Removal of cadmium (Cd-II) from aqueous solution using gas industry-based adsorbent

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Abstract

The present study is to determine the potential of gas industry sludge as a low-cost adsorbent for the removal of cadmium from aqueous solution. The batch experiments were conducted to optimize pH, contact time, adsorbent dose and initial metal ion concentration. The experiments were optimized at pH 5, contact time 60 min and an adsorbent dose of 0.75 g/100 ml with constant agitation at the room temperature. The result of batch experiment was further discussed for the removal of cadmium from adsorbent. The adsorption data were correlated with Freundlich and Langmuir isotherms to know the interaction of sludge adsorbent with cadmium. High value of regression coefficient ($R^2 > 0.95$) indicated that the adsorption data are fitted well with both Langmuir and Freundlich isotherms. The adsorption capacity of 25 mg/g was found to be quite good for removal of the cadmium from aqueous solution. Desorption efficiency showed highest removal with HCl (42.83%); followed by HNO_3 (38.26%), EDTA (37.73%) and H_2SO_4 (36.83%). The removal and desorption efficiency of the gas industry-based adsorbent was found to be comparable and even more suitable than the chemically modified adsorbents. The present study provides a low-cost and environmentally friendly adsorbent, which can be used as an alternative for the treatment of wastewater.

Keywords Adsorption isotherm · Batch reaction · Cadmium · Gas industry sludge · Heavy metals

1 Introduction

Heavy metal pollution is a major environmental concern at the global scenario and has limited the availability of fresh water for the increasing population [1, 2]. The changing water quality has become a serious problem for the entire nations of the world [3]. The heavy metals are non-biodegradable, cause damage along the trophic level and create serious problems due to their persistence, toxicity and bioaccumulation nature [4–7]. Some of the heavy metals are biologically essential and act as a micronutrient for plants and relatively harmless, but some are purely toxic (i.e. arsenic, beryllium, cadmium and lead). These include metallic elements having atomic weight in the range 63.5

and $200.6 \text{ g mole}^{-1}$ with a specific gravity greater than 5 g cm^{-3} [8].

The heavy metals are contributed by different sources, and their high concentration may cause serious health hazards [9, 10]. Cadmium (Cd-II) is one of the heavy metals having main sources such as the wastewater, industrial effluents, paint, plastic, synthetic fertilizer, metal coatings industry [11], atmospheric deposition and rocks [12]. This metal is very harmful, and the human being is susceptible from residential or occupational exposure even at very minute concentration (0.5 mg/L). High level of cadmium in the wastewater may lead to various ailments [13]. International Agency for Research on Cancer (IARC) has classified that the cadmium and its compounds are carcinogenic in nature [14]. The ingestion and inhalation of cadmium may

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cause dysfunction of the kidney and severe damage to the lungs; and instead, its chronic exposure may lead to death [1, 15]. A high proportion of cadmium in the soil may produce oxidative stress and nutritional deficiencies [16–18].

The wastewater treatment has become a worldwide problem and concern for the policy makers and environmentalists [19]. Various conventional physiochemical processes are used for the removal of heavy metals from wastewater such as coagulation, flocculation [20], precipitation [21], adsorption [22], membrane filtration [23], electrolysis [24], photocatalytic degradation [25], ion exchange [26], oxidation, reduction [27] and solvent extraction [28]. Different researchers have indicated the strength and limitation of these techniques [1, 29–31]. Among these, adsorption method does not require a huge input of chemical reagents that create other environmental and health problems to deal with secondary pollutants. Adsorption studies have investigated the low-cost adsorbents from agricultural and industrial wastes and found them to be much effective for the removal of heavy metals from wastewater [32–34]. Some of these studies modified the adsorbents with various chemicals to increase the sorption capacity [29, 35, 36], but still there is a stringent need to explore low-cost and high-potential adsorbent that could have appreciable removal efficiency and desorption capacity for the recovery of heavy metals from the wastewater.

In the present study, the gas industry sludge has been used without any chemical treatment to prepare gas industry-based adsorbent (GIBA), which was further used in a batch experiment for the removal of cadmium (Cd-II) from aqueous solution. The study covered a series of batch experiments for the optimization of pH, contact time, adsorbent dose and initial metal ion concentration; followed by the desorption process to check the reusability of adsorbent for further use. The Langmuir and Freundlich isotherms were used to describe the adsorption of Cd-II on the adsorbent. Thus, the present study aims to explore the potential of gas industry sludge for the removal of heavy metal from synthetic solution of cadmium. The adsorption isotherm provides input to better understand the interaction of adsorbate on the adsorbent that can further be helpful to design the batch system for wastewater treatment.

