Biosorptive uptake of arsenic(V) by steam activated carbon from mung bean husk: equilibrium, kinetics, thermodynamics and modeling

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Abstract The present investigation emphasizes on the biosorptive removal of toxic pentavalent arsenic from water using steam activated carbon prepared from mung bean husk (SAC-MBH). Characterization of the synthesized sorbent was done using different instrumental techniques, i.e., SEM, BET and point of zero charge. Sorptive uptake of As(V) over steam activated MBH as a function of pH (3–9), agitation speed (40–200 rpm), dosage (50–1000 mg) and temperature (298–313 K) was studied by batch process at arsenic concentration of 2 mg L⁻¹. Lower pH increases the arsenic removal over the pH range of 3–9. Among three adsorption isotherm models examined, Langmuir model was observed to show superior results over Freundlich model. The mean sorption energy (E) estimated by Dubinin–Radushkevich model suggested that the process of adsorption was chemisorption. Thermodynamic parameters confer that the sorption process was spontaneous, exothermic and feasible in nature. The pseudo-second-order rate kinetics of arsenic gave better correlation coefficients as compared to pseudo-first-order kinetics equation. Three process parameters, viz. adsorbent dosage, agitation speed and pH were opted for optimizing As(V) elimination using central composite design matrix of response surface methodology (RSM). The identical design setup was used for artificial neural network (ANN) for comparing its prediction capability with RSM towards

As(V) removal. Maximum arsenic removal was observed to be 98.75% at sorbent dosage 0.75 gm L⁻¹, pH 3.0, agitation speed 160 rpm and temperature 308 K. The study concluded that SAC-MBH could be a competent adsorbent for As(V) removal and ANN model was better in arsenic removal predictability results than RSM model.

Keywords Biosorption · Arsenic · Mung bean husk · Thermodynamic parameters · RSM · ANN

Introduction

The presence of Arsenic (As, Z = 33) in terms of most plentiful element in the human body, seawater and earth's crust is 12th, 14th and 20th position, respectively (Mandal and Suzuki 2002). As an ultra-trace nutrient, arsenic deficiency leads to inhibited growth, although beyond its necessary level it causes toxic effect on plants, animals, and human beings. Inorganic arsenics are proven to be carcinogens (Ng 2005). A worldwide recognized problem related to arsenic contamination in drinking water connected with 21 countries; the largest population being risked are Bangladesh followed by West Bengal in India (Jiang et al. 2012). World Health Organization (WHO) recommended that 10 µg L⁻¹ is the limit for utmost permissible intensity of arsenic in drinking water. Geological source such as natural reaction and volcanic emissions are the main causes of arsenic pollution where other factors such as the burning of fossil fuels, mining, smelting, and pesticide use by human activities are also responsible for arsenic toxicity (Bissen and Frimmel 2003).

Till date, several methods have been attempted such as coagulation/precipitation (Song et al. 2006), coagulation/filtration (Wickramasinghe et al. 2004), adsorption

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(Asadullah et al. 2014), oxidation (Borho and Wilderer 1996), ion exchange (Urbano et al. 2012), membrane/reverse osmosis (Fogarassy et al. 2009), and electro-coagulation (Hansen et al. 2007) to eliminate arsenic from water. Among these technologies, adsorption is preferably a good choice due to its operational easiness, cost effectiveness, eco-friendly nature and accessibility of various types of adsorbent (Ranjan et al. 2009).

For the last couple of decades, activated carbons as an adsorbent has been widely used for arsenic removal from water (Kalderis et al. 2008; Mohan and Pittman 2007; Mondal et al. 2008) as it possesses significant surface area with micro/mesopore structure along with extensive adsorption capacity. Commercial activated carbons are expensive and it is the main drawback of using this adsorbent in all the application sectors (Roy et al. 2013). Recently, some of the low cost compounds such as zirconium oxide-coated marine sand, chir pine sawdust, bagasse fly ash, clay and magnetic chitosan-bamboo sawdust composites were employed while some of the biomasses were also used to remove arsenic from water (Khan and Sing 2010; 2013, 2014, Khan and Nazir 2015; Ali et al. 2014). Chemical activated carbon (linseed cake activated carbon and waste tyre activated carbon) was also used to eliminate arsenic from aqueous solutions (Khan et al. 2016, 2017). Here, in this paper we have tried to prepare a low cost agricultural residues mung bean husk as a precursor to prepare potential carbonized adsorbent that can deal with waste disposal problem as well as the possibility of recycling the waste towards its application in removal of toxic contaminant from aqueous solution (Budinova et al. 2009; Mondal et al. 2016a, c). *Vigna radiate* (Mung bean) seeds are an inhabitant from the Indian subcontinent and 90% of world's mung bean is produced in Asia, and India is the top producer of this crop (Mondal et al. 2015, 2016b).

The typical optimization of a removal process requires changing one variable while fixing the other variables at a certain level. This sort of optimization process is impractical as it requires a large number of experiments to be performed due to the possibility of different factorial combinations of the test variables (Chang et al. 2011). This sort of problem can be overcome by using appropriate optimization process like RSM and ANN.

The aims of the present study includes: (1) synthesis of steam activated mung bean carbon as an adsorbent (2) characterization of adsorbent material, (3) investigation of the effects of process parameters such as adsorbate solution pH, temperature, adsorbent dosage and speed of agitation on the adsorption of As(V) onto adsorbents, (4) Isotherm, kinetics and thermodynamics of the adsorption process and (5) finding an optimum condition of adsorption process using RSM and ANN and their comparison.

