

Evaluation of *Moringa oleifera* Carbon for the As(III) Removal from Contaminated Groundwater

Suman Barua¹, Ismail M. M. Rahman¹, M. Nazimuddin², and Hiroshi Hasegawa³

¹Department of Applied and Environmental Chemistry, Faculty of Science,
University of Chittagong,
Chittagong 4331, Bangladesh

²Department of Chemistry, Faculty of Science,
University of Chittagong,
Chittagong 4331, Bangladesh

³Institute of Science and Engineering,
Kanazawa University,
Kakuma, Kanazawa 920-1192, Japan

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ABSTRACT: Removal of trivalent arsenic from contaminated groundwater was studied using steam activated carbon prepared from the leaf, seed and pod of *Moringa oleifera* (MO) plant of the indigenous-cultivar of Bangladesh. Batch adsorption experiments were performed as a function of contact time, adsorbent doses and variants. The removal efficiency of the MO-leaf-carbon and MO-seed-carbon was substantial, while it was trivial for MO-pod-carbon. The pseudo-first- and second-order and intra-particle diffusion equations were used to evaluate the sorption mechanism of the MO carbon options. The MO is a common plant variety of the arsenic-affected Bengal delta. Therefore, it can be exploited as a cheaper resource of carbonaceous adsorbent for the economical removal of arsenic from the water.

KEYWORDS: *Moringa oleifera*, leaf, seed, pod, arsenic, water treatment.

1 INTRODUCTION

Arsenic, a toxic trace element, have incorporated in different environmental compartments, food chains and ultimately in humans due to the geogenic arsenic contamination of the groundwater [1]. The chemical forms of arsenic species determine the corresponding toxicity of arsenic in humans. Arsenite [As(III)] and arsenate [As(V)] are the most common water-soluble As-species as existed in the natural water systems, and the trivalent species is 10 times more toxic than the pentavalent variant. The As(III) mostly exists in reducing ground waters and hydrothermal waters, while As(V) is more commonly found in surface waters and oxidizing ground waters [2]. In terms of the extent of groundwater arsenic contamination and potable water exploitation source, the most people are at risk in Bangladesh followed by West Bengal, India. Hence, an unavoidable risk causing both cancerous and non-cancerous health effects is assumable to the mentioned populations due to the exposure to high level of arsenic [3, 4], unless the groundwater-supply chain is combined with any treatment procedures.

The treatment techniques available for the processing of arsenic contaminated groundwater for the end-of-the-pipe drinking water supply are several [5-8], which include the application of ion-exchange resins, membranes and adsorption onto coagulated flocs or sorptive media. Arsenic removal by low-priced adsorbents has been the most promising technique, which can be implemented within a simplistic scheme and require least maintenance or operating cost [9]. There is increasing research interest in using alternative adsorbents prepared from low-cost or of minimum economic value materials

for arsenic removal, such as, laterite [10], sugarcane bagasse [11], rice husk [12], sawdust [13], coconut husk, oil palm shell [14], neem bark [15], peat, compost, leaf mold, straw, wool fiber, soybean and cottonseed hulls, etc. [16, 17].

Biodegradability and low sludge production are some advantageous factors, which promote the use of natural coagulants for water treatment in developing countries [18, 19]. The use of natural coagulant, such as *Moringa oleifera* (MO) is better than other chemical coagulants [20, 21] mostly in terms of the amount of produced sludge [22], while its coagulation mechanism consists of adsorption and neutralization of the colloidal charges [20]. The MO, which is a small deciduous tree, is the most widely naturalized species of *Moringaceae* family [23, 24]. It is native in Asia Minor, Africa, India, Pakistan and Bangladesh [25, 26]. The potential of MO or other *Moringa* species-variants for the removal of arsenic [27-31], lead [32], cadmium [33], or other waterborne toxins [34-36] has been reported [22, 37]. However, the mentioned studies are mostly concentrated on the exploitation of *Moringa* seed.

The objective of the present work is to check the potential of MO carbon powder prepared from the different usable parts of an MO plant, e.g., leaf, seed and pod for the removal of trivalent arsenic from the contaminated groundwater, which has not been reported before.

2 MATERIALS AND METHODS

2.1 REAGENTS

Analytical reagent grade commercial products were used throughout. The As(III) stock solution (10 mg L^{-1}) was prepared from sodium arsenite (BDH Chemicals, India), and the working standards were prepared by dilution on a weight basis. Double-distilled deionized water was used throughout.

