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# Adsorption of Hexavalent Chromium Cr(VI) by Using Local Jordanian Sand (LJS) and Iron Dust-Sand Mixed Adsorbents

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#### Abstract

Hexavalent chromium is a known carcinogen. It is generated during the electroplating, leather tanning, mining, and photography industries cause harm to the environment and water resources. An attempt for cost effective adsorbent was made by utilizing sand and iron-sand mixed adsorbents. Sand adsorbent was found to exhibit remarkable adsorption capacity for hexavalent chromium. The extent of Cr(VI) removal was found to be dependable on the solution pH, Cr(VI) initial concentration, contact time, sand adsorbent dosage, and the iron content in iron-sand mixed adsorbent. The optimum pH for the removal was found to be 2. The highly removed concentrations by sand adsorbent ranged from 0 to 40 mg/l. It was found that as contact time increases the removal efficiency of Cr(VI) increases. Variation in sand adsorbent dosages indicated that as the adsorbent dosage increases the removal efficiency increases and it was reached to 65% at 40 hours. On the other hand, the effect of iron mixed with sand adsorbent was examined at two stages; first, at low Cr(VI) concentration solution of 20 mg/l, and, second, at high initial concentration of Cr(VI) up to 1000 mg/l. By using the optimal iron dosage, the removal efficiencies were increased to 99% within 1 to 3 hours at 20, 61, 171, 345 and 1000 mg/l of Cr(VI) initial concentrations.

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Keywords: Hexavalent chromium, adsorbent, adsorption, Sand adsorbent (LJS), iron-sand mixed adsorbent, removal.

#### 1. Introduction

Wastewater from metal finishing industries contains contaminants such as heavy metals, organic substances, cyanides, and suspended solids, at levels which are hazardous to the environment. One of these heavy metals pollutants generated from the industry is hexavalent chromium [Cr(VI)]; the pollutant of greatest concern because of its toxicity (Kirk and Othmer, 1980; and Costa and Klein, 2006). Concentrations as low as 0.5 ppm in solution and 5 ppm in soils can be toxic to plants; in contrast Cr(III) is generally only toxic to plants at very high concentrations and is necessary in animal nutrition. Chromate or Cr(VI) is 100 times more mobile and more toxic than Cr(III), (Sang et al., 2002). A number of treatment methods for the removal of metal ions from aqueous solutions have been reported, mainly reduction, ion exchange, electrodialysis, electrochemical precipitation, evaporation, solvent extraction, reverses osmosis, chemical precipitation and adsorption, (Patterson, 1985).

One potential strategy is to use low-cost natural materials as sorbents for the contaminants of concern. Due to their low cost these materials could be disposed of directly when their sorptive capacity is exhausted rather than regenerating and reusing them (Bajpai, 2001; Matin et al., 2003; and Vikrant and Pant, 2006).

Studies conducted by Schmidt, 1977 and Ellis, 1985 revealed a significant removal of water-borne heavy metals by sand filtration. These preliminary findings were later confirmed by several workers, who thoroughly investigated the adsorption of heavy metals on sand and other similar materials. Ghanayem, 1989, reported that soil and soil materials such as clay minerals could play an important role in eradicating various heavy metals. Similarly, Muhammad, et al., 1997. found that passing wastewater containing heavy metals through a slow sand filter may produce effluents complying with the world health organization (WHO) guidelines for drinking water. Very high removal efficiencies of Cr (VI) were also achieved by adsorption on kaolinite and illite (Ward, 1990). The transport and mobility of hexavalent chromium in soils were found mainly to be controlled by adsorption and reduction processes. Reduction of Cr (VI) is caused by the magnetite present in the soil as concluded by (Azizian, 1993).

Using iron in different states under especial techniques have been considered to reduce the content of chromium ions from industrial and wastewater effluents. Abdo et al. (1998) used scrap iron to reduce hexavalent chromium to trivalent chromium with simultaneous generation of electrical energy using a divided parallel plate cell and fixed bed electrodes. It was also reported that the reduction of hexavalent chromium to trivalent chromium using pure iron wire is effective in reducing Cr(VI) to Cr(III) under pH conditions of 2-3 as concluded by (Gould, 1982). Later, El-Shazly et al., 2005 studied the reduction of hexavalent chromium and its kinetics by using a fixed bed of scrap bearing iron spheres and they concluded that such technique reduces Cr(VI) to trivalent state successfully. Rodriguez and Martinez (2005) concluded that a high and fast reduction of hexavalent chromium from wastewaters was according to a study in a tubular electrochemical reactor utilizing the oxidation of iron to Fe(II) in acid solution to aid the overall process at different pH levels.