Materials and methods

Preparation of adsorbate solution

Sodium arsenate dibasic heptahydrate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) as arsenic sourced compound was purchased from Fisher scientific (USA) and was dissolved in deionized water (18.2 M Ω) to prepare adsorbate solution. Arsenic stock solution of 3 ppm equivalent was prepared and further diluted to the desired concentration with deionised water (Millipore Corp., USA). The adsorbate pH was adjusted using 0.1 N NaOH and 0.1 N HCl.

Preparation of sorbents and its characterization

The precursor mung bean husk used for preparation of activated carbon was purchased from Durgapur (India) local market. Distilled water was used to wash MBH several times followed by drying to obtain the desired precursor. In the first step of operation in a stainless steel cylindrical shelled furnace chamber, the precursor was gradually carbonized with the rate of heating of 55 °C per 15 min at 550 °C for 1 h. After carbonization, temperature was heated up to 650 °C into the furnace chamber and superheated steam was introduced at a pressure of 1.5–2.0 kg cm⁻² for 1 h for activation of carbon (Mondal et al. 2015). The prepared activated carbon was then placed to desiccators to cool down to room temperature. Characterization of the sorbent was done properly using different instrumental techniques such as SEM, BET, FTIR and point of zero charge.

Batch mode adsorption studies

The appropriate quantity of mung bean husk derived activated carbon for the adsorption of arsenic was estimated by the batch equilibrium study. Arsenic concentration of 2 mg L⁻¹ was taken in a fixed volume (100 ml) of liquid sample in all the tested batch sorption experiments in an incubator shaker (Model Innova 42, New Brunswick Scientific, Canada) at 310 K. Process optimization was done by altering the dosage (50 mg–1 g), pH (3–9) and agitation speed (40–180) for a preset contact time interval. The concentration of arsenic in a flask was measured by taking adsorbate solution at a specific time interval. Arsenic solution from flask was taken at a specific time interval and then centrifuged at 4000 rpm for 20 min. Supernatant solution collected at different time interval was analyzed for the presence of arsenic using arsenic kit (Merck, USA). Each batch experiment was conducted in triplicate and for data analysis purposes mean values were used. At equilibrium q_e (mg g⁻¹) conditions, the quantity of arsenic

compound adsorbed can be estimated according to the mass balance from the initial and final equilibrium concentrations by means of the following equation:

$$q_e = \frac{(C_i - C_e)V}{m}, \quad (1)$$

where C_i is the initial arsenic concentration (mg L^{-1}), C_e is the equilibrium arsenic concentration (mg L^{-1}), V is the volume of the arsenic solution (L) in contact with the adsorbent, m is the mass of the adsorbent (g).

The percent removal of arsenic was calculated using the following relation:

$$\text{Sorption}(\%) = \frac{(C_i - C_e)}{C_i} \times 100. \quad (2)$$

Control experiments were conducted in each batch study to confirm that arsenic was not adsorbed by the flask wall.

Models for kinetic and equilibrium studies

The evaluation of the adsorption kinetics was tested by subsequent models, which are pseudo-first order (Kumar et al. 2003), pseudo-second order (Ho et al. 2000) and Elovich (Chien and Clayton 1980).

The assessment of the equilibrium adsorption isotherm was checked by different models. These are: Langmuir (Mohan et al. 2011a), Freundlich (Mohan et al. 2011a), Temkin and Dubinin–Radushkevich (Dubinin 1960).

Removal process modeling

The optimization of As(V) elimination from water was done through RSM by the same process variables used in batch mode, viz. adsorbent dosage, pH and agitation speed. Concentration of As(V) was taken 2 mg L^{-1} in this modeling process. CCD was chosen from RSM to form experimental matrix to build up a suitable model for explaining the process of arsenic removal. Table 1 shows the variables range and levels used in this experiment. This modeling process had taken removal percentage of arsenic as the response of the system.

Quadratic equation to correlate response function and variables process can be mathematically done by the following equation:

$$x_j \quad \gamma = \beta_0 + \sum_{i=1}^k \beta_i x_i + \sum_{i=1}^k \beta_{ii} x_i^2 + \sum_{i=1}^k \sum_{j=1+1}^k \beta_{ij} x_i x_j + \varepsilon, \quad (3)$$

where β_0 is the constant coefficient, β_i , β_{ii} and β_{ij} are linear, quadratic and interaction coefficients effect, respectively, x_i , x_j are independent variables and ε is the error.

In CCD, three-dimensional response plots were generated that explain the interactive effect of independent variables with their corresponding effect.

Artificial neural network model works in a very complicated manner like the neurons of the nervous system of human to make a refined optimal decision based on the data or stimuli. Currently for engineering application, so many ANN models have been developed such as feed forward, multilayer perception (MLP), and radial basis function. The present investigation studied As(V) removal modeling using feed-forward three-layer back propagation algorithm with a linear transfer function.

ANN model essentially consists of three layers, i.e., input layers, hidden layers and an output layer. The independent variables are input and hidden layers, while dependent variables are output layer. ANN model is structured in such a fashion that all layers are interconnected by a processing unit called neuron, while the neurons are connected to each other by links (Fig. 1). Input layer takes the information from outer source and pass the information to the next layer (hidden) for processing of data. Input values were weighted individually before it passes through the hidden layer. The hidden layer processes the data and analyzes to produce the output based on the sum of the weighted input values (Khataee and Khani 2009). The mathematical equation to denote the net input Y_{ij} of the neuron j in the layer i is represented by

$$Y_{ij} = \sum_{k=1}^{n_{i-1}} W_{ijk} V_{i-1,k} + \theta_{ij}, \quad (4)$$

$$V_{ij} = g(Y_{ij}), \quad (5)$$

where V_{ik} is the input of neuron, W_{ijk} is the weight of connection and θ_{ij} is the bias of neuron. The neuron output V_{ij} is estimated using an activation function $g(Y_{ij})$. Equations (4) and (5) can be used to model nonlinear relationships between a set of input and output variables. It has to be noted that the training is performed using the input as well as output data taken from the system under study.