2.2 COLLECTION OF SAMPLES

The origin of the *Moringa oleifera* (MO) plants used in this study was a private grove located in the hilly area of the Chittagong district, Bangladesh. The mature pods of MO were collected and the seeds were separated from the pods. The MO leaves were also picked up from the matured MO plants. All the masses were sorted, sun-dried and ground to powder. The powdered samples were stored at $20 \pm 1 \text{ }^\circ\text{C}$ until further use.

2.3 PREPARATION OF THE MO CARBON

The carbonization of the MO plant parts (leaf, seed and pod) was performed in a muffle furnace at 200°C for 2 h. The carbon thus obtained was further treated following the process as described by the Warhurst *et al.* [38]. A set of three samples of treated carbon were prepared using leaf, seed, or pod of the MO plant ensuing the same procedure, followed by drying in an oven at 100°C . The products of each set were mixed together, homogenized, sieved, and stored in desiccators. The MO carbons representing the leaf, seed and pod parts are mentioned hereafter as MO-leaf-carbon (MO-LC), MO-seed-carbon (MO-SC) and MO-pod-carbon (MO-PC).

2.4 PREPARATION OF THE SIMULATED GROUNDWATER

Simulated groundwater resembling the typical groundwater characteristics available in the arsenic-prone regions of Bangladesh is prepared as described by Leupin *et al.* [39]. The composition is given in the Table 1. The simulated water was spiked with 0.5 mg L^{-1} of As(III).

Table 1. Composition of the simulated groundwater (GW) and the typical groundwater in Bangladesh (BGD-GW)

| | Simulated GW | Typical BGD-GW ^a |
|------------------------------------|--------------|-----------------------------|
| pH _{ini} | 7.0 ± 0.05 | 7.0 ± 0.2 |
| HCO ₃ ⁻ (mM) | 8 | 7.8 ± 2.7 |
| Ca (mM) | 2.5 | 1.9 ± 1.4 |
| Mg (mM) | 1.6 | 1.3 ± 0.8 |
| Si (mg L ⁻¹) | 20 | 19.2 ± 4.7 |
| P (mg L ⁻¹) | 2.0 | 1.47 ± 1.48 |
| As (mg L ⁻¹) | 0.5 | 0.199 ± 0.166 |
| Fe (mg L ⁻¹) | 0 | 5.3 ± 4.8 |
| Mn (mg L ⁻¹) | 0 | 0.57 ± 0.75 |
| DOC (mg L ⁻¹) | 0 | 3.3 ± 2.8 |

^a Data source: Leupin et al. [39] based on the database of BGS and DPHE [40]

2.5 ARSENIC REMOVAL EXPERIMENTS USING THE MO CARBON

A series of batch studies was performed by adding the MO carbons (MO-LC, MO-SC and MO-PC) at varying dose (1 to 3 g L⁻¹). The solution pH was maintained at 8.0 ± 0.5 and was adjusted using either HCl or NaOH (1 mol L⁻¹). The mixtures were stored in stoppered tubes, and were agitated at room temperature (25 ± 2 °C) using an end-to-end shaker at a fixed speed of 300 ± 20 rpm for various time intervals (0–24 h). The supernatant was separated from the solid residue via filtration with Whatman filter paper. An atomic absorption spectrometer (AAS) of the iCE 3000 Series (Thermo Scientific, Franklin, MA, USA) was used to determine the As(III) concentration in the supernatant. All the experiments were conducted in triplicates, and an averaged value is reported.

3 RESULTS AND DISCUSSION

3.1 EQUILIBRIUM STUDY: EFFECT OF CONTACT TIME

The change in the residual arsenic concentration with contact time with varying adsorbent doses is shown in Figure 1. The pattern indicates that the adsorption of As(III) by MO carbons is a rapid process and often reaches equilibrium within 12 to 24 h. The observed trend confirms that the removal of As(III) by MO carbon occurs at a higher rate in the first 5 hours, while it becomes slower at the subsequent hours approaching gradually to the equilibrium state. The comparative difference between the concentration of As(III) in the bulk solution and the number of the adsorbent sites are high at the initial hours, which causes a rapid uptake. The rate tends to be slowed down as the saturation of the adsorbent sites occurs at the late hours. The decrease in As(III) concentration with time occurs up to 12 h and then the curves seem to be flattened, i.e., an equilibrium condition is achieved.

