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# Modeling and Simulation of Fixed Bed Column for Arsenic Removal using Iron Ore and PAN Fiber Adsorbents

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**ABSTRACT: Globally, arsenic (As) is most significant and serious pollutant in water and causes severe consequences to human health. Owing to this, As removal from wastewater has become a serious concern at present time. Compared with other As removal techniques, adsorption has been selectively and effectively employed. For an economical process offering an adequate performance of As removal, the choice of natural and cheaper adsorbents plays a key role. In this work, adsorption performance of fixed bed adsorption column using indigenous adsorbents i.e., iron ore and modified Polyacrylonitrile (M-PAN) fiber was investigated experimentally. Consequently, Aspen Adsim® software was used to simulate fixed bed adsorption process. The simulation results were compared with the experimental observations obtained on lab-scale for the validation purpose and found good resemblance. It was found that overall performance of fixed bed column was significantly influenced by design and operating parameters such as bed depth (adsorbent), column diameter, initial As concentration and flow of feed. Three different values were selected for the initial concentration of As, *i.e.,* 6.67 × $10^{-6}$ mol/lit, 1.3 ×$10^{-5}$ mol/lit and 2.0 ×$10^{-5}$ mol/lit, depth of bed depth was taken as 10 cm, 20 cm and 30 cm. The feed flowrate was varied in the range of from 0.05 lit/min to 0.15 lit/min with step-size interval of 0.05 lit/min. The obtained results suggested that, for a given condition, increasing bed depth and initial concentration of as increased the removal efficiency, whereas the opposite effect of feed flowrate was obtained. Compared with iron ore, the M-PAN fiber exhibited maximum efficiency in terms of bed service time.**

**Keywords:** Arsenic, Iron ore, modified PAN fiber, Adsorption.

## I. INTRODUCTION

Likewise the other developing countries, disposal of domestic contaminated water and industrial waste water has been a major concern in Pakistan [10]. Water contamination due to presence of heavy metals is one of the key environmental issue in the developing world, since, a low concentration of such metal can lead the strong toxicity [23]. Arsenic (As) is also a hazardous element which is widely found in the groundwater in two forms as As(III) and As(V) [6, 22, 30]. The presence of arsenic in earth's crust is about 00.00021%. The use of As contaminated water for drinking purposes leads severe impacts on human health directly and indirectly (when it is used for irrigation or cattle breeding purposes) [31]. Furthermore, the discharge of wastewater in freshwater bodies results in an increasing concentration of water pollution [25]. Mostly, the presence of As in underground water is resulted by activities of arsenoparite rocks and sand soils, in which the minerals are released. Which also contains traceable concentration of As when water is taken out by tube wells and borings. According to the standards of World Health Organization (WHO) and US Environmental Protection Agency (EPA), the safe limit of As in water is considered to be 10 ppb [15]. Groundwater reservoirs in Pakistan especially in the province of Sindh are highly enriched with As concentration, thus the local community has been severely affected by several diseases. Moreover, in many localities, groundwater is the major source for household use and agricultural purpose. Therefore, removal of As from groundwater has become a major concern.

Over a past two decades, development of a cost effective technologies has been a major concern with the purpose of As removal from water systems. For removal of arsenic from surface water many physicochemical methods have been employed, *i.e.,* adsorption, coagulation, reverse osmosis, lime softening and precipitation. Adsorption is popular and most efficient [1, 17] among said technologies and mainly used due to simplicity in operation, cost effectiveness, environment friendly and presence of numerous adsorbents (naturally and synthetically) [28]. In a previous study, it was observed that an increase in specific surface area of adsorbent increased the performance of adsorption [9].

To date, a variety of adsorbents have been used with purpose of As removal such as Activated alumina [14, 26], Iron Oxide coated sand [12, 29], Activated carbon [8, 11], Granular $Fe(OH)_3$ [19], hybrid adsorbent embedding iron-manganese sludge [19] and so on. Moreover, low-cost naturally occurring adsorbent are efficient to remove arsenic from laterite available in soil was found to have good arsenic removal capacity for both As (III) and As(V). As reported in past, the laterite soil has exhibited the maximum adsorption capacity for As(III) which was 1.38 g/Kg in a batch process which is comparable to activated alumina (1.41 g/Kg). Arsenic removal capacity of laterite soil can further be improved by its modification. Memon *et al.*, [16] used chemically activated hematite iron ore and investigated experimentally the effect of various parameter on As removal performance. Natural and indigenous iron ores have also been used as economical and promising adsorbent for the removal of As. Also, their advantageous In a study of our group, the iron ores were found more effective to reduce the As content of different water sources [3]. Khan and Imteaz [12] investigated the As adsorption characteristics of different naturally available sand adsorbents and found that a remarkable decrease in As concentration was achieved as that by commercially available adsorbents. For the prediction of the wave front of the adsorption shape or the column breakthrough, the fixed bed column was considered as an important one as determined for the span of operational life and regeneration time of the bed. At present, fixed bed adsorption technique for As removal has received a reasonable attention [7] and several studies have been reported to generate valuable data for design of industrial applications [5]. The key parameters which control the overall performance of adsorption are temperature, pH, inlet concentration of As, bed height, flow rate and *etc.* For an effective adsorption process, it is essential to incorporate the effect of all these parameters. Since the experimental investigations are quite time and efforts taking, however, in this regards, numerical study could be a good choice, and the obtained results can be used as key guideline for scaling up any process. Previously, a number of studies have been performed numerically on As removal. Rodríguez-Romero *et al.*, [24] employed artificial neural network-based adsorption models to simulate the As adsorption processes. Pal *et al.,* [18] developed the dynamic mathematical model to study the removal of As from drinking water by chemical coagulation-precipitation and validated experimentally. Process simulation and modeling using Aspen Adsim software can also open a new horizon, and it has been rarely reported for As removal processes.