In this present study, same input, viz. adsorbent dosage, pH and agitation was taken from RSM to model the ANN process. CCD matrix generated from RSM along with the output results was used in ANN modeling to train output and input layer. Matlab (version 7.12) toolbox of Neural Network was used to calculate the ANN data.

Results and discussion

Characterization of activated carbon

The BET-specific surface area and micro-pore volume were estimated to be $405 \text{ m}^2 \text{ g}^{-1}$ and $0.2853 \text{ cm}^3 \text{ g}^{-1}$,

Table 1 CCD with experimental and predicted values

Variable	Unit	Symbols	Range					Observed	RSM	ANN
			$-\alpha$	-1	0	1	$+\alpha$			
pH		<i>A</i>	1.295	3	5.5	8	9.704			
Agitation speed	rpm	<i>B</i>	45.683	90	155	220	264.31			
Dosage	mg	<i>C</i>	127.28	400	800	1200	1472.72			
	<i>A</i>	<i>B</i>	<i>C</i>	<i>A</i>	<i>B</i>	<i>C</i>				
1	0	0	0	5.5	155	800	95	94.85	94.79	
2	0	0	$+\alpha$	5.5	155	1472.71	87.5	86.76	87.51	
3	0	0	0	5.5	155	800	95	94.85	94.795	
4	0	0	0	5.5	155	800	94	94.85	94.795	
5	-1	1	1	3	220	1200	91	91.58	90.97	
6	$+\alpha$	0	0	9.70	155	800	87.5	87.65	87.749	
7	1	1	-1	8	220	400	84	83.84	83.974	
8	0	0	0	5.5	155	800	95	94.85	94.795	
9	1	-1	1	8	90	1200	87	86.98	86.981	
10	$-\alpha$	0	0	1.29	155	800	95	94.2	94.999	
11	0	0	0	5.5	155	800	95	94.85	94.795	
12	-1	1	-1	3	220	400	90	90.48	90.019	
13	1	1	1	8	220	1200	87.5	87.93	87.504	
14	0	$-\alpha$	0	5.5	45.68	800	87.5	87.41	87.485	
15	-1	-1	1	3	90	1200	87.5	88.13	87.512	
16	0	0	0	5.5	155	800	95	94.85	94.795	
17	-1	-1	-1	3	90	400	89	89.03	89.098	
18	0	0	$-\alpha$	5.5	155	127.28	84	84.08	83.997	
19	1	-1	-1	8	90	400	85	84.88	84.907	
20	0	$+\alpha$	0	5.5	264.31	800	90	89.43	90.004	

respectively. Adsorbents surface charge can be estimated by the help of point of zero charge (pH_{pzc}) and experimentally it was observed to be 8.6. FTIR spectrum of SAC-MBH and SAC-MBH along with adsorbed As(V) molecule was shown in Fig. 2a, b, respectively. The process of adsorption has been influenced by the presence of functional groups such as carboxyl, hydroxyl, sulfate, and amino groups on the surface of an adsorbent. The SAC-MBH exhibited different bands at 770 cm^{-1} (C–H bend), 1068 cm^{-1} (C–N stretch), 1215 cm^{-1} (C–H bend) and 1384 cm^{-1} (nitro group). Sifting of some bands such as $770.39\text{--}769.25\text{ cm}^{-1}$, $1637.6\text{--}1629.22\text{ cm}^{-1}$ and $3393.85\text{--}3390.59\text{ cm}^{-1}$ suggests that C–H bend, N–H bend stretch and N–H stretch may be participating for the adsorption of arsenic onto SAC-MBH.

One of the vital parameters in the process of adsorption is specific surface area of adsorbent, and pore size is helpful to know adsorbates binding chances with the functional group into the activated carbon. The surface structure of the raw activated carbon was found to be porous, irregular and uneven shaped where porous

structure was highly heterogeneous in nature associating with high surface area in SEM micrograph as depicted in Fig. 3a. It was shown from the Fig. 3b that the pores number was reduced, and the heterogeneous structure helps to capture the As(V) molecule inside the activated carbon surface to facilitate absorption. The surface texture became smooth due to filling up of the pores by As(V) molecule adsorption. The point of zero charge of the adsorbent was estimated to be 8.6 that point out the ‘H’ category of carbon. H category carbons have a lesser density of oxygen-containing functional group, more hydrophobic and showing strong acid adsorption capacity (Mohan et al. 2011b).

Effect of adsorbent dosage

Experimental results suggest that the percentage removal of the As(V) species increased (Fig. 4a) with increased dosages of adsorbent (0.5–0.75 g). Increase in adsorbent dosage means the availability of more surface area which, in turn, indicates the presence of more active binding sites

Fig. 1 Topology of the ANN architecture

to adsorb the adsorbate molecules (Lewinsky 2007). Further increase in dosages beyond 0.75 g mass did not cause any significant increment in the elimination tendency of As(V). This is due to the adsorption of almost all As(V) onto the adsorbent surface and attaining of equilibrium between the As(V) adsorbed to the adsorbent and those As(V) left in the solution. The results of this study are the following similar type of pattern reported as findings by other researchers (Li et al. 2012; Roy et al. 2013). The optimum value of adsorbent dosage was 0.75 g L^{-1} and for the rest of the experiment this value was used.