It is a matter of fact that searching for a new nonconventional water resource in Jordan has the first priority. This research will focus on examining a potential lower cost treatment technique of the industrial wastewater for reuse applications or for a safe disposal of treated industrial wastewater.

#### 2. Materials and Methods

A local Jordanian sand sample (LJS) of 25 kg was prepared by washing and dried at 104°C and then used for all experimental investigations, while, the iron dust was obtained from a dust collection system of a steel shot blasting facility. A 3000 ppm stock solution of hexavalent chromium Cr(VI) was prepared by dissolving the required quantity of solid chromium trioxide in distilled water. Solutions of the desired concentrations were then prepared by diluting the appropriate volumes of the stock solution using distilled water. 1N hydrochloric acid was utilized for washing and rinsing of lab equipments used in the current study. Meanwhile 0.1N sulfuric acid and A lab grade of 0.1M sodium hydroxide solution were used for adjustment of pH of Cr(VI) solutions investigated. Four sets of batch experiments were carried out to investigate the effect of the solution acidity, adsorbate initial concentration, adsorbent dosage and iron dust-sand ratio. All batch experiments were carried out at room temperature and a rotation speed of 150 rpm. An analytical digital balance (Mettler AE200, USA), with 0.1 mg readability and 205 gram capacity was used to weigh the required amounts of chromium trioxide as well as sand and iron dust.

Prior to analysis, samples of Cr(VI) solution were filtered through a 0.45  $\mu$ m membrane filter and their pH was adjusted to a value of 8.5 so that trivalent chromium would form an insoluble precipitate (i.e., chromium hydroxide, Cr(OH)<sub>3</sub>), which could then be easily removed, and thus the solution becomes with the lowest total chromium concentration. This acidity (8.5) is the optimum pH value at which the solubility of chromium hydroxide is minimal. The filtrate was then analyzed colorimetrically for the determination of the remaining concentration of Cr(VI) according to the Standard Methods for the Examination of Water and Wastewater, 15th edition and using a direct reading spectrophotometer (HACH, model DR2000) (APHA 1989). All reagents used were of AR grade (HACH, USA). A microprocessor pH meter (model pH 3000.WTW) was used to measure the pH.

# 3. Results and Discussion

# 3.1. Effect of solution pH

Figure (1) shows the variation of hexavalent chromium concentration with contact time at different starting pH values while Figure (2) represents the percentage removal with time at the same pH values.



The removal of Cr(VI) from solution increases as the solution pH decreases. This increase lessens as the pH increases until no removal achieved at pH values above 7. The top line in Figure (1) as well as the bottom one in Figure (2) represent a concurrence of four lines of pH values of 10, 9, 8, and 7, which reveals that almost no removal of Cr(VI) takes place at these pH values. At pH 6, there is a slight removal of Cr(VI). Furthermore, the results indicate that the rate of removal of Cr(VI) significantly increases at pH values below 3 until it reaches maximum at pH 1 while the time required to reach equilibrium conditions increases. For example, a removal percentage of 5% was achieved at pH 6, while 45% removal was obtained at solution pH 1, at contact time of 3 hours (see Figure 2).





It is clear from that the maximum attainable removals of Cr(VI) occur at pH range of 1 to 2, which indicates that no significant removals are achieved by lowering the pH further, as shown in Figure (3).

#### 3.2. Effect of contact time

At a given pH, as can be seen in Figures (2) and (1), as the contact time increases the remaining Cr(VI) concentration rapidly decreases. As time elapsed, this behavior becomes less pronounced until a state of equilibrium is reached. Similarly, both the removal efficiency and chromium uptake firstly increase as treatment time increases until equilibrium conditions are approached, which means that further increase in contact time results in an insignificant metal removal or uptake. This practically implies no more chromium removal takes place (see Figure 2). The percentage removal of Cr(VI) from aqueous solution increases rapidly and reaches a removal efficiency of 50% within 5 to 6 hours from the beginning of treatment process which implies that most of removal was achieved first, After that time, the percentage removal of Cr(VI) increases slowly by time increasing, reaches a maximum of 65% till 40 hours. This means that a further increase in contact time has a negligible effect on the percentage removal or metal uptake.