In the context of above discussion, this work presents experimental and numerical investigation on As removal performance of the different adsorbents (including the naturally available indigenous iron ore and modified Polyacrylonitrile fiber (M-PAN)) from the contaminated water. The process simulation was performed using Aspen Adsim software. Simulation results were compared with those experimental measurements obtained at lab scale. Effects of various operating parameters, such as initial arsenic concentrations, feed flowrates, and bed height were investigated on the performance of the adsorption column. Taking into account the economical aspect of process, the major significance of this work is to employ a natural and indigenous iron ore and compare its performance with modified PAN fiber. Moreover, the results of process simulation & modeling through Aspen Adsim software will provide useful guideline in order to scale-up the process at industrial scale.

## II. MATERIALS AND METHODS

### *A. Simulation and column specification*

As pen ONE engineering suite is widely used on industrial scale for the process simulation purposes. Aspen Adsim® is one important application of Aspen ONE engineering suite which covers all adsorption related simulations. During process simulation input data was chosen on the removal of arsenic by using two different types of adsorbents i.e. iron ore and M-PAN fiber. The model of the adsorption column used for simulation study is shown in Fig. 1. Feed stream was introduced from the bottom of column. The flow was assumed as ideal flow, while contaminated water having arsenic contamination was introduced from the bottom of the adsorption column. The temperature was also assumed constant and maintained at 30℃ in the present simulation. The diameter of the column was 2 cm whereas the bed height of the column was 10 cm. Biased Upwind Differencing Scheme (BUDS) was selected with 50.0 numbers of the nodes for the numerical computations.

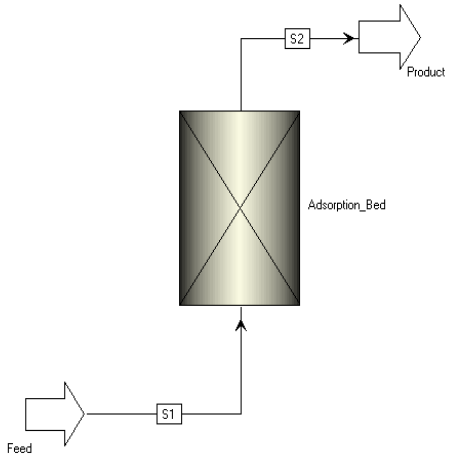


**Fig. 1.** Aspen AdsimV10 model for fixed-bed adsorption As.

### *B. General assumptions*

In process simulation, the pressure drop of the liquid phase was assumed to be constant, a plug flow was taken into account and deal mixing occurring in the liquid phase was considered. Finally, a lumped mass transfer rate (LLR) was applied with liquid or solid film resistances.

### *C. Model Equations*

**Model assumption (liquid)**

LLR was selected as a linear function for components of liquid phase concentration and solid-phase loading as given below.

$$\rho_s \frac{\partial w_i}{\partial t} = MTC_s\left(c_i - c_i^*\right) \qquad (1)$$

### *D. Material balance*

The equation of material balance equation for liquid-phase adsorption is given as,

$$-\varepsilon_i E_i \frac{\partial^2 c_i}{\partial z^2} + \frac{\partial}{\partial z}(\nu_i c_i) + \varepsilon_i \frac{\partial c_i}{\partial t} + \rho_s \frac{\partial w_i}{\partial t} = 0 \qquad (2)$$

In this equation, the first term is dispersion term, the second term indicates the convective force, the Mass transfer from the liquid to solid phase is given as,

$$\frac{\partial w_k}{\partial t} = MTC_{sk}\left(w_k^* - w_k\right) \qquad (3)$$

*E. Convection with constant dispersion*

The material balance was estimated in simulation with convection model. Constant dispersion was assumed in convection for the bed and the equations were solved with constant dispersion coefficient. The used dispersion coefficient was remained constant throughout the bed layers for all the selected species.

*F. Selection of Isotherms*

The designing of adsorber is highly dependent on the provided data of adsorptions isotherm. The separation of liquid with adsorption is driven by constant driving forces in the equilibrium process of adsorption. The actual inlet conditions or loading conditions are same as that of liquid phase compositions due to equilibrium conditions. When the feed adsorption isotherms are given for the specified components, the performance of the bed model could be predicted by the equations given by Langmuir as follows:

$$w_i = \frac{IP_{1i} IP_{2i} c_i}{1 + IP_{2i} c_i} \quad (4)$$

Where the $IP_1$ = (1/ $q_{max}$ $K_L$) and IP2=1/$q_{max}$ in which $q_{max}$ is the adsorbent capacity and $K_L$ is the equilibrium constant.

*G. Varying parameters in simulations*

The performance of adsorption column was assessed through dynamic simulations by varying important operating parameters including depth of bed, flow-rate of feed stream and feed stream arsenic concentration. For investigating the effects, two different materials were used as adsorbent materials i.e., iron ore and modified Polyacrylonitrile (M-PAN) fiber. The information of all simulated cases is given in Table 1.

**Table 1: Design matrix of Simulation cases.**

| Case no. | Initial concentration mol/lit | Feed flow lit/min | Bed height Cm | Adsorbent used |
|---|---|---|---|---|
| 1-3 * | $6.67 \times 10^{-6}$ (500 ppb) | 0.05 | 10, 20, 30 | Iron Ore |
| 4-6 * | | 0.10 | | |
| 7-9 * | | 0.15 | | |
| 10-12 | $1.3 \times 10^{-5}$ (1000 ppb) | 0.05 | 10, 20, 30 | |
| 13-15 | | 0.10 | | |
| 16-18 | | 0.15 | | |
| 19-21 | $2.0 \times 10^{-5}$ (1500 ppb) | 0.05 | 10, 20, 30 | |
| 22-24 | | 0.10 | | |
| 25-27 | | 0.15 | | |
| 28-30 | $6.67 \times 10^{-6}$,<br>$1.3 \times 10^{-5}$,<br>$2.0 \times 10^{-5}$ | 0.05 | 20 | M-PAN Fiber |
| 31-33 | $6.67 \times 10^{-6}$,<br>$1.3 \times 10^{-5}$,<br>$2.0 \times 10^{-5}$ | 0.10 | 20 | |
| 34-36 | $6.67 \times 10^{-6}$,<br>$1.3 \times 10^{-5}$,<br>$2.0 \times 10^{-5}$ | 0.15 | 20 | |

**Note:** * The cases 1 to 9 (highlighted) are also experimentally investigated and then results are compared for validation.