Effect of solution pH

The solution pH is a significant factor for wastewater treatment process. Figure 4b presents the activated carbon behavior over the pH range of 3–9 and it is easily understood that the optimum arsenic elimination is achieved at pH 3.

The solution pH affects the activated carbon by dissociating the functional groups on the adsorbent surface active site by changing the surface charge (pHpzc) of the activated carbon. On the other hand, the solution pH affects the target pollutant by changing degree of ionization (pKa) and speciation of the arsenic. Since the pHpzc of SAC-MBH surface was 8.6, below the pHpzc value the adsorbent surface becomes positive in a solution. On the other side, As(V) has dissociation constant $\text{pK}_a = 6.94$, thereby at $\text{pH} > 6.97$ it is protonated where at $\text{pH} < 6.97$ it is

deprotonated. Thus, it can be said that at less than pH 6.97, As(V) remains in solution in anionic form (H_2AsO_4^-), so at this stage arsenate becomes negatively charged while SAC-MBH becomes positively charged, so electrostatic attraction occurs and this was most likely the adsorption mechanism (Liu et al. 2012). As the pH increases, adsorbent surface becomes less positive ($\text{pH} < \text{pHpzc}$) or more negative ($\text{pH} > \text{pHpzc}$). These two factors cumulatively decrease electrostatic attraction while increasing repulsion that leads to decrease in As(V) removal at higher pH.

Effect of agitation speed

Agitation speed is a significant parameter for As(V) removal by SAC-MBH. The adsorption study was carried out at variable agitation speed ranging from 40 to 200 rpm that has been depicted in Fig. 4c. Increasing agitation speed causes increased removal of As(V) and reached a maximum removal value at 160 rpm (95%). This optimum agitation speed confirms usability of all the surface active sites of adsorbent to uptake arsenic. It was an optimum speed of agitation to assure the availability of all the surface binding sites for arsenic uptake. During slow agitation speed, SAC-MBH did not spread in the solution properly, instead it conglomerated in the solution; thus, beneath the apex layers of adsorbent it hide many active sites so apex layer is only available for adsorption and in this process hidden layers are not present to participate in the adsorption process, since they had no proper contact with As(V).

Fig. 2 **a** Before and **b** after adsorption of As(V) onto SAC-MBH

Fig. 3 **a** Before and **b** after adsorption of As(V) onto the surface of SAB-MBH on SEM at $\times 500$ magnification

Fig. 4 Effect of **a** adsorbent dosage, **b** pH, **c** agitation speed, and **d** temperature on the adsorption of As(V) by SAB-MBH

In case of higher agitation speed, As(V) particles have not enough time to make a bond with SAC-MBH due to random collisions among the participant particle. Similar type of agitation speed pattern was also found by the other researchers (Liu et al. 2012; Shafique et al. 2012).

Effect of temperature

In general, the adsorption process is affected by the temperature. Influence of temperature on the arsenic adsorption

by the sorbent was investigated at four different temperatures (298, 303, 308 and 313 K).

As(V) removal was observed to increase from 95 to 98.75% with increase in temperature as depicted in Fig. 4d. It indicates that As(V) removal by adsorption onto SAC-MBH was encouraging at elevated temperatures. Since by further increasing the temperature the percentage removal of the adsorbate remained constant. Increasing temperature speed up the As(V) molecule while slower the retarding forces on the molecules; thereby, percentage removal of

the sorption capacity of the adsorbent had been enhanced. Percentage removal of arsenic molecule was also increased due to pore size changes and increase in kinetic energy of the adsorbents. This type of trend was found by other authors (Pokhrel and Viraraghavan 2008).

Kinetic studies

In the process of adsorption, kinetics was done to understand two important factors: one is equilibrium time and other is adsorption mechanism. To study the mechanism of arsenic adsorption, three kinetic models were fitted. The kinetic parameters of the fitted model are presented in Table 2.

Adsorption kinetics of arsenic was investigated in the present investigation using three models, namely Lagergren pseudo-first order, pseudo-second-order and Elovich equation. Fitting and evaluation of the three models into the adsorption kinetics study ultimately give the finest model to explain the adsorption mechanism at different temperature.

Theory of kinetics

Lagergren pseudo-first-order kinetics and second-order kinetics models are mathematically explain in their linear form by the following Eqs. (6) and (7), respectively:

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303} t, \quad (6)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t, \quad (7)$$

where q_e and q_t are the quantity of arsenic adsorption at an equilibrium and time t (mg g^{-1}); k_1 (min^{-1}) and k_2 ($\text{g mg}^{-1} \text{min}^{-1}$) are the pseudo first order and pseudo second order rate constants, respectively.

Table 2 Kinetic parameters for As(V) removal using activated MBH carbon

T (K)	298 K	303 K	308 K	313 K
q_{exp} (mg g^{-1})	0.253	0.26	0.253	0.263
Pseudo-second order				
K_1 (min^{-1})	0.395	0.510	1.575	1.575
q_{cal} (mg g^{-1})	0.272	0.283	0.272	0.283
R^2	0.989	0.995	0.999	0.999
Elovich equation				
a ($\text{mg g}^{-1} \text{min}^{-1}$)	0.365	0.537	305.182	305.182
b (g mg^{-1})	26.315	27.027	52.631	52.631
R^2	0.834	0.930	0.931	0.931

Experimental conditions: adsorbent dosage 0.75 g; pH 3, agitation speed 160 rpm; temperature 308 K

The linear plot of $\log(q_e - q_t)$ versus t and t/q_t versus t gives model applicability of pseudo first order and pseudo second order kinetics (Fig. 5a), respectively. The kinetic parameters k_1 (min^{-1}) and k_2 ($\text{g mg}^{-1} \text{min}^{-1}$) were obtained from the slope and intercept of the above-mentioned linear plot by applying Eqs. (4) and (5).

