

## Removal of airborne hexavalent chromium using alginate as a biosorbent

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**ABSTRACT:** Airborne hexavalent chromium has been classified as a human respiratory carcinogen and long term exposure has been known to cause ulceration and perforation of the nasal septum, bronchitis, asthma, and liver and kidney damage. Chromium electroplating plants are the major sources of atmospheric chromium and packed-bed scrubbers are the common control devices used to reduce emission of chromic acid mist from electroplating bathes. The feasibility of a new method to remove this pollutant using alginate beads as a biomass derivative was investigated by one factor at a time approach and Taguchi experimental design. Polluted air with different chromium mist concentrations (10-5000 µg/m<sup>3</sup>) was contacted to alginate beads (3.3-20 g/L), floating in distilled water with adjusted pH (3-7), using an impinger at different temperatures (20 and 35°C), and various velocities (1.2 and 2.4 m/s). Although there were no statistical significant differences between factor levels, the higher ions removal efficiencies were achieved at lower levels of air velocities, pollution concentrations, higher levels of pHs, temperatures, and sorbent concentrations.

**Keywords:** Alginat; Biosorbent; Chromium mist; Control; Electroplating; Novel system

### INTRODUCTION

Hexavalent chromium, Cr (VI), has been classified as a human respiratory carcinogen and allergen (Ashley *et al.*, 2003; Guertin *et al.*, 2005; Hara *et al.*, 2010; Nickens *et al.*, 2010). Long term exposure to airborne Cr (VI) has been known to cause ulceration and perforation of the nasal septum, bronchitis, asthma, and liver and kidney damages in exposed workers (Kumar *et al.*, 2005; Costa and Klein, 2006). Cr (VI) emission is associated with a number of industrial sources including metal plating, tanning, chromate ore processing and spray painting operation; combustion sources such as automobiles and incinerators (Ashley *et al.*, 2003; Park *et al.*, 2004; Babel and Opiso, 2007; Pandey *et al.*, 2010). Thus, controlling of chromium emission is an important issue

in both industrial hygiene and environmental protection (Nwachukwu *et al.*, 2010).

Chromium electroplating plants are the major sources of atmospheric chromium (Kuo and Wang, 2002; OSHA 2006; Tirgar *et al.*, 2007), and Packed-Bed Scrubbers (PBS) are the common controlling devices used to reduce emission of chromic acid mist from electroplating bathes. Such industries should spend considerable money for scrubber wastewater treatment. Treatment of industrial effluents having less than about 0.1 g/L of dissolved metal ions by conventional processes presents several disadvantage, such as high operational cost, low efficiency, and generation of solid waste-often toxic, that may require safety disposal protocols or even further treatment (Ibanez and Umetsu, 2002; Aliabadi *et al.*, 2006; Abdel-Ghani and Elchaghaby, 2007; Goyal *et al.*, 2008; Nameni *et al.*, 2008).

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The capacity of biological entities to uptake heavy metal ions from dilute aqueous solutions is well documented and it can be considered as an alternative to be used under conditions where conventional processes lost competitiveness (Park and Park, 2006; Rafati *et al.*, 2010; Sethuraman and Balasubramanian, 2010; Vinodhini and Das, 2010). The biological treatment could be an alternative method to clean-up industrial wastewaters containing heavy metals. However, these processes are very sensitive to the characteristics of the effluent, as temperature, pH and chemical composition.

The use of dead biomass and biomass derivatives (biosorbent materials) for removal of heavy metals from aqueous solutions has been widely studied in recent years. These systems are less expensive than the traditional physicochemical processes. They do not need nutrients and are resistant to the physical-chemical properties of heavy metal solutions (Araujo and Teixeira, 1997).

Alginate, an exopolymer extracted mainly from brown seaweeds, has been used for a long time in several industrial applications. Among recent application, alginate has been used as a cell immobilization material and as a biosorbent material for removal of divalent heavy metals, such as  $\text{Cu}^{2+}$ ,  $\text{Co}^{2+}$ ,  $\text{UO}_2^{2+}$  and  $\text{Zn}^{2+}$  from aqueous solutions. Alginate showed good metal sorption efficiency (Araujo and Teixeira, 1997; Veglio *et al.*, 2002).

There are few reports about airborne heavy metal removal using a biomass derivative. Thus, in this project, alginate beads efficiency (as a biosorbent) for airborne Cr(VI) removal was studied in Public Health School and Public Health Research Institute, Tehran University of Medical Sciences, Iran (2006-7). The aim of this work was to investigate the feasibility of developing a method to remove airborne Cr (VI) mist.