*H. Experimental investigations*

A limited number of experiments was conducted to investigate As removal performance and the breakthrough curves were obtained under the similar conditions as described in case 1-9 (Table 1). These measurements were used to validate the model. The experimental setup (depicted in Fig. 2) consisted of a small column filled with the adsorbent (iron ore) to the height of 10 cm (which is known as bed height). A solution with As content of 500 ppb ($6.67 \times 10^{-6}$ mol/lit) was synthesized and used a stock solution. The prepared solution was introduced into column from the top under the condition of controlled flow rate using a parasitic pump.

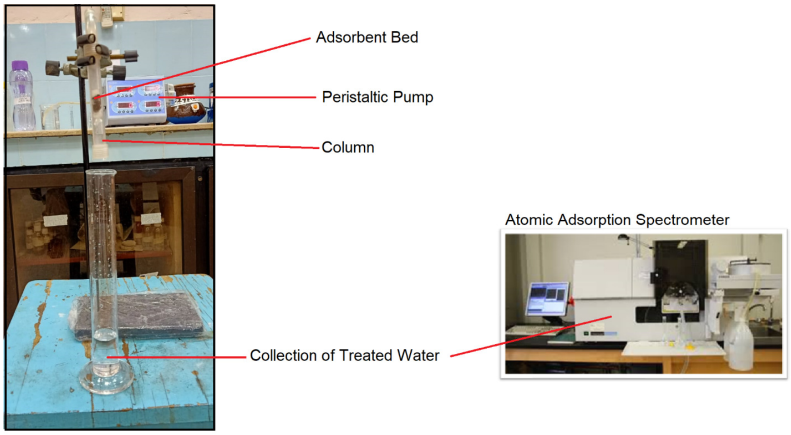


**Fig. 2.** Experimental Setup for column study.

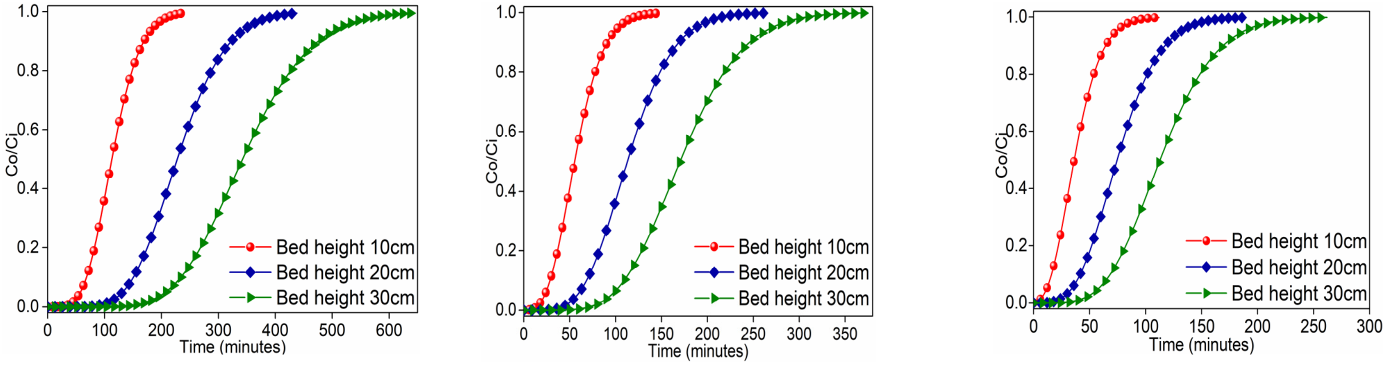
At the time interval of 10 mins, the samples were obtained periodically and through the use of atomic absorption spectrometer (A Analyst 700, Perkin Elmer, USA), their As content was determined. The experimental observations were recorded and used for model validation.

## III. RESULTS AND DISCUSSION

### *A. As removal against different bed height*

The adsorption efficiencies are usually shown through the breakthrough curves, which is the ratio of the outlet concentrations of adsorbed material to the inlet concentrations, as described through (Co/Ci). The effect of bed height on arsenic removal using iron ore as adsorbent are shown with fixed inlet concentration of arsenic ($6.67\times10^{-6}$ mol/L) in Figs. 3(a-i) at different bed depths of adsorbent (10 cm, 20 cm, and 30 cm) with various feed flow rates (0.05 lit/min, 0.10 lit/min and 0.15 lit/min).