Elovich kinetic model assumes that the active sites present on the solid surface are heterogeneous in nature and have different activation energy for chemisorptions. Generally, this model is one of the most practical to understand chemisorptions process.

The following mathematical equation helps to understand the model

$$q_t = \frac{1}{\beta} \ln(\alpha\beta) + \frac{1}{\beta} \ln t \quad (8)$$

where α is the initial sorption rate constant ($\text{mg g}^{-1} \text{min}^{-1}$) and β is the desorption constant (g mg^{-1}). The linear plot of q_t versus $\ln t$ gives the slope and intercept, from where the value of the parameters α and β is obtained. The value of $1/\beta$ indicates the sites available for adsorption.

Prediction of best kinetic model for As(V) adsorption

The suitability of the kinetics model was judged by their correlation coefficients (R^2) which has been tabulated in Table 2. At first, the experimental equilibrium data were analyzed by pseudo-first-order equation. The experimental data were found to be very unfavorable for first-order kinetic model to all temperatures (Figure not shown). Therefore, it was not justifiable to use the model of As(V) kinetics to superheated steam activated carbon.

The experimental data were then fitted into pseudo-second-order equation. The correlation coefficients value was found to be close to unity or unity where the calculated q_e values were close to experimental q_e values at all temperature tested in this experiment. Therefore, the excellent fit of the data in the model as well as the kinetic parameters suggests that As(V) adsorption kinetics followed pseudo-second-order kinetics.

Using last experimental data, Elovich equation model was verified. The correlation coefficient did not show good linearity but the constant α and β increase with increasing temperature; thus, it can be said that both the rate of chemisorptions and availability of adsorption surface would increase.

Equilibrium studies

The equilibrium adsorptive studies were done by applying different adsorption isotherm to find the relationship between the adsorption of adsorbate by adsorbent and remaining adsorbate concentration in the solution. The

Fig. 5 **a** Pseudo-second-order kinetics, **b** Langmuir isotherm and **c** Gibbs free energy change versus temperature of As(V) onto SAB-MBH

parameters found from the equilibrium model show some adsorption mechanism insight, properties and affinity of the adsorbent surface.

The isotherm of Langmuir model provides information about the adsorptive capacity and broad equilibrium behavior. The following equation used to describe the model

$$\frac{C_e}{q_e} = \frac{C_e}{Q_m} + \frac{1}{K_L Q_m}, \quad (9)$$

where C_e is the equilibrium adsorbate concentration at liquid phase (mg L^{-1}), q_e is the equilibrium adsorbate concentration at solid phase (mg g^{-1}), Q_m is the maximum adsorption capacity (mg g^{-1}), and K_L is the adsorption equilibrium constant (L mg^{-1}).

The plot between $\frac{C_e}{q_e}$ vs C_e of Langmuir isotherm indicates the isotherm constant value of K_L and Q_m from the slope and intercept of the graph, respectively (Fig. 5b).

Freundlich equilibrium model is associated with heterogeneous surface where low to intermediate concentration ranges are functional. The following equation can satisfactorily describe the model

$$\log q_e = \log K_F + \left(\frac{1}{n}\right) \log C_e, \quad (10)$$

where K_F and n are Freundlich constants linked to adsorption capacity and adsorption intensity, respectively. These two constants can be estimated from the intercept and slope of the linear plot of $\log q_e$ vs $\log C_e$.

The indication of adsorption favorability depends on the value of n .

Temkin isotherm model signifies the heat of the adsorption and the adsorbent–adsorbate interaction. For the Temkin isotherm model, following equation is used

$$q_e = B_T \ln K_T + B_T \ln C_e, \quad (11)$$

where q_e is the quantity of As(V) adsorbed at equilibrium (mg g^{-1}), B_T is the Temkin adsorption isotherm constant or heat of adsorption, where B_T is the (RT/b) , R is the universal gas constant and T is the temperature in kelvin, K_T is the Temkin equilibrium binding constant or the maximum binding energy (L mg^{-1}), and C_e is the adsorbate concentration at equilibrium liquid phase (mg L^{-1}).

The values of B_T and K_T were calculated from the plot of q_e against $\ln C_e$.

The Dubinin–Radushkevich (D–R) model is generally expressing the adsorption mechanism. It is applied to differentiate between physical and chemical adsorption. It is expressed by the subsequent equation

$$\ln q_e = \ln q_d - 2\beta \varepsilon^2, \quad (12)$$

where q_d is the Dubinin–Radushkevich isotherm constant related to the degree of sorbate sorption by the sorbent surface (mg/g), β is the mean free energy coefficient of adsorption, $\varepsilon = \text{Polanyi potential} = RT \ln(1 + 1/C_e)$, where R is the Gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$) and T is the temperature (K).

The mean free energy of the each adsorbate molecule can be mathematically expressed by the following equation (Kundu and Gupta 2006):

$$E = \frac{1}{\sqrt{2\beta}}, \quad (13)$$

E is the mean free energy of the adsorption (E).

The comparative analysis of the four isotherm models was done on the basis of linear coefficient R^2 value and the isotherm was validated by Chi square test.