## MATERIALS AND METHODS

### Airborne Cr (VI) generation

The predominant constituent in chromium-electroplating bath is  $\text{CrO}_3$ , although these might be amount of additives (usually 1 %  $\text{H}_2\text{SO}_4$ ) used in shop (Kuo and Wang, 1999). Thus, an electroplating bath was prepared as the source of Cr (VI) mist emission in pilot scale (Fig. 1). Detail of this system which equipped with a special sampling chamber and had a homogenous airborne Cr (VI) atmosphere, has been reported in our previous paper (Tirgar *et al.*, 2006a).

### Chemicals

Sodium alginate [Sigma-Aldrich Co.] was used. All the other chemicals were analytical grade and purchased from Merck Co.

### Beads preparation

Calcium alginate beads were prepared by dropping a 3 % (w/v) sodium alginate aqueous solution to a

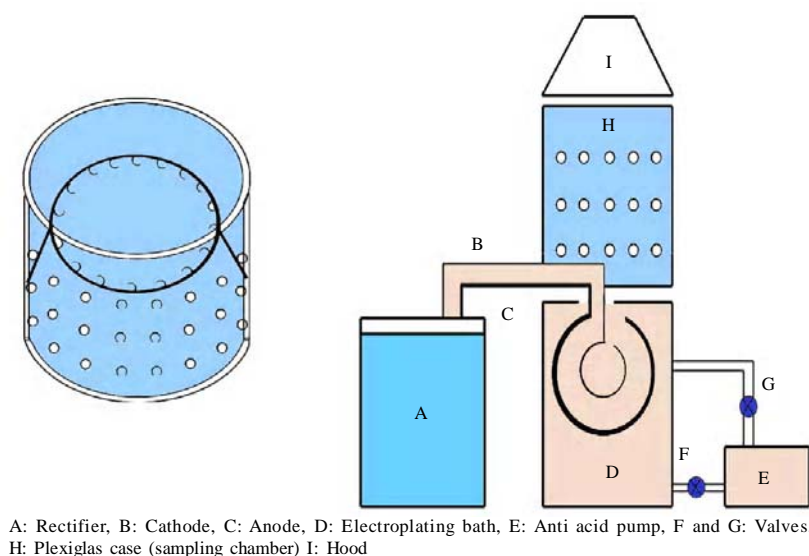


Fig. 1 : Set up of the mist generation system and sampling chamber



stirred 2 % (w/v)  $\text{CaCl}_2$  solution. Beads were stirred in  $\text{CaCl}_2$  solution on the orbital shaker at 250 rpm for 24 hours. Then, they were washed three times with distilled water and kept in a 2 %  $\text{CaCl}_2$  solution at 4 °C. Immediately before use, the calcium alginate beads were washed again three times with distilled water and the excess water was absorbed on paper filter (Araujo and Teixeira, 1997; Göksungur, *et al.*, 2003).

#### Bead characterization

The mean diameter of beads was determined in 10 groups of 50 alginate beads using a micrometer ( $2.87 \pm 0.06$  mm). The mean weight of the beads was determined using 10 samples of 20 beads ( $14.81 \pm 0.71$  mg). The solid content of the alginate beads were weighted before and after drying [ $7.04 \pm 0.32$  % (w/w)]. The biosorbent was dried in an oven at 60 °C over night (for 24 h).

#### Metal binding capacity

In order to find out optimum removal condition and Cr (VI) binding capacity, two sampling trains were used simultaneously (Fig. 2).

#### Sampling train with biosorbent

Sampling train with biosorbent (Biosorbent train) consisted of the following compartments: A) a special and standard air sampler named Midget Impinger (SKC, Blandford Forum, UK) containing alginate beads and 15 mL water with adjusted pH in thermostatically controlled water bath (Fig. 2). B) a 37-mm polystyrene

filter cassette (part B) containing a 5.0  $\mu\text{m}$  pore size polyvinyl chloride (PVC) membrane filter (MSA, Pittsburgh, USA). C) a personal sampling pump (Model 224-PCXR3; SKC, Blandford Forum, UK) calibrated for air flow rate of 1 or  $2.0 \pm 0.1$  L/min.

#### Sampling train without biosorbent

Sampling train without biosorbent (Water train) was completely the same as biosorbent train except Midget Impinger contained 15 mL only water with adjusted similar pH. Sampling duration for all samples were the same, i.e. 60 min.