#### 3.3. Effect of Cr(VI) initial concentration

The remaining Cr(VI) concentration was plotted against contact time as shown in Figure (4). As Cr(VI) initial concentration decreases the remaining concentration at equilibrium conditions decreases. Most of the removal of Cr(VI) occurred within the first 6 hours of the experiment.



As exhibited by Figure (4), the high removal of Cr(VI) at low initial concentration could be due to the availability of a sufficient number of free adsorption places onto adsorbent surface compared to the concentration of the adsorbate, which necessarily lead to prompt chromium removal from solution. Minor reduction in total removal efficiency (2.5%) is observed at 40 mg/l. As the initial concentration increases, the number of vacant adsorption sites becomes progressively less sufficient and subsequently the removal of the metal considerably decreases. Furthermore, as evident by Figure (4) equilibrium conditions (i.e., maximum chromium removal) are approached faster as the initial concentration of Cr(VI) increases, probably because the concentration of adsorbate specie tends to far exceed the available adsorption sites, which are likely become completely occupied by chromium specie, thus, decreasing the driving force for further adsorption.

Table 1: shows the removal percentage (Rem  $\ddot{}$ ), metal uptake, sand capacity and the remaining concentrations of Cr(VI) at equilibrium conditions.

Table (1): Removal percentage (Re %), metal uptake (q,), adsorption capacity and equilibrium concentrations for sand adsorbent using different Cr(VI) initial concentrations (20, 40, 60, 80 and 100 mg/l).

Co Ce (mg/L) (mg/L)		Removal Efficiency (R <sub>em</sub> %)	Metal uptake(q <sub>t</sub> ) (mg/g)	Adsorption Capacity (L/g)*	
20	7	65	0.104	0.008	
40	15	62.5	0.200	0.008	
60	29	51.67	0.240	0.008	
80	58	24.44	0.176	0.008	
100	90	10	0.080	0.008	



Removal efficiency (%) versus Cr(VI) initial concentration (mg/l) was illustrated in Figure (5). The highest removal percentages occurred in the range of 20 to 40 mg/l of Cr(VI) initial concentrations, and this may be due to the increase in the adsorption gradient which is generated between the adsorbent as solid phase and adsorbate (i.e, Cr(VI) solution) as a results of high availability of chromium ions in liquid phase. This implies that, high adsorption capacity is considered to be maximized at a Cr(VI) initial concentration of 40 mg/l, similar results were obtained by Santhy and Selvapathy (2004) who studied the removal of heavy metal adsorption onto activated carbon. The low removals of hexavalent chromium observed at high initial concentrations may be attributed to the limited number of adsorption sites available at the sand surface which is the key factor in the treatment process rather than Cr(VI) initial concentration. The repulsion among the chromium species at high initial concentrations may also play a role in the reduced removals obtained at high initial concentrations. These results are in agreement with those reported by (Lalvani et al., 1998; Santhy and Selvapathy, 2004; Baig et al., 2003; and Banerjee et al., 2004). As the initial concentration further increases, the removal efficiency rapidly declines until it reaches about 10% at an Cr(VI) initial concentration of 100 mg/l. This results in agreement with the findings reported by Lalvani et al. (1998) and Santhy and Selvapathy (2004). A plot of the metal uptake against the initial concentration is shown in Figure (6).



Such plot has a prime importance in practice, since it provides means of finding the optimum combination of initial concentration and contact time in order to achieve the removal of Cr(VI) required to comply with effluent standards. Figure (6) shows that, at first, the uptake varies linearly with the initial concentration until a maximum value is attained where further increase in the initial concentration results in a substantial reduction in the uptake. As shown, the metal uptake increases with the initial concentrations from 20 to 60 mg/l and then it starts to decline at higher Co values. This means that the maximum uptake of sand adsorbent is 0.248 mg/g at initial solution pH of 2 and an initial Cr(VI) concentration of 60 mg/l.

# 3.4. Effect of adsorbent dosage

Twelve sand adsorbent samples were tested. An initial adsorbent dose of 10 g/l was experimented and then gradually increased by 10 g/l each time up to 125 g/l. An initial solution pH of 2 was used throughout the ten experiments. The experimental results are displayed in Table (2).

Table (2): Adsorbent dose, Cr(VI) removal %, Equilibrium Concentrations (C<sub>e</sub>) and uptake (q<sub>t</sub>). [In batch reactor where Cr(V) Co is 20 mg/L, pH is 2 and liquid volume is 2 liters].