The Chi square test (χ^2) was calculated by the following equation:

$$\chi^2 = \sum \frac{(q_{\text{exp}} - q_{\text{mea}})^2}{q_{\text{mea}}} \quad (14)$$

where q_{exp} is the experimental amount of arsenic concentration at equilibrium and q_{mea} is the measured or calculated amount of arsenic concentration at equilibrium.

Table 3 shows the diverse isotherm parameter with their corresponding values and from the Langmuir isotherm it was seen that the R^2 value is close to unity or unity as compared to the other three isotherm models. The R^2 value indicates the fit of experimental data into the liner isotherm equation; the higher the R^2 value the better is the goodness of fit.

Table 3 Isotherm parameters for As(V) removal using activated MBH carbon

	Values
Langmuir	
Q_{max} (mg g^{-1})	0.36
K_L (L mg^{-1})	0.3385
R_L	0.5963
R^2	0.999
χ^2	0.016
Freundlich	
K_F ($\text{mg g}^{-1}(\text{mg L}^{-1})^{-1/n}$)	0.7378
n_F	3.2679
R^2	0.723
χ^2	5.702
Temkin	
B_T	0.054
K_T (L mg^{-1})	1474.83
R^2	0.839
χ^2	0.043
D–R model	
q (mg g^{-1})	0.345
β	1.11×10^{-3}
E (kJ mol^{-1})	21.218
R^2	0.984
χ^2	0.378

Experimental conditions: adsorbent dosage 0.75 g; pH 3, agitation speed 160 rpm; temperature 308 K

The error factor when considering, it is shown that low χ^2 value in Langmuir model has better represented the equilibrium data into the equation. An outstanding fit of the adsorption data into the Langmuir isotherm model established that the adsorption is monolayer where each molecule of adsorption had identical activation energy and negligible sorbate–sorbate interaction (Kannan and Sundaram 2001). Lastly to confirm the results, Langmuir adsorption model was checked by the R_L value which is a dimensionless constant or separation factor or equilibrium parameter. R_L is computed by means of the subsequent equation

$$R_L = \frac{1}{1 + K_L C_0}, \quad (15)$$

where C_0 is the initial As(V) concentration (mg L^{-1}).

The value of R_L indicates the isotherm nature which may be irreversible ($R_L = 0$), favorable ($0 < R_L < 1$) or unfavorable ($R_L = 1$). Investigational data suggest the value of R_L was in between 0 and 1; which confirm adsorption was favorable and excellent fit of Langmuir model to describe the investigational data. Apart from Langmuir model, the parameters of D–R model have also some significance due to its R^2 value (0.984) and low Chi square value (0.378). From D–R model, the magnitude of E

Table 4 Comparative adsorptive capacity of prepared SAC-MBH and other different adsorbent found in the literature towards the adsorption of As(V)

Adsorbent	Adsorptive capacity (mg g ⁻¹)	References
Iron oxide/activated carbon composite	0.4123	Yao et al. (2014)
Iron oxide-coated sand	0.043	Thirunavukkarasu et al. (2002)
Untreated GAC	0.038	Gu et al. (2005)
Steam activated charcoal from mung bean husk	0.36	Present study
Aluminum-loaded coral limestones	0.15	Ohki et al. (1996)
Granular ferric oxide	0.004	Badruzzaman et al. (2004)
Olivier soil	0.42	Zhang and Selim (2005)
Coconut shell carbon with 3% ash	2.4	Lorenzen et al. (1995)
Commercial nanomagnetic	0.205	Türk et al. (2010)
HBC-Fe ₃ O ₄ -MnO ₂ bio composite	1.46	Zhu et al. (2015)
Magnetic nanoparticle	0.485	Shipley et al. (2010)

Table 5 Thermodynamics parameters for As(V) removal by using SAC-MBH

Temperature (K)	ΔG (kJ mol ⁻¹)	ΔH (kJ mol ⁻¹)	ΔS (J mol ⁻¹ K ⁻¹)
298	-2.301	-76.909	266.9
303	-4.151		
308	-6.027		
313	-6.125		

Experimental conditions: adsorbent dosage 0.75 g; pH 3, agitation speed 160 rpm; temperature 308 K

(21.218 kJ mol⁻¹) indicates the process followed chemisorptions adsorption. The maximum adsorption capacity of As(V) by SAC-MBH has been compared with other adsorbents found in different literature (Table 4).

Thermodynamic studies

Thermodynamic behavior of a system can be understood by the help of the following equation:

$$\Delta G^0 = -RT \ln K_c, \quad (16)$$

$$K_c = \frac{C_a}{C_e}, \quad (17)$$

$$\Delta G^0 = \Delta H^0 - T\Delta S^0, \quad (18)$$

where K_c is adsorption distribution coefficient, C_a and C_e are the equilibrium arsenic concentrations on the adsorbent and in the solution, respectively. ΔG^0 is the Gibbs free energy change, ΔH^0 is the enthalpy change and ΔS^0 is the entropy change (Yao et al. 2010).

A plot of ΔG^0 versus T gives the slope and intercept, from that the values of ΔS^0 and ΔH^0 can be measured (Fig. 5c).

Gibbs free energy for the adsorption of As(V) by SAC-MBH was obtained from the Eq. (15) and presented in Table 5.

The negative value of ΔH^0 strongly supports the exothermic nature of adsorption while the negative value of ΔS^0 suggests that the process was enthalpy driven. At all temperatures, the negative values of ΔG^0 confirmed the spontaneous and feasible adsorption process. By increasing the temperature, the value of ΔG^0 becomes more negative that suggests that the higher temperature makes the adsorption favorable.