The comparative uptake rate of As(III) in the adsorbent phase can be expressed using the term q_t (mg g⁻¹). The initial As(III) concentration (C_i) (mg L⁻¹), the concentration of As(III) in solution at any time (C_t) (mg L⁻¹), the total volume of the solution (V_f) (L) and the mass of the adsorbent (m) (g) are used to calculate the q_t using the following equation [41]:

$$q_t = (C_i - C_t) \frac{V_f}{m} \quad (1)$$

The trend in the change of the q_t of the MO carbons (MO-LC, MO-SC and MO-PC) at different doses (Figure 2) indicate that the As(III) uptake rate in the adsorbent phase has a reciprocal relationship with the adsorbent doses. The pattern might be attributable to the step up in the adsorbent sites with the increasing dose. Hence, the most suitable dose of MO carbon to treat the contaminated groundwater will be required to be determined based on the initial arsenic content of the raw water.

A comparison of the As(III) removal efficiencies of the MO carbons (MO-LC, MO-SC and MO-PC) at different doses is illustrated in the Figure 3. The As(III) removal performance of the MO-PC is considerably low compared to the other two options, and is not recommended as a viable option for the said purpose. A moderately comparable rate of the As(III) removal was achieved with the MO-LC or MO-SC, and the order can be shown as MO-SC > MO-LC for being to be more precise. Therefore, any of those options can be chosen based on the resource conditions.

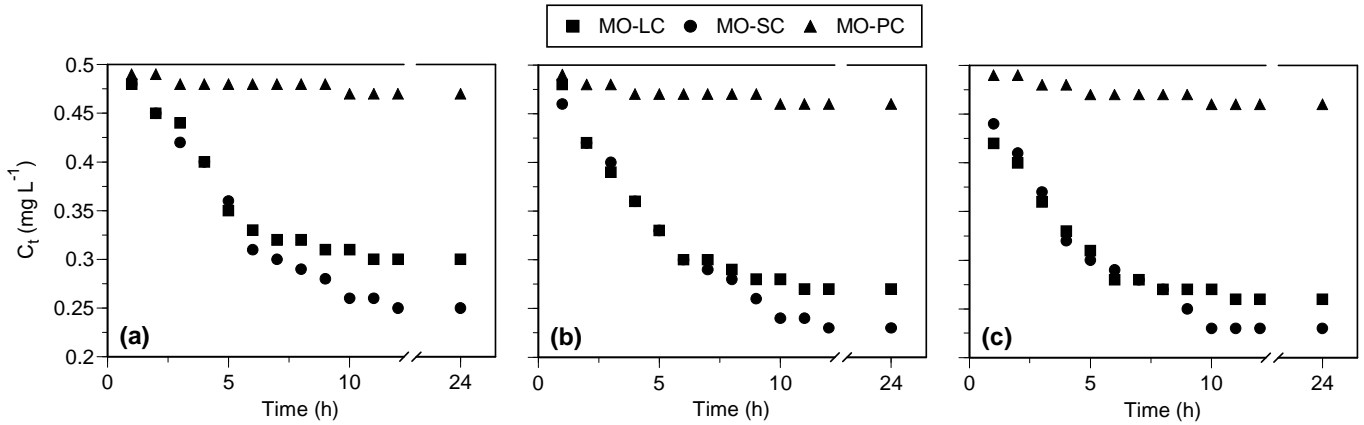


Fig. 1. The effect of contact time on the As(III) uptake rate at varying MO carbon doses: (a) 1 g L^{-1} , (b) 2 g L^{-1} , (c) 3 g L^{-1} MO-LC, MO-leaf-carbon; MO-SC, MO-seed-carbon; MO-PC, MO-pod-carbon t , 0 – 24 h; C_0 , 0.5 mg L^{-1} ; T , $25 \pm 2 \text{ }^\circ\text{C}$; $n = 3$

