



Influence of Adsorption Process Parameters on the Removal of Hexavalent Chromium (Cr(VI)) from Wastewater: A Review

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Abstract

In recent years, the release of heavy metals into the aquatic environment has become a major issue. Numerous socio-economic problems are caused due to the presence of several heavy metals in wastewater. Hexavalent chromium (Cr (VI)) is one of the major heavy metal present in the wastewater which comes from various industries such as fertilizers, pesticides, metal cleaning, dyes and pigment, especially in tannery industry. Numerous methods have been employed for the removal of Cr (VI) from wastewater. Adsorption has been reported as a suitable method due to its high efficiency, low cost, generation of minimum chemical sludge and reusability of the prepared adsorbents. In this review, various adsorption process parameters such as solution pH, adsorbent dosage, temperature and initial Cr (VI) concentration have been reviewed on the removal efficiency of Cr (VI) from wastewater. The percentage removal of Cr (VI) strongly depends upon pH of the solution and the optimum pH range was found to be 1.0-4.0. The reusability of the used adsorbents has also been discussed. It is comparatively good for practical applications. It can be concluded that the most of the adsorbents have good regeneration capability. This review paper suggested that the adsorption process parameters had an important role on the removal efficiency of Cr (VI) from wastewater.

Keywords: Adsorption, Wastewater treatment, Hexavalent chromium, Reusability

1 Introduction

The discharge of heavy metals into the water bodies has become a major environmental concern over the world (1-2). Amongst all the heavy metals, hexavalent chromium (Cr(VI)) has been considered one of the major pollutant which is soluble in water and has deleterious effects on environment as well as human health (3-4). The major sources of Cr(VI) in wastewater/waterbodies are electroplating, textile dyeing and especially tanning processing industries (5). Hexavalent chromium is highly toxic substance and mutagenic to the most organisms and is recognized to cause cancer; it also causes lung carcinoma in human lives [2]. The Cr(VI) is present in the excess quantity in water/wastewater beyond a permissible limit which results in skin irritation, ulcer formation, liver damage and pulmonary congestion [1]. World Health Organization (WHO) recommended the maximum permissible limit of hexavalent chromium concentration is 0.05 mg/L in wastewater [1]. According to United States Environmental Protection Agency (US EPA), the acceptable level of Cr(VI) is 0.05 mg/L for potable water and is 0.1 mg/L for inland surface waters. Considering this limit, it is necessary for industries to control their chromium level

in wastewater so that Cr(VI) could be come in the acceptable levels before discharge in to aquatic environment (6-10). There are following four stages of the conventional chromium treatment processes as (8): (a) Cr(VI)–Cr(III) reduction, (b) at higher pH, precipitation of Cr(III) as Cr(OH)₃, (c) settle down of the insoluble metal hydroxide, and (d) disposal of the dewatered sludge.

Numerous methods such as ion exchange, electrochemical reduction, precipitation, adsorption and reverse osmosis have been reported in literature for the treatment of chromium-laden wastewater. Between all the above methods, adsorption has been considered to be an appropriate method for the removal of Cr(VI) from wastewater (11-15). The some advantages of the adsorption process are its ease of operation, effectiveness and economic viability.

In the last two decades, various new materials as adsorbents such as tea waste (1), modified groundnut hull (4), treated waste newspaper (6), activated neem leaves (13) and banana peel (15) efficient adsorbents have developed for the removal of Cr (VI). Mostly adsorbents have shown their higher adsorption efficiency towards the treatment of wastewater due to their lesser time. But,

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some adsorbents have limited applications because of their low adsorption capacity and high contact time. The purpose of this review article is to investigate the influences of various adsorption process parameters i.e. solution pH, adsorbent dosage, temperature and initial Cr(VI) concentration on the removal efficiency of Cr(VI) from wastewater. A summary on the reusability of the used materials as efficient adsorbents has also been presented in this review article.