Experiment No.	Sand dose (g/l) (1)	Amount of sand (g) (Adsorbent) (m <sub>6</sub> ) (2)	Equilibrium concentration (Ce) (mg/l) (3)	Removal % (4)	Uptake adsorbed of Cr (IV) qt = (Ci-Ce)*V/ms (mg /g) (5)
1	10	20.00	18.00	10.00	0.200
2	20	40.00	16.70	16.50	0.165
3	30	60.00	15.00	25.00	0.166
4	40	80.00	14.00	30.00	0.150
5	50	100.00	13.00	35.00	0.140
6	60	120.00	12.00	40.00	0.133
7	70	140.00	11.00	45.00	0.128
8	80	160.00	9.80	51.00	0.127
9	90	180.00	8.70	56.50	0.125
10	100	200.00	8.40	58.00	0.116
11	112.5	225.00	7.60	62.00	0.110
12	125	250.00	7.00	65.00	0.104

Metal uptake of chromium was maximized at low adsorbent doses. A metal uptake of 0.200 mg/g sand was obtained using 20 g/l of adsorbent compared to an uptake of 0.104 mg/g sand by a dose of 125 g/l. The literature reviewed reveals that the metal uptake was found to decrease with increasing adsorbent dose, which is in compliance with the results of the present study (Fadali et al., 2004 and Gupta and Babu, 2006). As seen in Figure (7), there should be a critical adsorbent dose that results in a maximum removal efficiency, which ought to be seeked for a given adsorbate concentration, (Fadali et al., 2004; Gupta and Babu, 2006; and Santhy and Selvapathy, 2004).



# 3.5. Effect of iron dose

Due to the previously demonstrated incompatibility of sand as an adsorbent for highly concentrated hexavalent chromium solutions, iron dust (in certain proportions) was mixed with sand in order to enhance the removal of Cr(VI). Iron and its compounds are the most commonly encountered reductants for the treatment of Cr(VI)-containing liquid effluents (Carl and PuIs, 1994; Chang, 2004; Eary and Rai, 1988; and Scott et al., 1998). Two sets of experiments were carried out. In the first set, a low initial concentration was tested while in the second one highly concentrated solution was treated. The overall iron-sand dose was always kept constant at 125 g/l throughout the experiments. The concentrations of the solutions treated and the iron doses studied as well as the results obtained at Cr(VI) initial concentration of 20 mg/l are displayed in Table (3). Table (3): Effect of iron dust dose on Cr(VI) adsorption. [Cr(VI) C<sub>o</sub> is 20 mg/L, solution pH of 2]

				Iron cont	ent			
	Mixture No 1	Mixture No 2	Mixture No 3	Mixture No 4	Mixture No 5	Mixture No 6	Mixture No 7	Mixture No 8
Iron dose (g/L)	6.25	12.50	18.75	25.00	31.25	37.50	43.75	50.00
Iron amount in 2L (g)	12.50	25.00	37.50	50.00	62.50	75.00	87.50	100.00
Time (Hours)			Hexavalen	t Chromiun	n Concentra	tion Cr(VI)	(mg/L)	
0	20	20	20	20	20	20	20	20
1	1.00	0.90	0.70	0.30	0.10	0.09	0.08	0.05
2	0.80	0.80	0.30	0.20	0.09	0.08	0.08	0.04
3	0.70	0.50	0.30	0.10	0.90	0.08	0.05	0.02
4	0.70	0.40	0.20	0.09	0.07	0.07	0.05	0.01
5	0.60	0.30	0.10	0.09	0.07	0.07	0.04	0.01
6	0.40	0.30	0.09	0.09	0.05	0.05	0.03	0.01
10	0.30	0.20	0.09	0.06	0.04	0.04	0.02	0.01
20	0.15	0.10	0.08	0.06	0.04	0.03	0.02	0.01
35	0.15	0.10	0.08	0.06	0.04	0.03	0.02	0.01

It was founded that Cr(VI) removal of 96% at initial concentration (Co ) of 20 mg/l can be achieved in 2 hours by utilizing iron-sand system (dust dose 6.25 g/l) compared to about 65% removal in 40 hours using the same quantity of sand. The high removals obtained, in case of iron-sand system, can be mainly attributed to the chemical reduction of Cr(VI) to Cr(III) and adsorption of Cr(VI) by iron metal. In addition to its high reaction affinity to Cr(VI), iron dust (it is composed of very fine particles) expectedly provides very large reactive surface area, thus, enhancing further eradication of hexavalent chromium from solution. The reduction reaction of dichromate by iron is a heterogeneous reaction and pH dependent. It can also be quantitative and extremely fast (Carl and PuIs, 1994; Chang, 2004; and Eary and Rai, 1988).