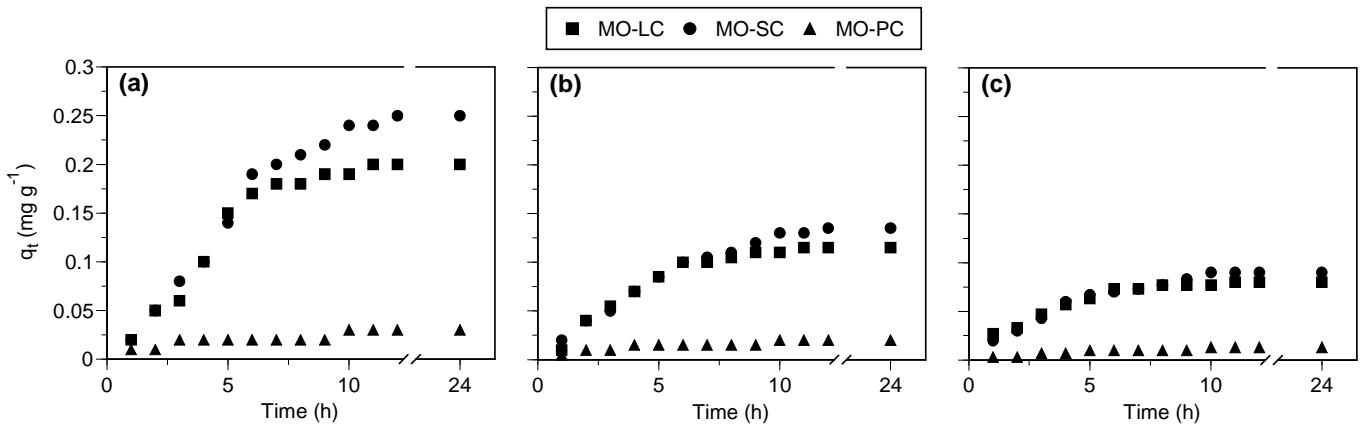


Fig. 2. The change in the q of the MO carbon variants with time at varying MO carbon doses: (a) 1 g L^{-1} , (b) 2 g L^{-1} , (c) 3 g L^{-1} MO-LC, MO-leaf-carbon; MO-SC, MO-seed-carbon; MO-PC, MO-pod-carbon t , 0 – 24 h; C_0 , 0.5 mg L^{-1} ; T , $25 \pm 2 \text{ }^\circ\text{C}$; $n = 3$

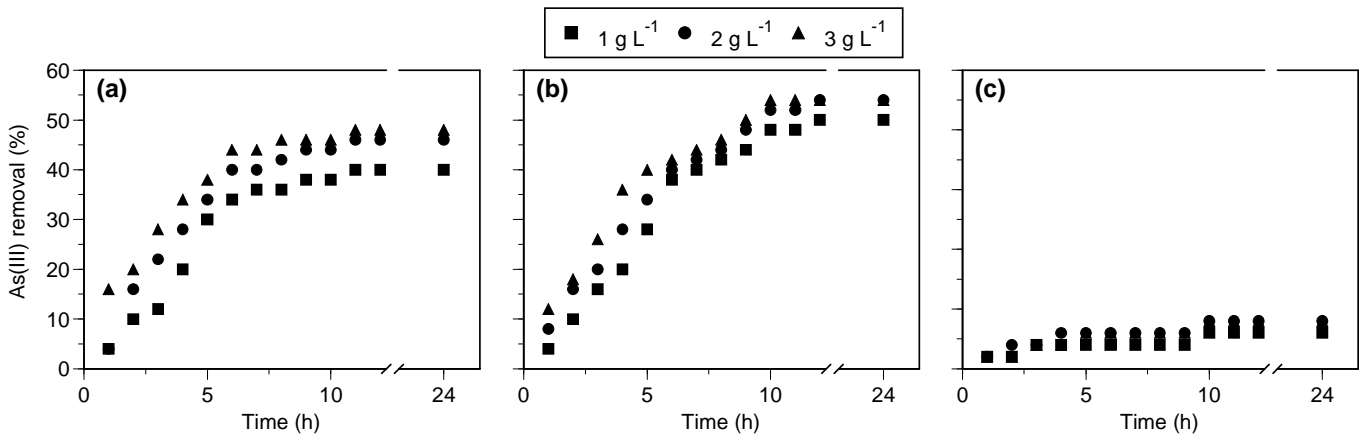


Fig. 3. The arsenic removal efficiency of the MO carbon variants: (a) MO-LC, (b) MO-SC, (c) MO-PC MO-LC, MO-leaf-carbon; MO-SC, MO-seed-carbon; MO-PC, MO-pod-carbon t , 0 – 24 h; C_0 , 0.5 mg L^{-1} ; T , $25 \pm 2 \text{ }^\circ\text{C}$; $n = 3$

3.2 ADSORPTION DYNAMICS

The kinetics of arsenic uptake by MO carbons is evaluated using the Lagergren’s pseudo-first-order and Ritchie’s pseudo-second order rate equations as derived using the adsorbent phase concentrations of As(III) (mg L^{-1}) against time (h) data.