2 Influence of various adsorption parameters on the removal efficiency of Cr (VI)

2.1 Influence of solution pH

Literature has reported that solution pH is a major factor affecting the adsorption efficiency of Cr(VI). The solution pH dependency of metal adsorption is generally linked to the functional groups of the adsorbents surface and ionic forms of metal ions in the solution (6, 16). It had been reported that maximum Cr(VI) removal efficiency was found under acidic conditions (1, 6, 9, 10, 17, 18-20). The summary of work done in the area of adsorption process for the removal of Cr(VI) from wastewater in recent years has been presented in Table 1. The reduction of Cr(VI) to Cr(III) may comprise generally two main mechanisms as: (1) the reduction of Cr(VI) to Cr(III) by electron-donor groups of the adsorbent surface and the reduced Cr(III) forms complexes with materials (2) the adsorption-coupled reduction of Cr(VI) to Cr(III) takes place on the sites of adsorbents (1, 17, 18). In literature, Cr(VI) is easily reduced to the Cr(III) due to its high redox potential value (1.3 V at standard state) under acidic conditions (1). In this regards, Nigam et al. (1) have studied the effect of pH over a pH range of 2–10 on the removal of Cr(VI) at an adsorbent dosage of 6 g/L and temperature of 30°C. Almost 97% removal was obtained at an optimum solution pH of 3.9 in 240 min of processing time. Owulude et al. (4) have investigated the removal of hexavalent chromium from aqueous solutions using modified groundnut hull as an efficient adsorbent at various pH. It was observed that acidic conditions are more relevant for the removal of Cr(VI) due to the presence of HCrO_4^- as a dominant species on to easy contact to the binding sites on the adsorbents so that electric attraction with H^+ ions on the surface of adsorbent can adsorbed higher metal ions. Therefore, maximum Cr(VI) removal of 96% was found at a low pH of 2.0. Chen et al. (20) have studied the effect of solution pH on the Cr(VI) removal from wastewater using modified corn stalks. They have reported that maximum adsorption of hexavalent chromium (99.5%) was obtained at pH value of 2.3.

Hence, it is very important to optimize the solution pH for each and every prepared new material as an adsorbent in order to achieve the maximum Cr(VI) removal efficiency from the wastewater.

2.2 Influence of adsorbent dosage

Adsorbent dose is another major important factor in the adsorption process which regulates the amount of metal ion removal and the economics of process. In the adsorption, adsorbent dosage to be used can be considered broadly by carrying out experiments where the amount of adsorbent is varied when other conditions are kept constants (20-22). It had been reported that there are various ranges of the different adsorbents dosage (shown in Table 1). In a literature study of Cr(VI) removal using biomass of *Agaricus bisporus*, it was seen that an adsorbent

dosage of 10 g/L was enough to acquire maximum removal of chromium (92.5%) where initial Cr(VI) concentration was 50 mg/L with temperature of 20°C and agitation speed of 150 rpm (22). It had been reported that the Cr(VI) removal percentage was enhanced from 74 to 99% for an increase in dosage from 0.5-1.5g/L due to the number of available sites of adsorption and surface area increased (20). Usually, it was understood that chromium removal efficiency was enhanced with increase in the adsorbent dosage up to an optimum as the number of vacant sites available for uptake of heavy metals. Nevertheless, adsorbent dosage cannot keep increasing to attain higher adsorption because of cost becomes a limiting factor and therefore adsorbent dosage is required to optimize for adsorption process.

2.3 Influence of temperature

Temperature plays an important role in deciding the removal efficiency of Cr(VI) in the adsorption process (4, 5, 12, 14). It is necessary to identify the suitable temperature to get good adsorbent efficiency. The nature of the adsorption can be assumed depending on the process responds with variation in employed temperature. The adsorption of heavy metals increases with an increase in the temperature therefore a process/reaction is endothermic in nature (5, 12, 17) whereas adsorption of metals decreases for an increase in the temperature therefore a process/reaction is exothermic in nature (8). In addition to this, to determine the process whether it is endothermic or exothermic; the thermodynamic parameters such as ΔG° , ΔH° and ΔS° need to be calculated. It has been reported that the values of change in entropy (ΔS°) and change in enthalpy (ΔH°) can be calculated from the intercept and slop of plot $\ln K_{eq}$ v/s $1/T$ (1, 5). The positive values of ΔH° show that the adsorption process is endothermic in nature and vice-versa (1). Various researchers have reported the temperature effect over a range of 10 - 60°C for the various adsorbents towards the Cr(VI) removal from wastewater (1-5,12, 14-15, 17). Nigam et al. (1) have studied the effect of temperature over a range of 20 to 40°C on the removal of Cr(VI) using tea waste as an adsorbent and observed that the maximum of 97% removal of chromium was obtained at a temperature of 30°C. The percentage removal of Cr(VI) was enhanced for an increase in the temperature from 20 to 40°C, which may be due to the increase in diffusion rate of the metal ion. Similar results were also reported by other study that the percentage removal of chromium was increased with an increase in the temperature suggesting that the adsorption process is an endothermic process (3, 5, 12, 14, 17). Apart from these studies, some studies have reported that the adsorption capacity decreased for an increase in the temperature advising that the adsorption process was exothermic process (4, 8). Hence, it is very important to find the temperature conditions so that process could be identified that it is either endothermic or exothermic in nature.