2 Materials and methods

2.1 Preparation of gas industry-based adsorbent (GIBA)

The sludge samples were collected from Indraprastha Gas Limited, New Delhi, India. The samples were transported

to the laboratory and kept in oven at 103 °C for 24 h and thereafter grinded using agate mortar. Then the samples were sieved, marked as gas industry-based adsorbent and stored in desiccator.

2.2 Preparation of synthetic solution of cadmium for batch reaction and optimization

The stock solution of Cd-II (1000 mg/L) was prepared by dissolving cadmium chloride ($\text{CdCl}_2 \cdot \text{H}_2\text{O}$) in double-distilled water and further diluted to prepare different solutions of various concentrations. The batch experiment was conducted to check the influence of pH, adsorbent dose, contact time and initial metal ion concentration (Cd-II) on adsorption efficiency. In case of pH optimization, the stock solution (100 mg/L) was taken in titration flask and pH was adjusted in the range of 2–7 using 0.1 N HCl and 0.1 N NaOH and calibrated with buffer solutions (pH 4.0, 7.0, 9.2). The experiment was conducted in a fixed adsorbent dose of 1.0 g/L, and the stock solution was agitated using orbital shaker at 150 rpm for 3 h and constant temperature (30 ± 2 °C). Finally, the solution was centrifuged at 4000 rpm for 15 min; and after filtration, the supernatant was taken to check the residual concentration of cadmium (Cd-II) with the help of atomic absorption spectrophotometer (Lab India AA-7000 series). The removal efficiency (%) of Cd-II was determined [37] using following equation (Eq. 1):

$$\text{Removal efficiency (\%)} = \left(\frac{C_0 - C_1}{C_0} \right) \times 100, \quad (1)$$

where C_0 represents the initial concentration of Cd-II and C_1 corresponds to its final concentration after the batch experiment. The optimized pH obtained from the above experiment was used to check the effect of contact time (15–180 min) and adsorbent dose (2.5–12.5 g/L). In the last run of batch experiment, the removal efficiency of Cd-II was checked using optimum pH, adsorbent dose and contact time with varied concentrations (20–200 mg/L) of Cd-II in aqueous solution, while the speed of orbital shaker (150 rpm) and the temperature (30 ± 2 °C) of the working environment were kept constant. Removal efficiency was calculated using a formula (Eq. 1) as stated above, and the uptake capacity of the Cd-II was also calculated using Eq. 2 as [38]:

$$Q_{\text{up}} (\text{mg/L}) = \frac{v(C_0 - C_f)}{M}, \quad (2)$$

where Q_{up} is the uptake capacity of Cd-II adsorbed on the GIBA, C_0 is the initial concentration (mg/L) of the Cd-II in aqueous solution and C_f is the final concentration of the

Cd-II after using v (volume) of the metal solution (L) with M (mass) of adsorbent (g) used in the experiment.

2.3 Desorption study

Desorption studies were carried out to check the reusability of GIBA with respect to four desorbing agents such as HCl, H_2SO_4 , HNO_3 and EDTA; all having same molarity (0.1 M). Cd-II-loaded adsorbent (1 g) was taken within a desorbing agent 100 ml (0.1 M) using 250-ml Erlenmeyer flask, and a contact time of 30 min was applied with a stirring rate of 150 rpm at constant temperature ($30 \pm 2^\circ C$). Desorption efficiency was calculated [39] using Eq. 3 as:

$$\text{Desorption efficiency} = \frac{\text{Amount of metal ion desorbed}}{\text{Amount of metal ion adsorbed}} \times 100 \quad (3)$$

3 Results and discussion

3.1 pH optimization

The adsorption of cadmium (Cd-II) using GIBA in the present study was found to be varied at different pH levels, which has also been reported by many researchers using different types of adsorbents [12, 40, 41]. The pH value was optimized for the present batch experiment. The per cent removal and uptake capacity (mg/g) at different pH levels are shown in Fig. 1. There is an increase in removal and uptake capacities of Cd-II up to pH level of 5. The per cent removal and uptake capacity at this pH value were found to be 85.69% and 88.07 mg/g, respectively. There was no appreciable change in the uptake and removal capacities that helped to select optimum pH 5, and thereafter, the increasing pH level was considered less economically

Fig. 1 Effect of pH on per cent removal and uptake capacity of cadmium

viable. Adsorption studies conducted by many researchers with cadmium also showed optimization level close to pH 5.0 [32, 42–44].