Figure (8) shows that as the iron content increase in adsorbent mixture Cr(VI) in the overall metal removal increases with the enhancement of the removal being less pronounced at high iron doses. The flat part of the curve in Figure (8) at high iron contents shows that equilibrium conditions are reached or no reaction takes place due to the depletion of Cr(VI) ions.

Table (4) shows the effect of employing the hybrid system on the remaining concentration of Cr(VI) using an iron dose of 31.25 g/l and different initial concentrations (20-1000 mg/l). The iron dose of 31.25 g/l was chosen for treating highly concentrated solutions because higher doses result in an insignificant enhancement in the Cr(VI) removal as clearly seen from Table (3).

Table (4): Effect adsorption. [Bate gram iron & 93.7	t of iron th reactor, s 5 gram of s	dust mix w solution pH and)].	rith sand or is 2, and irc	n high concenti on-sand adsorbe	ration of Cr(V) nt dose is (31.2			
Time (Hours)	Hexavalent chromium concentration Cr(VI) (mg/L)							
0	20.0	61.0	171.0	345	1000			
1	0.8	1.0	1.2	2.2	4			
2	0.90	0.95	0.9	2	2			
3	0.70	0.82	0.4	1.8	1			
4	0.50	0.70	0.2	1.2	0.9			
5	0.02	0.03	0.81	1	0.8			
6	0.02	0.03	0.10	1	0.8			
10	0.02	0.03	0.10	1	0.8			
20	0.02	0.03	0.10	0.5	0.8			
35	0.02	0.03	0.10	0.5	0.8			
40	0.02	0.03	0.10	0.5	0.8			

As can be seen, the iron–sand system can also be the very effective for the treatment of highly concentrated Cr(VI) solutions. All Cr(VI) initial concentrations are reduced to at least 4% of their initial concentrations in the first hour of the treatment. Also, almost complete removals of Cr(VI) are a achievable for initial concentrations of 20 and 1000 mg/l in 3 and 4 hours respectively.

Figure (9) compare the removal efficiencies of Cr(VI) at different retention times by using sand and iron-sand systems at the same initial Cr(VI) concentration of 60 mg/l. Again, results confirm the advantages of using the hybrid system over the sole sand adsorbent.



The process of Cr(VI) removal by using different iron types was classified as a physiochemical process (Carl and PuIs, 1994; Gang et al., 2005; and Rodriguez and Martinez 2005). This implies that these types of processes involve both chemical reduction reaction and physical adsorption at the same time. Cr(VI) is usually adsorbed onto outside the surface of iron before being reduced in the presence of electrons donated by the metal (Chang, 2004; and Eary and Rai, 1988). Accordingly, iron can be considered as a good adsorptive material in addition to being a very strong reductant.

# 4. Conclusion

Several conclusions can be drawn from this work. Up to 65% removal of Cr(VI), from dilute solutions, could be achieved by using the sand investigated as an adsorbent. Cr(VI) removal was found to depend upon solution pH, contact

time, adsorbent dosage as well as Cr(VI) initial concentration. The removal of chromium was found to be pH dependent. Maximum removals were obtained at pH range of 1-2. The removal of Cr(VI) was found to initially increase with time elapsed until equilibrium conditions are approached. 50% of the removal occurs in the first five hours of treatment. The metal uptake was also found to similarly vary with increasing contact time. The removal efficiency of hexavalent chromium (i.e., adsorption and/or reduction-precipitation) was found to decrease with increasing Cr(VI) initial concentration over concentration range studied. In addition, it was found that the equilibrium concentration of Cr(VI) increases with increasing initial concentration. Furthermore, the metal uptake linearly varies with the increasing initial concentration up to a point after which it becomes inversely proportional to initial concentration. The removal efficiency of Cr(VI) was found to almost linearly vary with the adsorbent quantity whereas the metal uptake as well as Cr(VI) equilibrium concentration decrease with increasing sand dose

Much higher removals of Cr(VI) (up to 99%) could be gained by the iron dust to the adsorbing medium due to the reduction and different adsorption mechanisms of hexavalent chromium by iron metal.

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