2.4 Influence of initial Cr (VI) concentration

The initial Cr(VI) concentration in wastewater samples is another important factor which influences the adsorption capacity of the metal ions. Previous studies have reported that the percentage removal efficiency of Cr(VI) is decreased at higher initial concentration of the Cr(VI) (3, 5, 13, 16, 20).

Table 1: Summary of work done on Hexavalent Chromium Cr(VI) removal from wastewater using different adsorbents

S. No.	Adsorbent	Treated Volume, mL	Ranges of Process parameters studied				Optimum conditions	Cr(VI) removal (%)	References
			pH	Adsorbent dosage	Temperature (°C)	Initial Cr(VI) Concentration			
1.	Tea waste	200	2-11	2-10 g/L	20-40	141-455 mg/L	pH = 3.9, adsorbent dosage = 6 g/L, T = 30°C, initial Cr(VI) concentration = 141 mg/L, time = 240 min	96.7	Nigam et al. (1)
2.	Synthesized chitosan from crab shell	100	3-8	1-5 g/L	30-60	1345±1 mg/L (real effluent)	pH = 3, adsorbent dosage = 5g/100mL, T = 60°C, time = 60 min	99.9	Hossain et al. (2)
3.	Activated carbon from Tamarind wood	100	1-6	1-5 g/L	10-50	10-50 mg/L	pH = 1, adsorbent dosage = 2g/L, T = 30°C, time = 40 min, , initial Cr(VI) concentration = 30 mg/L	96	Acharya et al. (3)
4.	Modified groundnut hull	100	1-8	10-50 mg	10-40	12.5-18 mg/L	pH = 2, initial Cr(VI) concentration = 13 mg/L, time = 60 min, T = 28°C	96	Owalude et al. (4)
5.	Paper mill sludge	100	2-6	1.5-7 g/L	20-45	10-100 mg/L	pH = 4; adsorbent dosage = 3.5g/L, time = 180 min, T = 45°C, initial Cr(VI) concentration = 10mg/L	98.4	Gorzin et al. (5)
6.	Treated waste newspaper	100	2-7	0.4-4g/100mL	–	5-70 mg/L	pH = 3, adsorbent dosage = 3g/L, time = 60 min	64	Dehghani et al. (6)
7.	Ocimum americanum L seed pods	50	1-6	–	–	100-200mg/L	pH=1.5, adsorbent dosage = 8g/L, time = 120min, initial Cr (VI) concentration = 50mg/L	100	Levankumar et al. (7)
8.	Fertilizer industry waste material	10	1-3	1-10g/L	30-50	10-100 mg/L	pH = 2, adsorbent dosage = 4 g/L, T= 30°C, time = 70 min, initial Cr (VI) concentration = 100 mg/L	65	Gupta et al. (8)
9.	Oak wood charcoal	40	1-7	0.05- 0.6g	–	2 mM	pH = 2, adsorbent dosage = 0.2g, time = 120 min, T = 25±1°C, initial Cr(VI) concentration = 2 mM	75	Pehlivan et al. (11)
10.	Neem leaves	25	1-11	4-16g/L	-	20-700 mg/L	pH =1-3, adsorbent dosage = 10 g/L, T = 30°C, initial Cr(VI) concentration = 50 mg/L	99	Babu et al. (13)