3.2 Effect of adsorbent dose and contact time

To study the effect of adsorbent dose and contact time, the batch experiment was conducted at initial cadmium concentration of 100 mg/L, optimized pH (5), temperature ($30^\circ C$) and agitation speed (150 rpm). The adsorbent dose was found to be varied from 0.25 to 1.25 g/100 ml and contact time from 15 to 180 min, respectively. The result revealed that the adsorption rate increased from 83.761 to 87.215%, when contact time was increased from 15 to 180 min at the adsorbent dose of 0.25 g/100 ml. Also in the similar time frame, an increase in per cent removal was observed from 85.021 to 91.345%, 86.575 to 91.760%, 87.32 to 92.93% and 88.542 to 93.070% at 0.50, 0.75, 1.00 and 1.25 g/100 ml dose of adsorbent, respectively (Fig. 2). Figure 3 shows that the uptake capacity increased from 33.504 to 34.886 mg/g, 17.004 to 18.151 mg/g, 11.543 to 12.235 mg/g, 8.73 to 9.29 mg/g and 7.083 to 7.446 mg/g at 0.25, 0.50, 0.75, 1.00 and 1.25 g/100 ml dose of adsorbent, respectively, when contact time was increased from 15 to 180 min. There was no appreciable increase in the removal per cent for cadmium after 60 min.

It was observed that the per cent removal of cadmium from aqueous solution increased with increase in adsorbent dose from 0.25 to 0.75 g/100 ml. However, such an increase in per cent removal was not observed after increasing the adsorbent dose. This was due to “solid concentration” effect, which explains that the quantity of a metal adsorbed per unit quantity of adsorbent decreases significantly with increasing the mass

Fig. 2 Effect of adsorbent dose and contact time on cadmium removal by gas industry-based adsorbent

Fig. 3 Effect of contact time on removal and uptake capacities from gas industry-based adsorbent

of adsorbent per unit volume [45]. It was observed that the concentration of cadmium adsorbed on the gas industry-based adsorbent increased with time. During the initial stage of sorption, a large number of vacant sites were available for adsorption. It was also observed that with the increase in adsorbent dose, more surface area became available for adsorption due to increase in active sites on the adsorbent; then making easier penetration of metal ions to the adsorption sites [46]. After a lapse of time, the remaining vacant surface sites

occupied difficulty due to repulsive forces between the solute molecules on the adsorbent surface.

3.3 Effect of initial cadmium concentration

To study the effect of initial metal ion concentration, the optimized value of pH (5.0), contact time (180 min) and adsorbent dose (0.75 g/100 ml) were used in batch experiment. The cadmium concentration increased from 20 to 200 mg/L, and the per cent removal decreased from

Table 1 Effect of initial cadmium concentration on the per cent removal and uptake capacity of cadmium by gas industry-based adsorbent

Cadmium concentration (mg/L)	Residual cadmium concentration (mg/L)	Removed cadmium concentration (mg/L)	% removal	Uptake capacity (mg/g)
20	0.153	19.847	99.24	2.646
40	1.853	38.147	95.37	5.086
60	3.229	56.771	94.62	7.569
80	6.124	73.876	92.35	9.850
100	9.214	90.786	90.79	12.104
150	20.55	129.45	86.30	17.260
200	36.85	163.15	81.58	21.753

Fig. 4 Desorption efficiency of various desorbing agents for the removal of Cd-II from gas industry-based adsorbent

99.24 to 81.58%, whereas the uptake capacity increased from 2.646 to 21.753 mg/g (Table 1). This is because of more binding sites were available at a low concentration of metal ions, but as the concentration of metal ions increased, then the ions started to compete for available binding sites [47].

3.4 Desorption study

Recovery of metal ions adsorbed on the adsorbent is one the most important aspects for the development of a successful adsorption process [48]. The maximum desorption efficiency was observed to be 42.83% with gas industry sludge-based adsorbent using 0.1 M HCl as eluent (Fig. 4). The desorption efficiency of adsorbent for cadmium was found to be in the following order as $\text{HCl} > \text{HNO}_3 > \text{EDTA} > \text{H}_2\text{SO}_4$.

