biosorbent dose also determine the uptake capacity of biosorbent per unit of mass. To investigate this biosorbent dose was ranging from 0.5 g to 4 g at optimum pH value of 5 and the concentration of solution was 1 g/50 mL. Fig. 2 represents the result that 97 % of As(III) was removed from 1 g of biosorbent dose. In this dose sufficient quantity of active sites present to interacts with As(III) ions in the solution. With increase in the dose of biosorbent material, no change was observed in the percentage removal of As(III). This effect was due to the aggregation of biosorbent material present in the mixture. Therefore, the optimum dose was taken 1 g for further experiments.

Fig. 2. Effect of biosorbent dose on percentage removal of As(III)

Contact time: To study the contact time for biosorption of As(III) by ranging time interval from 5 to 120 min. Fig. 3 represents the result obtained from the experiment. Result shows that within 5 min, the 80 % of biosorption took place and it was observed that 97 % of adsorption attained by increasing the reaction time up to 30 min. This rapid increase in adsorption was due to biosorption sites available on the surface of biosorbent. Further there was gradual or insignificant increase in biosorption had observed. Therefore, the optimum contact time for As(III) removal was 30 min.

Fig. 3. Effect of contact time for biosorption of As(III)

Initial As(III) ion concentration: The effect of As(III) concentration on removal of adsorbate was studied by varying concentration of As(III) from 0.5 to 6 mg/L. Fig. 4 indicates the result that the percentage removal decreases with increase in concentration of As(III). 100 % removal was observed at 0.5 mg/L and 98 % of As(II) removal was observed at 1 mg/L which decreases to 71.60 % removal of As(III) at 6 mg/L. Initially, higher percentage removal was observed due to a lesser amount of metal ions present in the solution to approach free sites on the surface of biosorbent. But with the continuous increase in concentration of As(III) ion, the removal percentage get decreases due to decrease in availability of active free sites on surface of biosorbent. The optimum As(III) concentration was taken 1 mg/L. The uptake capacity of biosorbent, adsorbed As(III) ion per unit mass was calculated by following equation:

$$\text{Uptake capacity} = C_i - C_f \times \frac{V}{M} \quad (2)$$

where, C_i is the initial As(III) ion concentration (mg/L), C_f is the concentration of As(III) at equilibrium (mg/L), V is the volume of As(III) solution (L) and M is the weight of biosorbent (g). The uptake capacity of *Vanda* sp. for removal of As(III) was 0.215 mg/g.

Fig. 4. Effect of initial As(III) ion concentration on biosorption of As(III)

Interfering ion study: To study the influence of common ions interference in biosorption of As(III) by biosorbent were investigated. The interference of fluoride, nitrate, sulfate, phosphate, iron, copper, chromium and cadmium shows result that the percentage removal of As(III) were decreases up to 7-8 % but this difference of percentage removal was not so significant. In the presence of nickel and zinc, the percentage removal was observed the decreasing from 98 to 80 % for nickel and 85 % for zinc. But there is no significant changes were found in the adsorption by manganese. With the interference of lead, the removal efficiency was same as without interference that is 98 %.

FTIR analysis of unloaded and loaded biosorbent: FTIR analysis was obtained to collect the information of possible interaction between the metal ions (As (III)) and cell of biosorbent (*Vanda* sp.). The FTIR spectrum of unloaded and loaded biosorbent was shown in Fig. 5. In unloaded biosorbent, the strong broad band observed at 3430.79 and 2920.78 cm^{-1} were attributed to stretching -OH bond of alcohols and carboxylic acid respectively. The medium peak at 1629.30 cm^{-1} was assigned to primary amine (N-H). The strong band at 1514.05 cm^{-1} shows the presence of strong asymmetric stretching of N-O compounds. The peak at 1036.82 cm^{-1} may be attributed to C-N stretch of aliphatic amine. Another peak at 555.16 cm^{-1} indicates the presence of C-Cl of alkyl halides.