S. No.	Adsorbent	Treated Volume, mL	Ranges of Process parameters studied				Optimum conditions	Cr(VI) removal (%)	References
			pH	Adsorbent dosage	Temperature (°C)	Initial Cr(VI) Concentration			
11.	Ragi husk	50	2-4	2-5g/L	25-30	50-150 mg/L	pH= 2, adsorbent dosage = 2 g/L, T = 30°C, initial Cr(VI) concentration = 150 mg/L;	68.6	Premkumar et al. (14)
12.	Pre-boiled sunflower stem	50	2-7	0.2 -1g	-	10-70 mg/L	pH = 2, adsorbent dosage = 1g/50mL, T = 26±1°C, initial Cr(VI) concentration = 50mg/L, time = 180min	54.8	Jain et al. (16)
13.	Modified walnut shell	20	2-9	0.02-0.20 g	25-65	0.1-1.0 mM	pH = 2, time = 120min, adsorbent dosage = 0.1 g, T = 25±1°C; initial Cr(VI) concentration = 1 mM	75	Altun et al. (17)
14.	Modified red pine saw dust (tartaric acid modified saw dust)	30	3-10	0.1g	-	0.1-1.0 mM	pH = 3, T = 25±2°C, adsorbent dosage = 0.1g, initial Cr(VI) concentration = 1 mM, time = 120min	87.7	Gode et al. (18)
15.	Bone charcoal	50	1-11	0.5-2 g	—	5-25 mg/L	pH = 1, adsorbent dosage = 2gm, initial Cr(VI) concentration = 10 mg/L, time = 30min	90	Dahbi et al. (19)
16.	Modified corn stalks	50	1.8-5.2	0.5-4 g/L	25-50	100-400 mg/L	pH = 2.3, adsorbent dosage = 1.5g/L, initial Cr(VI) concentration = 100mg/L, T = 25±1°C, time = 60 min	99.5	Chen et al. (20)
17.	Biomass of Agaricus bisporus	100	1-7	3-15 g/L	20-40	25-125 mg/L	pH = 1, adsorbent dosage = 10g/L, initial Cr(VI) concentration = 50mg/L, T = 20°C	92.4	Ertugay et al. (21)
18.	Husk of Bengal gram	100	1-7	1-40 g/L	—	10-100 mg/L	pH = 2, adsorbent dosage = 1g/100mL, initial Cr(VI) concentration = 10mg/L, time = 180min	99.9	Ahalya et al. (22)

Table 2: Overview on the reusability of the prepared adsorbents by various researchers for the removal efficiency of Cr(VI)

S. No.	Adsorbent	Frequency of reusability	Optimum conditions	Cr(VI) Removal efficiency (%)	References
1.	Tea waste	5	pH = 3.9, adsorbent dosage = 6 g/L, T = 30°C, initial Cr(VI) concentration = 141 mg/L, time = 240 min	90.5	Nigam et al. (1)
2.	Treated waste newspaper	5	pH=3, adsorbent dosage=0.4g/250mL, initial Cr(VI) concentration =20mg/L, time= 60min	78	Dehghani et al. (6)
3.	Paper mill sludge	3	pH = 4; adsorbent dosage = 3.5g/L, time = 180 min, T = 45°C, initial Cr(VI) concentration = 50mg/L	68.34	Gorzin et al. (5)
4.	Synthesized chitosan from crab shell	5	pH = 3, adsorbent dosage = 5g/100mL, T = 60°C, time = 60 min	78	Hossain et al. (2)
5.	Banana peel	10	pH =2, time = 30min, initial Cr(VI) concentration = 1.92×10^{-5} M	92	Memon et al. (15)
6.	Chitosan/MWCNTs-COOH composite	5	pH =2.0, adsorbent dosage = 0.5 g/500 mL, T = 30°C, time = 30min	91	Huang et al. (23)
7.	Chitosan functional gel including with multiwall carbon nanotube and substituted polyaniline	3	pH =2.0, adsorbent dosage = 0.5 g/100 mL, initial Cr(VI) concentration = 6 mg/L	85	Kim et al. (24)
8.	40% Aliquat 336/60% PVC PIM	5	pH =4.0, adsorbent dosage = 0.13 g/100 mL, initial Cr(VI) concentration = 48 mg/L	30.83	Gherasim et al. (25)
9.	Chitosan grafted GO nano-composite	10	pH =2.0, adsorbent dosage = 2 g/L, initial Cr(VI) concentration = 50 mg/L	82	Samuel et al. (26)
10.	AC of Cornulaca monacantha stem	5	pH =2.0, adsorbent dosage = 60 mg/L, initial Cr(VI) concentration = 20 mg/L, contact time = 120 min, T = 65°C	89	Sharma et al. (27)