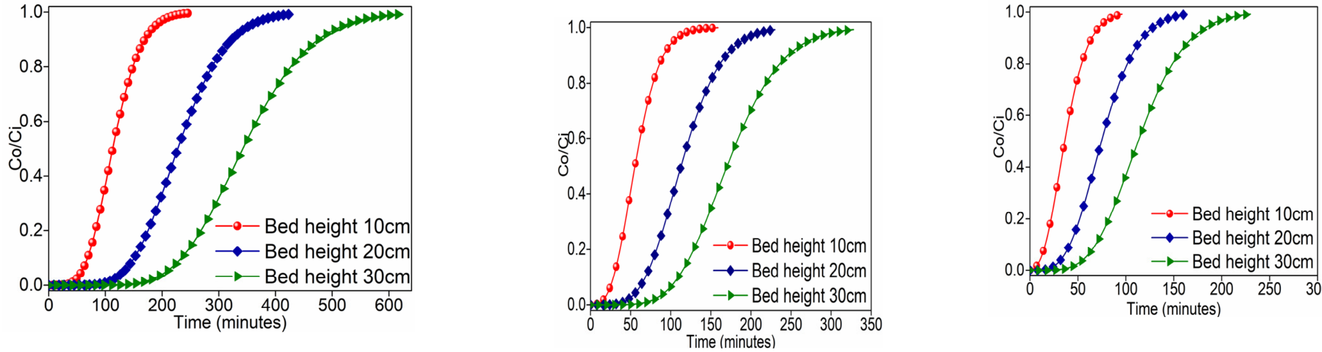
It was observed that higher bed depths require more time for the saturation of the bed. The probable reason of this effect is the increase in the binding sites availability due to increased bed depth. Moreover, with the increase of bed depth and hence increase in saturation time of bed, the adsorbent particles increased with the intra particles diffusion into the inner pores. Consequently, at 30 cm bed depth maximum time is required to saturate the bed. The depth of bed is associated with its dose, with increasing bed dose the As removal efficiency will also increase and *vice-versa*. The presently obtained trends are consistent to those obtained in past studies [13, 27]. Similar effect was observed with increased initial arsenic concentration in the feed. Increased initial concentration has more saturation efficiency of the bed.



**(a)** Feed flowrate 0.05 lit/min

**(b)** Feed flowrate 0.10 lit/min

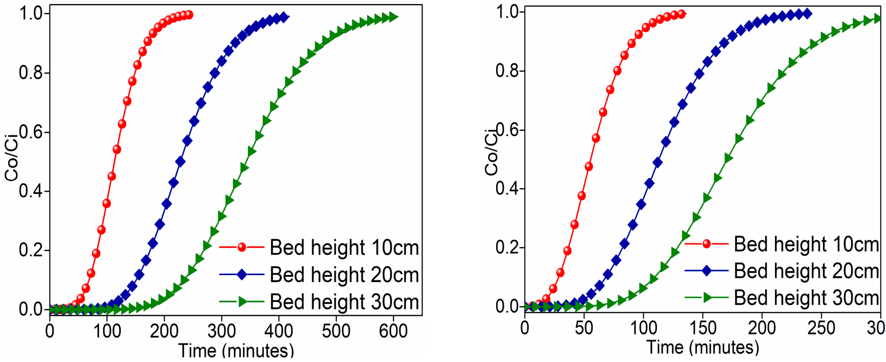
**(c)** Feed flowrate 0.15 lit/min



**(d)** Feed flowrate 0.05 lit/min

**(e)** Feed flowrate 0.10 lit/min

**(f)** Feed flowrate 0.15 lit/min



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**(g)** Feed flowrate 0.05 lit/min

**(h)** Feed flowrate 0.10 lit/min

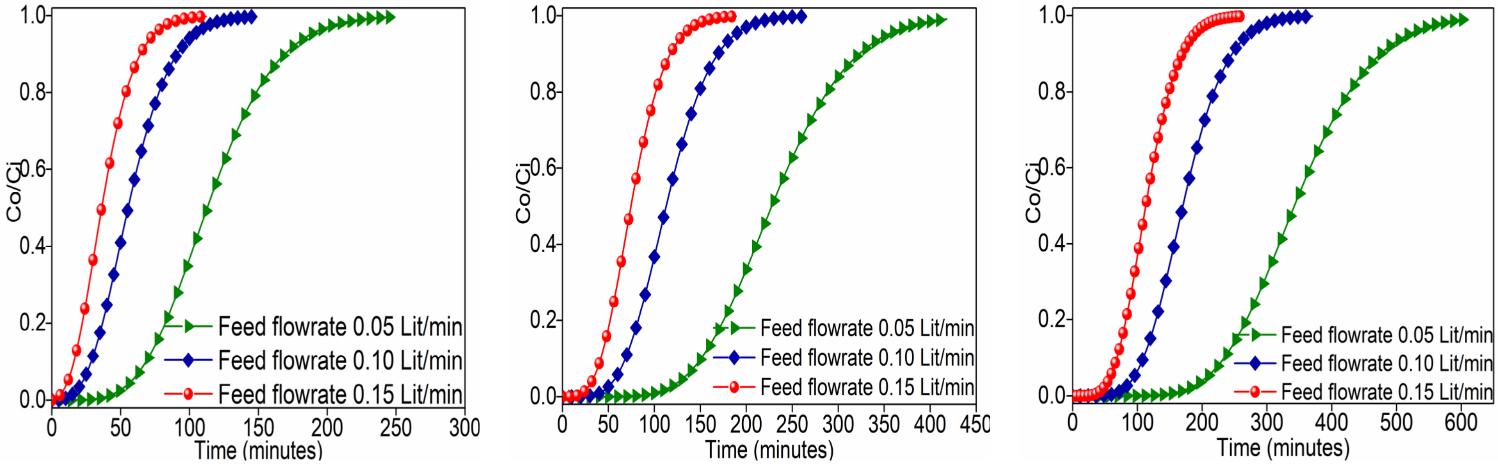
**(i)** Feed flowrate 0.15 lit/min

**Fig. 3.** Breakthrough curves against different bed heights; (a-c) @ As initial concentration of $6.67\times10^{-6}$ mol/lit, (d-f) @ As initial concentration of $1.3\times10^{-5}$ mol/lit and (g-i) @ As initial concentration $2.0\times10^{-5}$ mol/lit