Fig. 5. FTIR spectrums (a) unloaded biosorbent, (b) loaded biosorbent

The spectrum observed after adsorption of As(III) shows significant changes and shifting of peaks. The peak of O-H group at 3430.79 and 2920.78 cm^{-1} were shifted to 3451.33 and 2919.63 cm^{-1} , respectively, indicating As(III) chelation with -OH group present in the biomass surface [32]. After adsorption of As(III), a peak displayed at 1733.78 cm^{-1} due to increase in frequency. This band clearly reveals that there was involvement of carboxyl group in sorption of As(III) [33]. Another shifting observed from 1629.30, 1514.05 and 1036.82 cm^{-1} to 1630.89, 1322.52 and 1058.68 cm^{-1} , respectively, due to the complexation of nitrogen from amide group with As(III) [34]. The above results shows that there was interaction between As(III) and hydroxyl, carboxyl and amine group present on the surface of biosorbent.

Scanning electron microscopic and electron diffraction

X-ray: The *Vanda* sp. morphology structure of unloaded and loaded biomass was studied during biosorption process with the help of SEM. Fig. 6(a), shows that before adsorption of As(III), the rough surface and spherical clusters with large number of small pores were present on the surface. This surface structure may be provides sites for the sorption of As(III) [35]. Fig. 6(b) shows that the small particles adhered on the surface and reduction in pores was clearly visible after adsorption of As(III) in biosorbent. EDS analysis provides the confirmation of adsorption by elemental analysis of surface composition. Fig. 7(a) clearly shows that before adsorption, As(III) was not present in the structure of *Vanda* sp. But after adsorption in Fig. 7(b) As(III) was observed in the surface due to sorption of As(III) by biosorbent material. SEM-EDS result analysis conclude that there was biosorption As(III) by the *Vanda* sp.

Fig. 6. SEM analyses (a) unloaded biosorbent (b) loaded biosorbent

Fig. 7(a). EDS analyses of unloaded biosorbent

Fig. 7(b). EDS analyses of loaded biosorbent

Adsorption isotherm: The process of adsorption isotherm is the relation between amounts of adsorbate adsorbed on the surface of adsorbent at equilibrium condition. Isotherm studies describe about the capacity of unit mass of biosorbent to remove quantity of pollutants. Langmuir and Freundlich sorption isotherm had been studied.

The Langmuir adsorption isotherm explains about the sorption takes place at homogenous surface of the adsorbent. Its linearized form can be written as [36]:

$$\frac{C_e}{q_e} = \frac{1}{bq_m} + \frac{C_e}{q_m} \quad (3)$$

where, C_e is the equilibrium metal ion concentration (mg/L), q_e is the amount of As(III) adsorbed at equilibrium concentration (mg/g), q_m and b are the Langmuir constants of adsorption capacity (mg/g) and energy of adsorption (L/mg) respectively. The values q_m and b were calculated by slope and intercept values of the linear plots of C_e/q_e versus C_e (Fig. 8.) and were presented in Table-1. The correlation coefficient (R^2)

Fig. 8. Langmuir isotherm equation graph

TABLE-1
ADSORPTION ISOTHERM VALUES OF
BIOSORBENT FOR As(III) REMOVAL

Langmuir isotherm values			Freundlich isotherm values		
q_m (mg/g)	b (L/mg)	R^2	K_f (mg/g)	N	R^2
0.21	7.5	0.959	3.499	3.23	0.976

was found to be 0.95. The result favours that biosorption of As(III) by biosorbent were well fitted to Langmuir model. The adsorption capacity of *Vanda* sp. was found to be 0.21 mg/g for As(III).

The Freundlich adsorption isotherm explains adsorption on heterogeneous surface within the sorbent. Its linearized form [37] can be written as:

$$\log q_e = \log K_f + \left(\frac{1}{n}\right) \log C_e \quad (4)$$

where, K_f and $1/n$ were Freundlich constant representing sorption capacity (mg/g) and sorption intensity of the sorbent respectively. The values of K_f and n were calculated from the slope and intercept by plotting q_e versus C_e (Fig. 9). The values of K_f and $1/n$ obtained by equation were 3.49 mg/g and 0.30, respectively. The value of $1/n$ lies between 0 and 1 indicates the biosorption of As(III) and it was favorable under the studied conditions. The R^2 value of Freundlich model was higher than Langmuir model (Table-1), this result indicates that Freundlich model was properly able to describe the relation between amounts of sorbed As(III) and its equilibrium concentration in the solution.