Modeling of the process

Response surface methodology

The experimental design of the modeling of removal process was carried out and results were obtained according to CCD matrix (Table 1). Experimental data of As(V) removal process were fitted into different models (linear, interactive, quadratic and cubic) using two tests, i.e., the sequential model sum of squares and lack of fit test were carried to find the best fitted model and the model summary statistics are specified in Table 6. It was clear from the results that high value of determination of coefficient (R^2), adjusted determination of coefficient (R^2_{adj}), predicted determination of coefficient (R^2_{pre}) while low value of p values in quadratic model proved its competency among the different models. In addition to this, the experimental data were successfully covered in this model. The quadratic model was expressed in terms of coded variables (Eq. 19) and in terms of actual variables (Eq. 20).

$$\begin{aligned} \% \text{ Removal of As (V)} = & + 94.85 - 1.95 \times A + 0.60 \times B \\ & + 0.80 \times C - 0.63 \times A \times B \\ & + 0.75 \times A \times C + 0.50 \times B \times C \\ & - 1.39 \times A^2 - 2.27 \times B^2 - 3.33 \times C^2 \end{aligned} \quad (19)$$

$$\begin{aligned}
& \% \text{ Removal of As (V)} \\
& = +65.53489 + 1.66153 \times \text{pH} + 0.18179 \\
& \quad \times \text{Agitation speed} \\
& + 0.028224 \times \text{Dosage} - 3.84615\text{E} - 003 \times \text{pH} \\
& \quad \times \text{Agitation speed} \\
& + 7.50000\text{E} - 004 \times \text{pH} \times \text{Dosage} + 1.92308\text{E} - 005 \\
& \quad \times \text{Agitation speed} \times \text{Dosage} - 0.22226 \times \text{pH}^2 \\
& \quad - 5.37990\text{E} - 004 \\
& \times \text{Agitation speed}^2 - 2.08354\text{E} - 005 \times \text{Dosage}^2
\end{aligned}
\tag{20}$$

Quadratic model's statistical significance was checked using ANOVA (analysis of variance). The results of ANOVA for second order response surface model are shown in Table 6. Different parameters associated with quadratic model proved that the model is significant such as 1. High fishers F value (94.43). 2. Very low probability value (<0.0001). 3. Multiple correlation coefficient (R^2) value (0.9946) was fairly high so significant for Goodness of fit test. 4. Predicted multiple correlation coefficient value (0.9277) was in a reasonable

good agreement with the adjusted multiple correlation value (0.9778). 5. The lack of fit value is less than 0.05. 6. Coefficient of variance (CV) value (0.66) is low suggesting precision and reliability of the data. 7. The values of “probability $>F$ ” less than 0.0500 indicate that the model term is significant and 8 coefficients of the main, square as well as interaction effects of model terms are significant.

Effect of adsorbent dosage and pH on As(V) removal Prediction of As(V) removal by the combined effect of adsorbent dosage and solution pH is described in three dimensional plot (Fig. 6a). Increasing the adsorbent dosage while decreasing the pH of the solution increases the % removal of As(V). This phenomena occurs as more adsorbent dosage indicates more adsorbent mass so increases adsorbent surface area that ultimately leads to more sorption site for adsorption. The adsorbed As(V) amount increases with decrease in solution pH. The explanation of the fact is that when the solution pH is less than pH_{pzc}, then the adsorbent surface (pH_{pzc} = 8.6) becomes positively charged where the surface of arsenate

Table 6 RSM Model fit summary statistics and ANOVA analysis of RSM model

Source	Standard deviation	R^2	Adjusted R^2	Predicted R^2	PRESS	Remarks
RSM model fit summary statistics						
Linear	3.87	0.214556	0.067285	−0.11991	341.726	
2FI	4.21	0.246099	−0.10186	−1.02718	618.5689	
Quadratic	0.6	0.98833	0.977828	0.927697	22.06223	Suggested
Cubic	0.56	0.993758	0.980233	0.222053	237.3807	Aliased
Source	Sum of squares	Degree of freedom (df)	Mean square	F -value	Probability $> F$	
ANOVA						
Model	301.584	9	33.5	94.1	< 0.0001	Significant
A	51.86	1	51.86	145.65	< 0.0001	
B	4.929	1	4.92	13.84	0.0040	
C	8.678	1	8.68	24.37	0.0006	
AB	3.125	1	3.12	8.78	0.0142	
AC	4.5	1	4.5	12.64	0.0052	
BC	2	1	2	5.617	0.0393	
A	27.809	1	27.81	78.09	< 0.0001	
B	74.45	1	74.46	209.1	< 0.0001	
C	160.15	1	160.16	449.79	< 0.0001	
Residual	3.56	10	0.356			
Lack of fit	2.727	5	0.545496	3.27	0.1095	Not significant
Pure error	0.833	5	0.166667			
Total	305.14	19				
Std.dev.	Mean	C.V. %	R^2	Adj. R^2	Predicted R^2	
Model statistics						
0.596725	90.075	0.662	0.988	0.977	0.927	

Fig. 6 Three-dimensional plot showing the interaction effect of **a** pH and dosage, **b** agitation speed and dosage and **c** pH and agitation speed of SAB-MBH on removal of As(V)

($pK_a = 6.97$) becomes more negative at pH; thus, negatively charged anionic particles (arsenic molecules) strongly attract positively charged cationic surface of adsorbent and, thus, adsorption became more at low pH.