The Lagergren’s kinetics equation has been widely used for the adsorption of an adsorbate from an aqueous solution [42, 43]. The most applied style of the equation has been proposed by Trivedi *et al.* [44], which is as follows:

$$\ln(1 - q_t/q_e) = -k_1 t \tag{2}$$

The second-order empirical equation proposed by Ritchie [45] has been used to test the sorption of gases onto a solid, which has later been applied to the sorption systems of solution and solid [46, 47]. The linearized form of the equation [41, 48] is as follows:

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e} t \tag{3}$$

In Eq 2 and 3, q_t is the amount of arsenic adsorbed at any time (mg g^{-1}), q_e is the amount of arsenic adsorbed at equilibrium (mg g^{-1}), k_1 and k are the rate constants of a pseudo-first-order and pseudo-second-order adsorption reactions as expressed, respectively, in min^{-1} and mg min^{-1} .

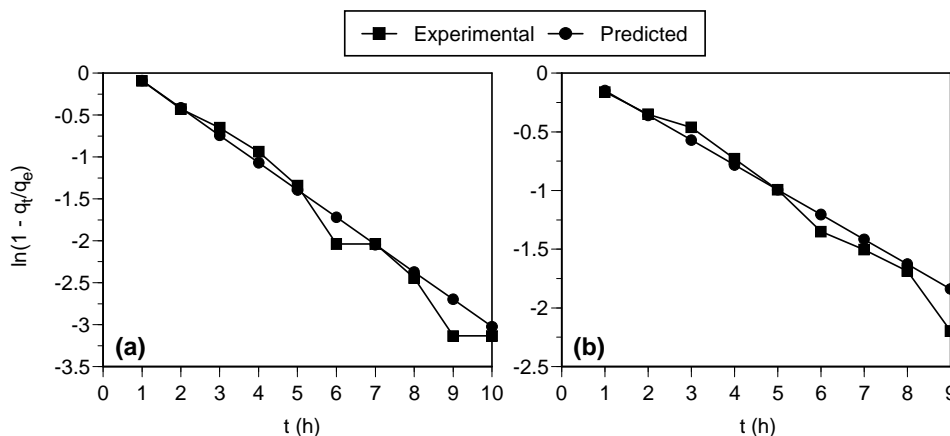


Fig. 4. Lagergren pseudo-first order kinetic plots for the MO carbon variants (dose 2 g L^{-1}): (a) MO-LC, (b) MO-SC MO-LC, MO-leaf-carbon; MO-SC, MO-seed-carbon. $C_0, 0.5 \text{ mg L}^{-1}$; $T, 25 \pm 2 \text{ }^\circ\text{C}$; $n = 3$