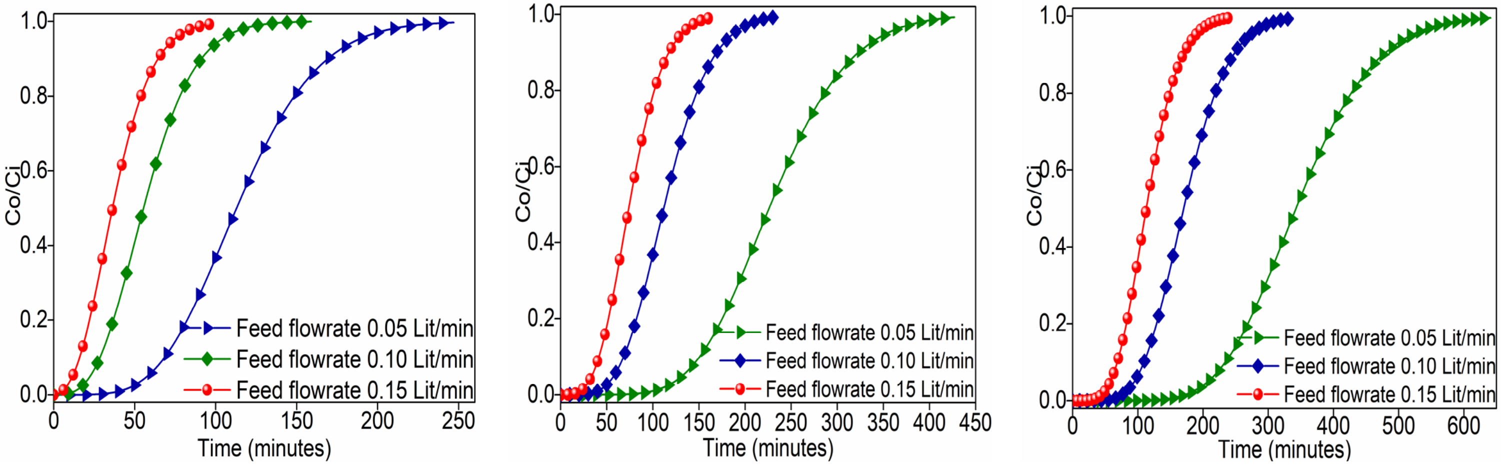
### *B. As removal at different feed flowrate*

There were inverse effects of flow rate on the saturation of bed, more the feed flowrate, lesser the time needed to saturate the bed, these results were consistent to those obtained previously [20]. Moreover, local adsorption materials and processes are also efficient for the removal of contaminants from water [20].

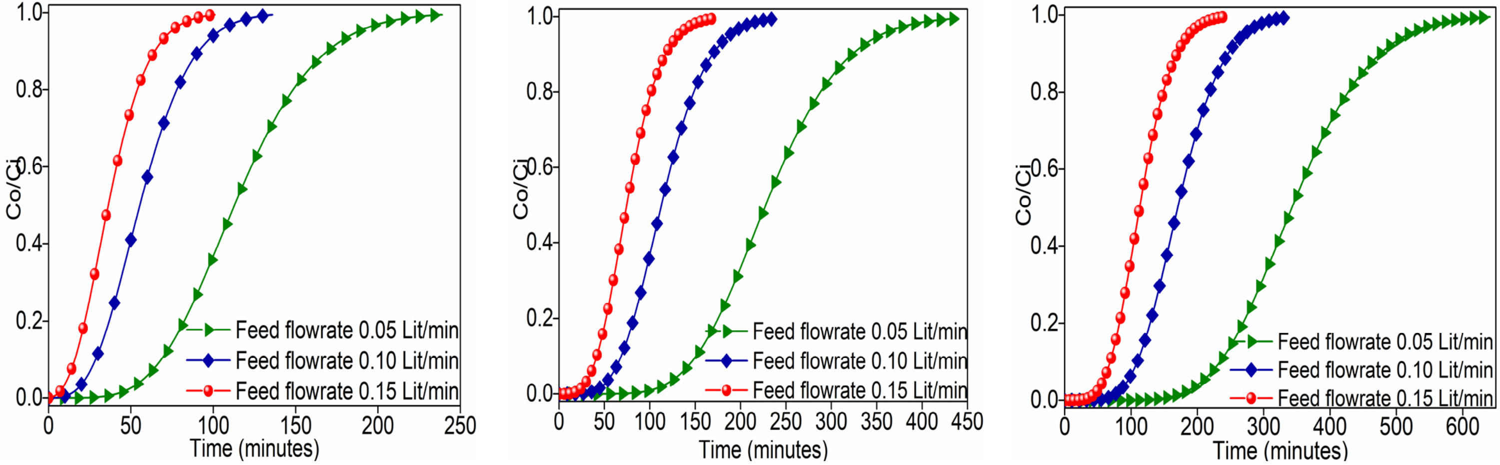
The effect of feed flowrate on As removal performance under the different conditions (bed height and initial concentration of As) is presented in terms of obtained breakthrough curves, which are plotted in Figs. 4(a-i). The obtained results demonstrated that, with increasing effluent flow rate, the breakthrough volume was decreased and *vice versa*. This could be attributed to the decreasing residence time as well as intimate contact over the bed of column. Simply, for a higher contact time, the probability of As ions to penetrate into the adsorbent pores increases.