Effect of adsorbent dosage and agitation speed on As(V) removal The combined effect of adsorbent dosage

and agitation speed was shown in the three-dimensional plot as indicated in Fig. 6b. Both independent variables have positive effect on the As(V) removal so increasing the both variables has synergistic effect on this process.

Effect of pH and agitation speed As(V) removal Figure 6c demonstrates the interactive effects of two independent

Fig. 7 **a** Regression plot of ANN model. **b** Residuals distribution patterns of RSM and ANN

Table 7 Trial and error method using transfer function (poslin and purelin) for % removal of As(V) by ANN model

Hidden layer algorithm	Output layer function	Correlation coefficient
Levenberg–Marquardt back propagation	Trainlm	0.9984
Resilient back propagation	Trainrp	0.965
Scaled conjugate gradient back propagation	Traincgp	0.588
Conjugate gradient back propagation with Polak-Ribiere updates	Trainscg	0.212
Gradient descent with momentum and adaptive learning rate back propagation	Traingdx	0.326

variables, i.e., pH and agitation speed. Regression analysis indicates the model term pH and agitation speed has net positive effect on As(V) removal. Collective p value (0.0393) of these two parameters is lesser than 0.1000 signifying the combined model terms.

Artificial neural network training for the optimization of the As(V) removal model

Input data were feed into the ANN architectures where algorithm (error back propagation) was used to train the process. The same experimental matrix of CCD was used to train the input and output data of ANN modeling of the process. Independent variables, i.e., dosage, pH and rpm, are selected as input nodes where dependent variables response or the removal of As(V) is the output nodes. In the

hidden layer, nonlinear transformations were done to test different types of algorithm to select the best network which gave R^2 (coefficient of correlation) that is very close to one.

Training of ANN network to optimize the removal process is a crucial step. The selection of POSLIN (positive linear transfer function) for input data where PURELIN was selected for output data to train the ANN. The best ANN network was selected by optimizing neuron present within the hidden layer. After feeding the data and running the software in ANN, we found the best correlation coefficient value (0.99) from the regression plot of the trained network (Fig. 7a). A high correlation coefficient that is closer to 1 signified the neural network modeling reliability with the experimental data. Among the different algorithm used in ANN (Table 7), Levenberg–Marquardt back

propagation (Trainlm) showed the best fitted algorithm to explain the ANN networking for the removal process.

Comparison of RSM and ANN network for the removal of As(V)

Utilizing RSM and ANN modeling process, linear and nonlinear multivariate regression problems are solved. The difference between the experimental value and model predicted value (RSM and ANN) is compared in Table 1. The residuals distribution pattern of RSM and ANN is shown in Fig. 7(b). Compared to RSM model, the residual fluctuation of ANN models is less diverge, normal and minute. Further both models are compared using three statistical estimators, i.e., root mean squared error (RMSE), coefficient of determination (R^2) and absolute average deviation (AAD)

$$\text{RMSE} = \left(\frac{1}{n} \sum_{i=1}^n (T_{\% \text{removal, predict}} - T_{\% \text{removal, exp}})^2 \right)^{1/2}, \quad (21)$$

$$R^2 = \frac{\sum_{i=1}^n (T_{\% \text{removal, experimental}} - \overline{T_{\% \text{removal, cal}}})^2}{\sum_{i=1}^n (T_{\% \text{removal, experimental}} - \overline{T_{\% \text{removal, cal}}})^2 + (T_{\% \text{removal, experimental}} - T_{\% \text{removal, cal}})^2}, \quad (22)$$

$$\text{AAD} = \left(\frac{1}{n} \sum_{i=1}^n \left(\frac{T_{\% \text{removal, predict}} - T_{\% \text{removal, exp}}}{T_{\% \text{removal, exp}}} \right) \right) \times 100, \quad (23)$$

where n is the number of points, $T\%$ removal, predict is the predicted value, $T\%$ removal, exp is the actual value, and the symbol ‘-’ is the average of the related values.

Statistical comparison (Table 8) showed that both models have good quality of data fitting and estimating capability, although ANN has some slight edge over RS. The advantage of ANN is its flexibility and non-requirement of standard experimental design. Its disadvantages include no combined parametric effect on the process. In general, ANN approach is a trustworthy, rational and

simulates non-linear form while overcoming the limitations of quadratic non-linear correlation assumption of RSM.

Conclusion

Isotherm, kinetics, thermodynamics and optimization of the process parameter associated with adsorption of pentavalent arsenic onto mung bean husk derived activated carbon have been studied in detail. Experimentally the maximum adsorptive removal of arsenic has been observed to take place at an adsorbent dosage 0.75 g, pH 3.0, agitation speed 160 and temperature of 308 K in case of 2 ppm As(V). Out of four isotherm models tested, the fitting order with respect to coefficient of correlation was Langmuir > D-R model > Temkin > Freundlich. The kinetic aspect of the data showed that the adsorption followed pseudo-second-order model. The heat of sorption was found to be (21.218 kJ mol⁻¹) which indicates that the sorptive removal process was chemisorption. Thermodynamic parameters suggest that the adsorption process was exothermic, spontaneous, and feasible in nature and higher

temperature was favorable for adsorption process. An effort has been made to model the removal process utilizing RSM and ANN network approaches. The process modeling done by ANN ($R^2 = 0.9969$) has some minute edge over RSM ($R^2 = 0.9883$) although in general both the models have some good optimizing capability.

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Table 8 Comparative statistical analysis of RSM and ANN

Parameters	RSM	ANN
RMSE	0.422	0.215
R^2	0.9883	0.99698
ADD (%)	0.00093	0.00011

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