3.5 Adsorption isotherm

The adsorption isotherm represents the relationship between the concentration of the solute in the adsorbent (q_e) and solution at equilibrium (C_e) [49]. Adsorption

Fig. 5 Langmuir adsorption isotherm for cadmium by gas industry-based adsorbent

capacities were calculated from linearized Langmuir isotherm using Eq. 4:

$$C_e/q_e = 1/q_{\max}b + C_e/q_{\max}, \quad (4)$$

where C_e is the concentration of adsorbate solution at equilibrium and q_e is amount adsorbed per unit of adsorbent at equilibrium. The Langmuir isotherm considers that all of the metal ions are chemically adsorbed in a fixed number on those free sites, which are energetically equivalent [50]. The Freundlich isotherm [51] was also used to express the linearized logarithmic (Eq. 5) as:

$$\log q_e = (1/n) \log C_e + \log K_f, \quad (5)$$

where q_e is the amount of metal adsorbed in (mg/g) and K_f and $1/n$ are the relative indicators of the adsorption capacity. The Langmuir and Freundlich isotherms are plotted in Figs. 5 and 6, which indicated the value of $R^2 > 0.95$. The equilibrium biosorption data followed both Langmuir and Freundlich isotherms with $R^2 > 0.95$ (Table 2). The maximum cadmium adsorption capacity ($q_{\max}$) with gas industry-based adsorbent was found to be 25 mg/g. The present study was further compared with other studies, which indicated that the GIBA is having appreciable adsorption capacity in comparison with the biosorbent materials and chemically modified adsorbents (Table 3).

Fig. 6 Freundlich adsorption isotherm for cadmium by gas industry-based adsorbent

Table 2 Langmuir and Freundlich constant for cadmium by gas industry-based adsorbent

Langmuir constant			Freundlich constant		
$q_{\max}$ (mg/g)	b (L/mg)	R^2	K_f (mg/g)	n (L/mg)	R^2
25	0.14	0.953	4.90	2.52	0.978

The batch experiment further revealed that the GIBA is having high removal potential of heavy metal (Cd-II) and can be considered as a low-cost adsorbent for the wastewater treatment.

4 Conclusion

A wide range of adsorbents are used worldwide for the wastewater treatment; however, a search for the low-cost and environmentally friendly adsorbent is still there. Many researchers have inferred the alternative low-cost

adsorbents including agricultural and industrial wastes, which can be potentially used for the removal and recovery of the heavy metals. The present study explored the potential of gas industry-based adsorbent (GIBA) in batch experiment after the optimization of various adsorption parameters such as pH, contact time, adsorbent dose and effect of initial Cd-II concentration from the aqueous solution. The adsorption capacity of present adsorbent is found to be very high in comparison to the agricultural by-products and chemically modified adsorbents as well as biosorption. The high removal and desorption efficiency of GIBA from the aqueous solution strengthen its future use as a low-cost adsorbent for the removal of heavy metals from the wastewater.

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Compliance with ethical standards

Conflict of interest All the authors declare that they have no conflict of interest.

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Table 3 Comparison of adsorption capacity ($q_{\max}$) of gas industry-based adsorbent with other adsorbents for the removal of cadmium

Sr. no.	Adsorbent type	pH	$[q_{\max}]$ (mg/g)	References
1	<i>Mucor rouxii</i>	4–5	8.5	Yan et al. [52]
2	<i>Gracilaria salicornia</i> (red algae)	5	18.0	Hashim and Chu [53]
3	Clarified sludge	–	36.23	Naiya et al. [49]
4	Almond shell	5–6	7.19	Mehrasbi et al. [54]
5	Iron oxide-coated sludge	–	14.7	Phuengprasop et al. [34]
6	Sodium bicarbonate-treated newspaper pulp	6.4	1.44	Ossman and Mansour [55]
7	Natural phosphate	5	26	Yaacoubi et al. [56]
8	Zeolite clinoptilolite	4	16.73	Dstan and Dehghani [57]
9	<i>Anacyclus nigellifolius</i> Boiss	6	69.14	Mohammad et al. [58]
10	Barley straw biochar	6	1.6	Jazini et al. [59]
11	<i>Caulerpa lentillifera</i>	5	4.7	Pavasant et al. [60]
12	Gas industry sludge-based adsorbent	5	25.0	Present study

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