**(a)** Bed height 10cm **(b)** Bed height 20cm **(c)** Bed height 30cm



**(d)** Bed height 10cm **(e)** Bed height 20cm **(f)** Bed height 30cm



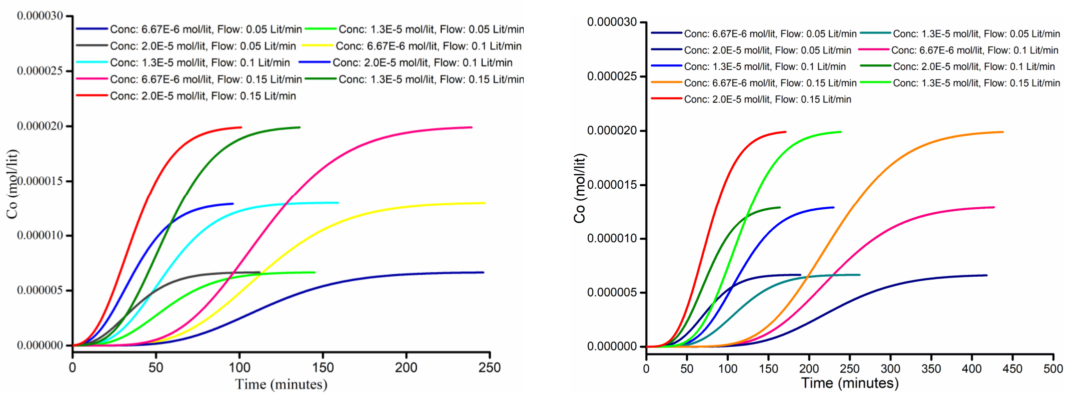
**(g)** Bed height 10cm **(h)** Bed height 20cm **(i)** Bed height 30cm

**Fig. 4.** Breakthrough curves against different feed flow rate; (a-c) @ As initial concentration of $6.67 \times 10^{-6}$ mol/lit, (d-f) @ As initial concentration of $1.3 \times 10^{-5}$ mol/lit and (g-i) @ As initial concentration $2.0 \times 10^{-5}$ mol/lit

Therefore, intra-particles diffusion is an important factor at a low feed flow rate over the entire adsorbent bed for As removal. The trend of As removal efficiency obtained against flow rate is similar to that reported by Dhoble *et al.*, [7].

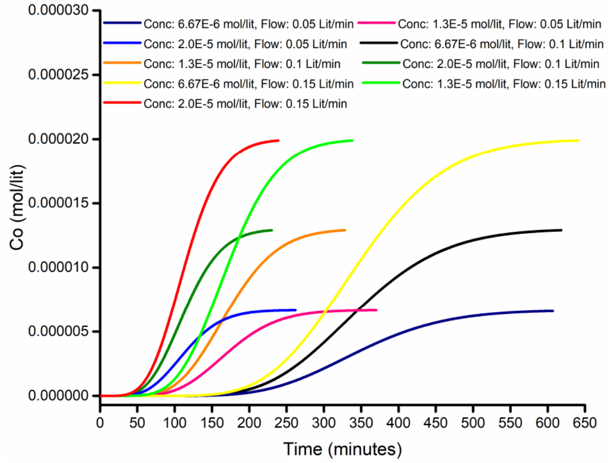
*C. As removal at different initial concentration of As*

The effect of inlet concentrations of arsenic ($6.67\times10^{-6}$ mol/lit, $1.3\times10^{-5}$ mol/lit and $2.0\times10^{-5}$mol/lit) on the outlet concentration of arsenic is shown in Figs. 5 (a-c) at 10, 20 and 30 cm bed heights with different selected flowrates (as per Table 1) using iron ore adsorbent. The results showed that, with increasing inlet As concentration, its concentration at outlet was decreased such that, at higher inlet concentration of As, its minimum concentration was obtained at outlet. This can be explained as, at higher arsenic concentration the adsorbent bed gets saturation state relatively in short time. Additionally, the results also demonstrated that for a given condition, the adsorption time was increased with increasing the height of bed. The As concentration in Azadirachta indica was observed 43.1 mg/kg by [21]. The presently obtained relationship between breakthrough volume and As initial concentration is similar with findings of Singh and Pant [27] and Asif *et al.* [2].



(a) 10cm bed height.

(b) 20cm bed height.



(c) 30cm bed height

**Fig. 5 (a-c)** Effect of As concentration feed under different feed flow rates

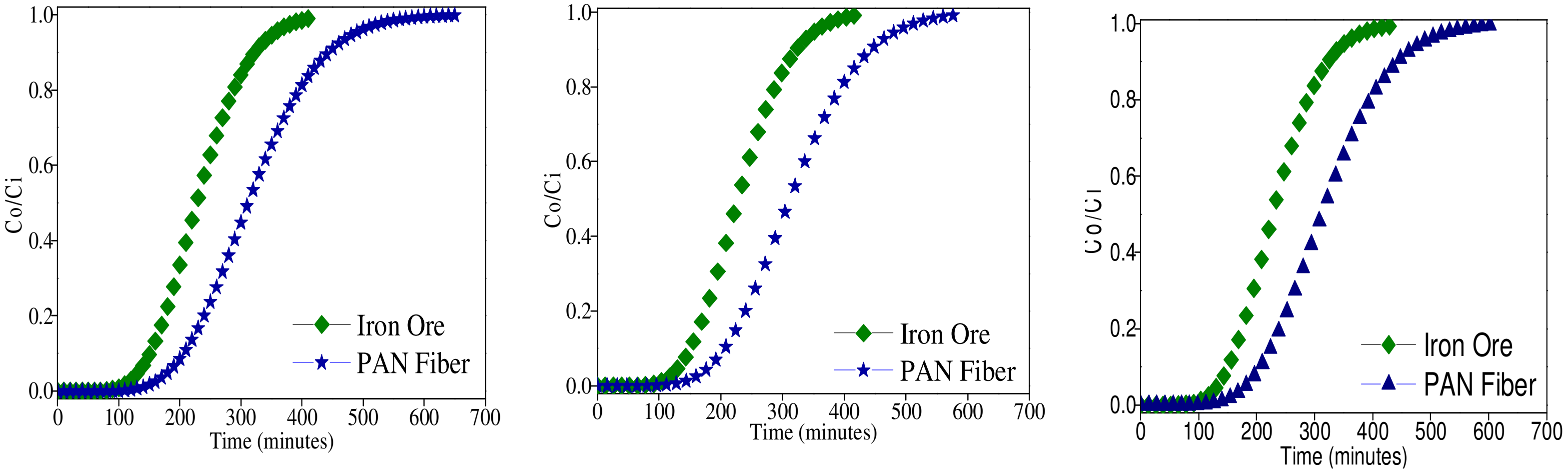
*D. As removal performance of different adsorbents*

To investigate the removal efficiency of arsenic with different adsorbent materials, few more simulations were performed with PAN fiber adsorbent. Only limited cases were simulated on selected materials as mentioned in Table 1.. As depicted in Figs. 6 (a-i) for any given condition, the M-PAN fiber exhibited the maximum As removal performance followed by iron ore. The As removal mainly depends on the few characteristics of adsorbents, such as large surface area hence more sites for chelation will be available offering higher adsorption capacity.