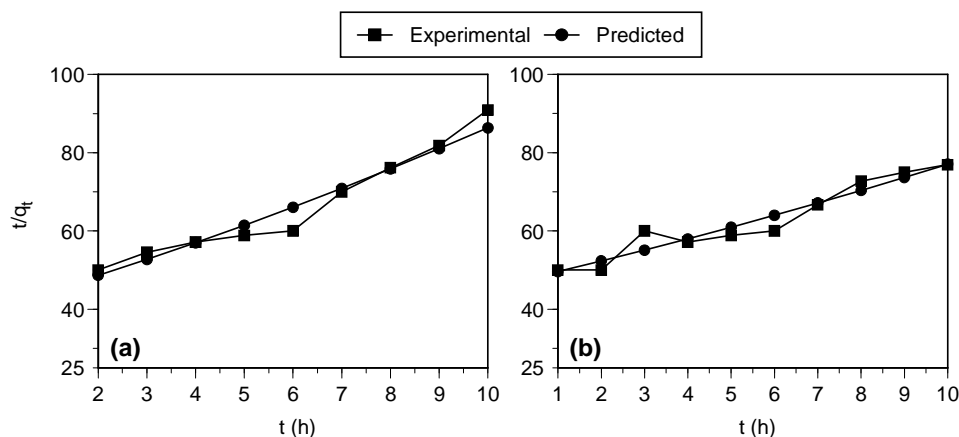


Fig. 5. Ritchie’s pseudo-second order kinetic plots for the MO carbon variants (dose 2 g L^{-1}): (a) MO-LC, (b) MO-SC MO-LC, MO-leaf-carbon; MO-SC, MO-seed-carbon. $C_0, 0.5 \text{ mg L}^{-1}$; $T, 25 \pm 2 \text{ }^\circ\text{C}$; $n = 3$

The batch kinetic data for the adsorbent phase was fitted to the Lagergren's pseudo-first-order and Ritchie's pseudo-second order models, which are shown in Figure 4 and 5, respectively. The regression calculation of the data-sets was performed using the non-linear optimization by Levenberg–Marquardt algorithm, and the most favorable fit between the experimental and theoretical data were observed for Ritchie's model. Therefore, it can be said that the pseudo-second order model best describe the behavior of As(III) adsorption by MO carbon variants.

3.3 ROLE OF TRANSPORT IN ARSENIC UPTAKE

The process of adsorption in a solid from solution might consist of the transport of the adsorbate from the bulk phase to the laminar liquid film at the surface, transport of the particles through the film and uptake into the interior of the adsorption sites. Besides the reaction kinetics, the transport processes, such as, film and pore diffusion also controls the mechanism of the adsorption. A plot of $\ln(C_t/C_i)$ versus time (Figure 6), based on Eq 4, was used to evaluate whether the film resistance to mass transfer controlled the As(III) uptake during the batch adsorption experiments with the MO carbons.

$$\ln \frac{C_t}{C_i} = -\frac{K_f W S_w}{V_f} t \quad (4)$$

In Eq 4, the K_f is the mass transfer coefficient for film diffusion, S_w is the specific external surface of adsorbent on weight basis, W is the adsorbent dosage, V_f is the volume of the fluid, C_i is the initial adsorbate concentration and C_t is the concentration of the adsorbate at any time. A linear pattern in the data-sets would indicate that the mechanism of adsorption is controlled by film diffusion [41]. However, all the data-trends confirms a non-linear pattern postulating that the rate of As(III) uptake was not controlled by the film diffusion. The solid-solution mixtures were agitated at a high speed (300 ± 20 rpm) during the kinetic experiments to ensure a high shear on the particle surface. As a result, the thickness of the film surrounding the particles was low and any possible rate-limiting effect due to the film diffusion was minimized [41, 49].

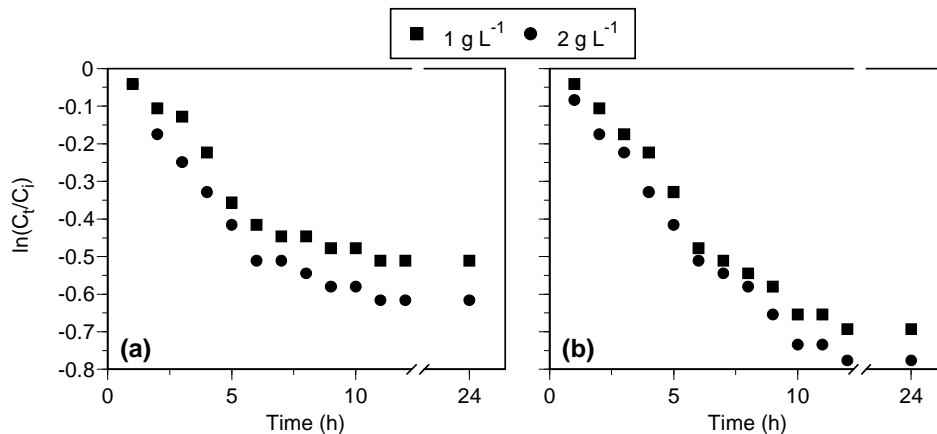


Fig. 6. Test for film diffusion as a rate controlling step in the As(III) uptake by the MO carbon variants: (a) MO-LC, (b) MO-SC
MO-LC, MO-leaf-carbon; MO-SC, MO-seed-carbon. T , 0 – 24 h; C_0 , 0.5 mg L^{-1} ; T , $25 \pm 2 \text{ }^\circ\text{C}$; $n = 3$