Fig. 9. Freundlich isotherm equation graph

Continuous flow experiment: The continuous flow experiment was conducted to investigate the maximum efficiency of *Vanda* sp. for the sorption of As(III), so it can also be used for industrial application. Packed bed column is classically used to improve the contact between two phases that is solid phase (adsorbent) and liquid phase [As(III) ion concentration]. The adsorption of As(III) in column experiment were represented by breakthrough curve. The breakthrough curve was plotted effluent concentration versus volume (L). Breakthrough point was obtained when the effluent concentration reaches to permissible limit. The effluent concentration solution was collected from upward flow rate of 13 mL/min after each empty bed contact time (EBCT) of 50 min. The solutions were collected and analyzed until the biosorbent was saturated with As(III) ion concentration in the column. This saturation point was known as exhaust point. The breakthrough curve and exhaust curve were plotted according to the results (Fig. 10), which

Fig. 10. Breakthrough and exhaust point graph

shows that 100 % of As(III) was removed till 2 bed volume of effluent. The breakthrough point was also observed after passing 2 bed volume of effluent. The breakthrough point biosorption capacity (BPBC) can be represented as:

$$BPBC = \frac{C_o - V_b}{M} \quad (5)$$

where, C_o is the initial As(III) ion concentration (mg/L), V_b is the breakthrough point, volume of effluent passed (L) and M is the total mass of the biosorbent taken for packed glass column (g).

The exhaust point reaches after passing 18 bed volume of effluent from packed glass column. The exhausted point biosorption capacity (EPBC) can be represented as:

$$EPBC = \frac{C_o - V_e}{M} \quad (6)$$

where, C_o is the initial As(III) ion concentration (mg/L), V_e is the exhausted point, volume of effluent passed (L) and M is the total mass of the biosorbent taken in packed glass column (g).

The calculated BPBC up to 2 bed volume effluent was 0.065 mg/g and the EPBC up to 19 bed volume effluent was 0.9 mg/g. The value of exhausted point biosorption capacity for As(III) was higher than biosorption capacity of batch study using *Vanda* sp.

Comparison of uptake capacity between *Vanda* Sp. with other sorbent: Uptake capacity of other sorbents of previous work was compared with the uptake capacity of recent study. Efficiency of *Vanda* sp. shows better uptake capacity among other results as shown in Table-2.

Conclusion

The biosorbent was proficient to remove As(III) from aqueous solution with uptake capacity of 0.9 mg/g by continuous flow method and 0.21 mg/g by batch mode study. Effect of pH was strong factor affecting on biosorption process and optimum pH value was 5. The sorption was rapid and equilibrium obtain within 30 min of contact time. Hydroxyl, carboxyl and amino groups were involved in biosorption of As(III) on the surface. SEM confirms the change in surface after adsorption and EDS analyses shows the presence of As(III) after

TABLE-2
COMPARISONS OF UPTAKE CAPACITY OF
Vanda sp. WITH OTHER ADSORBENT

Biosorbents	Uptake capacity		Ref.
	mg/g	pH	
Iron oxide-coated sand	0.1360	–	[38]
Zeolites	0.0170	–	[39]
Synthetic iron sulphide	0.2000	–	[40]
<i>Aspergillus niger</i>	0.2000	6.5	[41]
Activated alumina	0.1800	3.5	[42]
Char, oak bark	0.0074	–	[43]
Rice polish (an agricultural residue)	0.1470	7	[44]
<i>Saccharomyces cerevisiae</i>	0.6200	5	[45]
<i>Vanda</i> sp. (batch mode)	0.2100	5	Present study
<i>Vanda</i> sp. (column mode)	0.9000	5	Present study

sorption. R^2 value of Freundlich isotherm was higher than Langmuir isotherm. Freundlich model was well fitted and confirms heterogeneous multilayer adsorption on the surface of biosorbent.

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