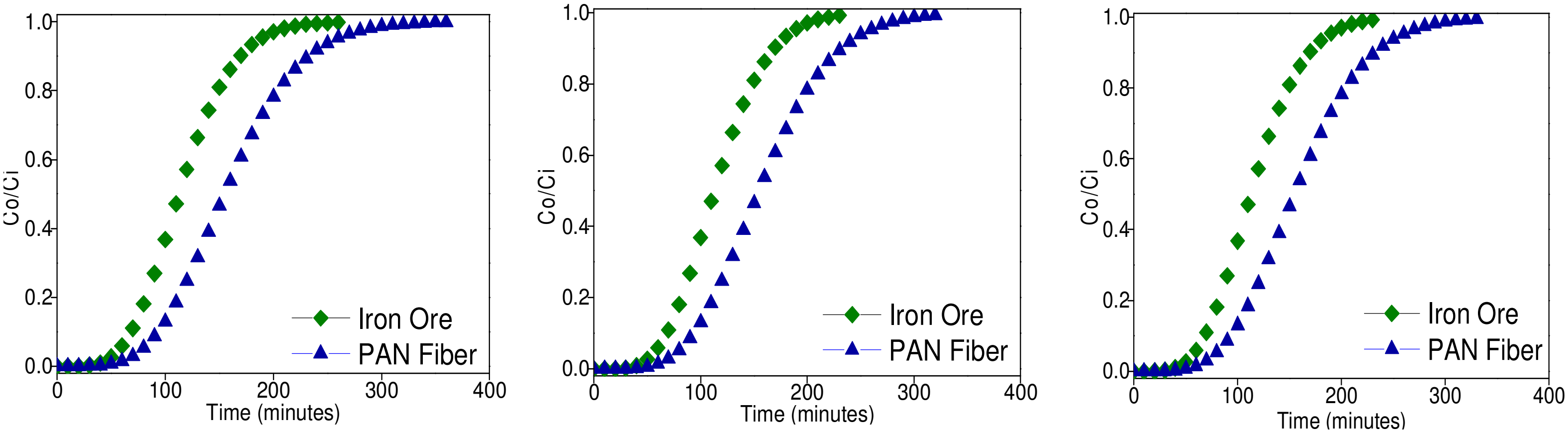
In this work, M-PAN was also used considering its modified surface with introduction of chelating groups like carboxyl, amine, imines and *etc.,* which is also reported in past [4]. Hence, the As removal performance exhibited by M-PAN was relatively superior.

## IV. MODEL VALIDATION

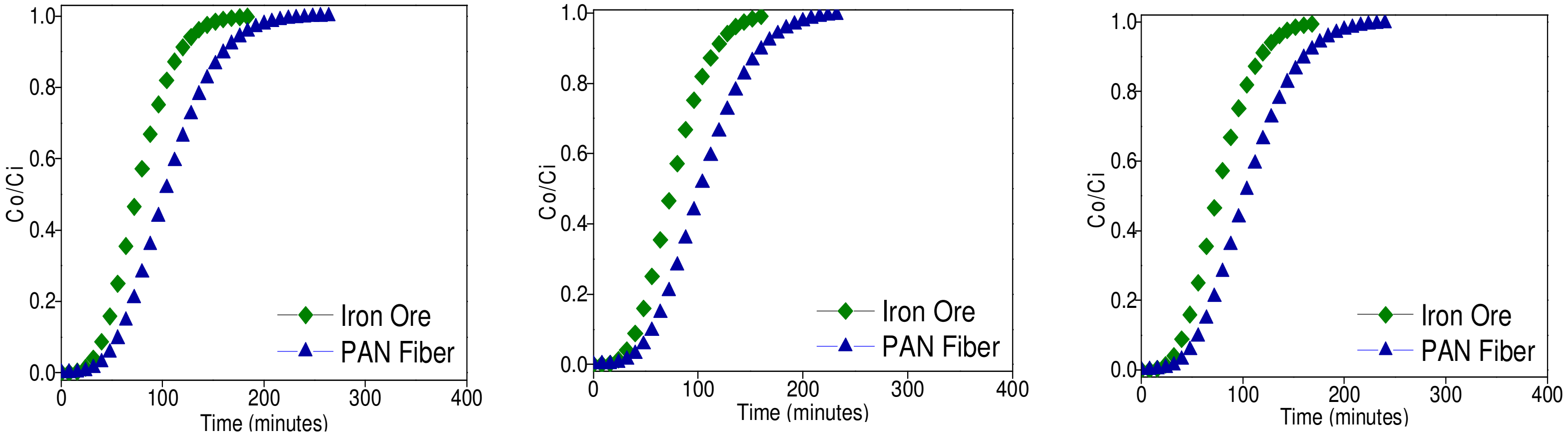
The simulation results collected in this study were compared with experimental results conducted on laboratory scale. The comparison of simulation results with experimental findings is shown in Fig. 7(a-c). The As removal efficiency was investigated experimentally for iron ore adsorbent only. Both the results were obtained in remarkable agreement with experimental results and the maximum error was less than 2%, revealing the reasonable accuracy of model.