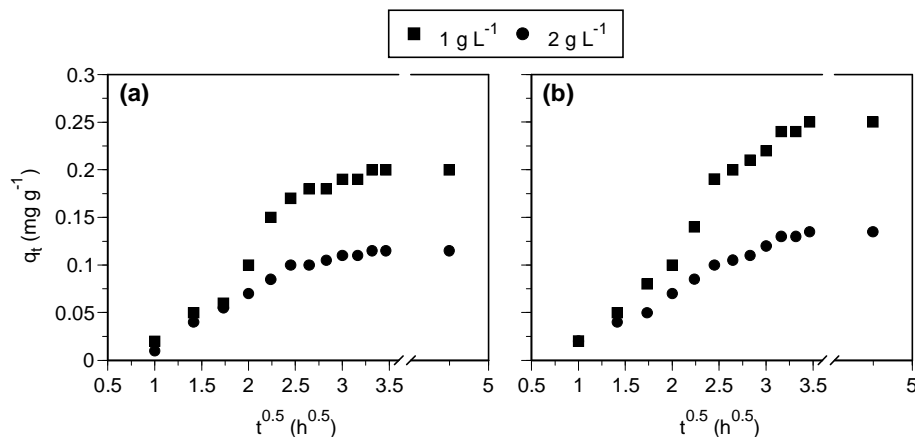


Fig. 7. Intra-particle mass transfer curve for the adsorption of As(III) by the MO carbon variants: (a) MO-LC, (b) MO-SC. MO-LC, MO-leaf-carbon; MO-SC, MO-seed-carbon. T , 0 – 24 h; C_0 , 0.5 mg L⁻¹; T , 25 ± 2 °C; $n = 3$

The intra-particle diffusion model [50, 51] have been used to describe the competitive adsorption occurs in a liquid-solid system. The initial rate of intra-particle diffusion can be calculated using the relationship between amount of adsorbed arsenic and square root of time (Figure 7):

$$q_t = k_{id} t^{0.5} \quad (5)$$

In Eq 5, q_t is the amount of arsenic adsorbed (mg g⁻¹) at time t (h) and k_{id} is the intra-particle diffusion rate constant (mg g⁻¹h^{-0.5}). The multi-linearity in the plots indicates that two or more steps occur in the adsorption processes. The initial sharper section of the plots represents the external surface adsorption or instantaneous adsorption stage. The next section of the plots is the gradual adsorption stage when the intra-particle diffusion is rate-controlled. The intra-particle diffusion starts to slow down due to the extremely low solute concentration in solution in the final step as represented by the plateau to the equilibrium. The linear portion of the plots for a wide range of contact time between adsorbent and adsorbate does not pass through the origin, which might be attributable to the difference of mass transfer rate in the initial and final stage adsorption [52]. Although the intra-particle diffusion is the major rate determining step, this variation from the origin or near saturation also represents that the contribution due to the surface adsorption cannot be ignored [53, 54].

4 CONCLUSION

The removal of As(III) from contaminated groundwater by MO carbon variants (leaf, seed and pod) is experimentally investigated in this study. It is explored that MO-LC and MO-SC can be effectively used for the As(III) removal, while the efficiency of the MO-PC is not significant. The As(III) uptake to the adsorbent phase occurred at a high rate in the initial hours followed by a slower subsequent removal rate with a gradual approach to a steady-state condition. The adsorption of As(III) can be better described by a pseudo-second order equation, e.g., Ritchie's model equation. It was confirmed that the intra-particle diffusion from the solution to the adsorption sites of the MO carbons can be considered as a major rate determining factor. *Moringa oleifera* can be harvested abundantly in the weather condition of the worst arsenic-affected area of Bengal Delta. Therefore, the use of *Moringa oleifera* derived carbonaceous materials for arsenic contaminated water treatment is expected to be economical and feasible. However, there is scope of improvement in the As(III) removal efficiency of the MO carbons via treatment with suitable chemicals. Moreover, column experiments are required to be conducted to explore the appropriate design of the As(III)-separation module, and a methodology should also require to be planned for the treatment of the spent MO carbons.

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