In this context, Acharya et al. (3) have studied the effect of initial feed concentration of Cr(VI) using adsorption at constant temperature of 30°C and pH of 6.5 and found that % removal of chromium decreased from 88 to 63% (at 3 g/L adsorbent dosage) with an increase in initial concentration of Cr(VI) from 10 to 50 mg/L. A similar observation was also made by Babu et al. (13), the initial Cr(VI) concentration was varied from 40 to 700 mg/L at the adsorbent dosage of 10 g/L. They have found that the percentage Cr(VI) removal decreased from 99.95% to 89.94% when Cr(VI) concentration increased from 40 to 700 mg/L. However, literature have reported that at higher concentration of the chromium, the adsorption capacity increases (12, 13, 20). Therefore, it is need to be reported in terms of adsorption capacity. In this regard, Hasan et al. (12) found that adsorption capacity increased for an increase in the initial concentration of Cr(VI). A similar trend was also found by Babu et al. (13). They have reported that adsorption capacity increases from when initial

Cr(VI) concentration increases. From the above studies, it is clear that % removal of Cr(VI) gets reduced for an increase in the initial concentration whereas adsorption capacity was found to be higher at higher concentration. Therefore, it is required to report the data in terms of percentage removal as well as adsorption capacity.

3 Overview on the reusability of used adsorbent

In the area of wastewater treatment, reusability of the prepared adsorbent is the most significant step because of untreated amount in excess may led to the disposal problems (1, 5, 6). The adsorbent can be recycled. The economy of the adsorption process depends upon how many times the prepared adsorbents can be reused without sacrificing its percentage removal efficiency. Many authors have reported on the reusability study of the adsorbents for the hexavalent chromium removal in recent years (1, 23-27). Nigam et al. (1) have studied about the reusability of the tea waste as an adsorbent and found that the %

removal of Cr(VI) was removed from 96.7% to 90.5% after five repeated cycles. Samuel et al. (26) have investigated the reusability test of the chitosan grafted graphene oxide (CS-GO) nano-composite and reported that the % removal of Cr(VI) was decreased from 96% to 82% after ten cycles. Sharma et al. (27) reported that the adsorption efficiency for Cr(VI) removal was dropped to 89.19% after five consecutive cycles. Kim et al. (24) have observed that the CS-MWNT-PAA-PADPA/FG showed nearly 85% removal efficiency of Cr(VI) even after three consecutive adsorption-desorption cycles. An overview on the reusability of used adsorbents reported in the literature is summarized in Table 2. Overall, it can be concluded that the percentage Cr(VI) removal decreases with an increase in the regeneration cycles of the used adsorbents.

4 Conclusions

The influences of various adsorption process parameters on the removal efficiency of Cr(VI) from wastewater using various adsorbents have been reviewed. There are some conclusions from this review as following:

- The percentage removal of Cr(VI) depends upon the solution pH and maximum removal efficiency of Cr(VI) from wastewater is found under acidic conditions. The optimum pH range for all adsorbents used in the current work is 1.0-4.0.
- Adsorbent dosage should be optimized for newly developed materials to achieve the maximum removal efficiency of Cr(VI) from wastewater because every prepared adsorbent have different adsorption capacity.
- It is needed to study the influence of temperature in the adsorption process for identifying the nature of adsorption/reaction (either endothermic or exothermic).
- From literature, lower concentration is more favorable for achieving the higher removal efficiency of Cr(VI) whereas adsorption capacity is found to be higher at higher initial Cr(VI) concentration.
- It is seen that reusability of the prepared adsorbent helps in the disposal problems and economy of the adsorption process.

Competing interests

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

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