**(a)** As concentration (6.67×10$^{-6}$ mol/lit) **(b)** As concentration (1.3×10$^{-5}$ mol/lit) **(c)** As concentration (2.0×10$^{-5}$ mol/lit)

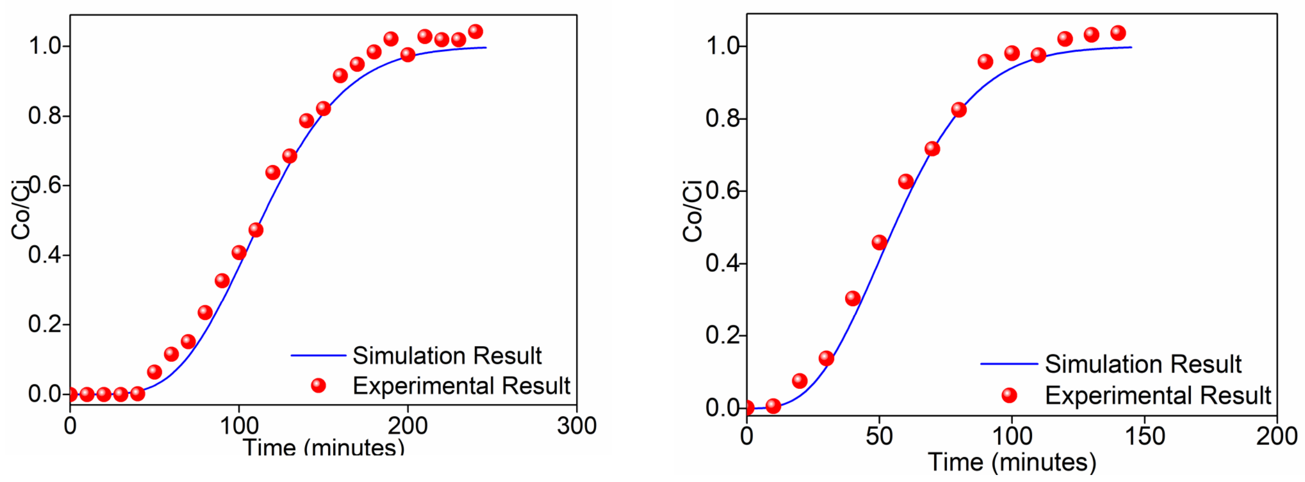


**(d)** As concentration (6.67×10$^{-6}$ mol/lit) **(e)** As concentration (1.3×10$^{-5}$ mol/lit) **(f)** As concentration (2.0×10$^{-5}$ mol/lit)



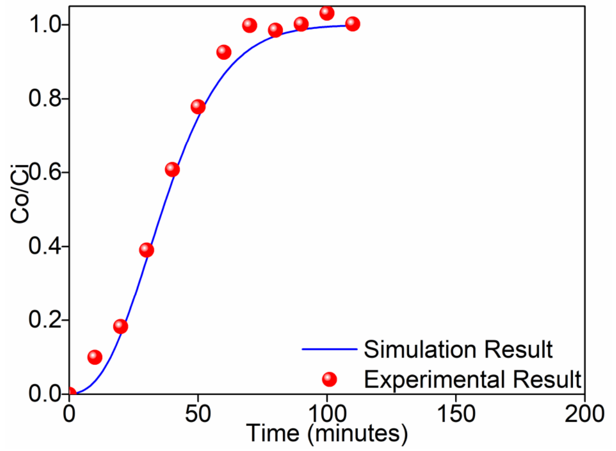
**(g)** As concentration (6.67×10$^{-6}$ mol/lit) **(h)** As concentration (1.3×10$^{-5}$ mol/lit) **(i)** As concentration (2.0×10$^{-5}$ mol/lit)

**Fig. 6.** As removal performance of different adsorbents at 20cm bed height and feed flow ; (a-c) @ feed flow of 0.05 Lit/min, (d-f) @ feed flow of 0.1 Lit/min and (g-i) @ feed flow of 0.15 Lit/min



(a) Feed flowrate 0.05 mol/lit

(b) Feed flowrate 0.10 mol/lit



**(c)** Feed flowrate 0.15 mol/lit

**Fig. 7. (a-c)** Validation of simulated results with experimental measurements for 10 cm bed height and As concentration of $6.67 \times 10^{-6}$ mol/lit (iron ore adsorbent)

## V. CONCLUSIONS

Removal of arsenic (As) from aqueous solution was modeled using Aspen Adsim® at 25℃. A fixed bed column was employed and As removal efficiency two different adsorbents, iron ore and M-PAN fiber was compared. Effect of key parameters such as, bed height, flow rate, initial As concentration was studied on As removal performance. The results were presented in terms of breakthrough curves. Following conclusions were drawn this study.

- For a given condition, the As removal efficiency was increased with increasing height of bed. For bed height of 30 cm, and the adsorption time of 150 minutes, As was almost completely removed when initial concentration was $6.67 \times 10^{-6}$ mol/lit.
- Flow rate led a negative effect on As removal efficiency.
- Compared with other adsorbents, the maximum removal performance was given by M-PAN fiber.
- For the model validation, few experiments were conducted, wherein, the conditions were, initial As concentration; $6.67 \times 10^{-6}$ mol/lit, bed height; 10 cm and feed flow rate; 0.05 lit/min, 0.10 lit/min, and 0.15 lit/min. As a result a remarkable validation of model was achieved.

The results obtained in this study will provide a generalized guideline to scale-up the As removal processes at industrial scale. The future recommendations of this study include the experimental and numerical investigation of As removal performance employing a broad variety of indigenous and cheaper adsorbents.

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**Conflict of Interest:** Authors do not have any conflict of interest to declare.

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