The difference between chromium concentration in biosorbent train and water train was considered as alginate -bounded chromium. The percent of adsorbed chromium ions was evaluated as  $[(\text{CW}-\text{CB})/\text{CW}] \times 100$ . CW and CB are the concentration of the metal ions in the water and biosorbent solution (mg/L) after sampling, respectively.

#### Optimization of biosorption conditions by one factor at a time approach

Effect of pH on the biosorption rate was investigated in the pH range 3.0-7.0. The pH was adjusted with 0.1 M  $\text{HNO}_3$  or  $\text{NaOH}$  at the beginning of the experiments. The effect of pollutant concentration was studied at three different levels (low, medium, and high) between 10-5000  $\mu\text{g}/\text{m}^3$ . Also the effect of biosorbent concentration was determined at three different levels (3.3, 10, and 20 mg/mL).

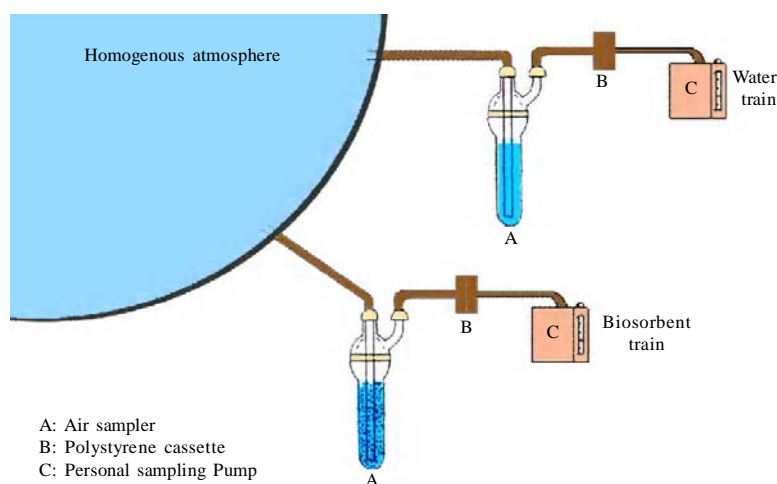


Fig. 2: Set up of the sampling trains



### Optimization of biosorption conditions by Taguchi method

To study the effects of important parameters on airborne Cr (VI) removal, five factors and two levels for each factor were studied by L16 ( $2^5$ ) Taguchi orthogonal array (Table 1) (Roy, 2001). The analyses of data were done by Qualitek-4 statistical software.

### Analytical method

The metal content in all experiments were measured using a Shimadzu AA 680 non-flame atomic absorption spectrometer. The instrument response was periodically checked with known standards. A calibration curve obtained with a correlation coefficient was 0.98 or greater. The samples were read three times and the mean value, as well as the relative standard deviation was computed. The samples were diluted as required to remain within the calibration linear range. The 357.7 nm wavelength was used for chromium ions studied (Cornelis, 1994).

## RESULTS AND DISCUSSION

### Primary optimization

Screening test results on pH, pollutant and sorbent concentrations using one factor at a time experimental design was presented in Fig. 3. The plots show the highest Cr (VI) removal obtained at pH 5, low pollution and medium sorbent concentrations. Other conditions including pH 6, medium pollutant and high sorbent concentration in the second preference of removal efficiency is considered to be used for secondary

optimization. The results presented below represent the mean values of at least three replications.

### Secondary optimization

Table 2 shows the descriptive data and the ANOVA test results for 64(16 runs with 4 replications) collected samples under the Taguchi experimental design. As indicated, there is no significant difference between the two levels of the main factors on Cr (VI) removal under the optimized levels ( $P>0.05$ ), also, there were no significant interactions between factors.

### Effect of pH

According to Park *et al.*, 2006. Cr (VI) can be removed from the aqueous solution by nonliving biomass through two mechanisms. In mechanism I (direct reduction), Cr (VI) is directly reduced to Cr (III) in the aqueous phase by contact with electron-donor groups of the biomass. However, reduction could occur only under strongly acidic conditions ( $\text{pH}<2.5$ ). Mechanism II (indirect reduction) consist of three steps: 1. The binding of anionic Cr (VI) ion species to positively charged groups present on the biomass surface; 2. The reduction of Cr (VI) to Cr (III) by adjacent electron-donor groups; 3. The release of the Cr (III) ions into the aqueous phase due to electronic repulsion between the positively charged groups and Cr (III) ions or Cr-binding. If there is a small number of an electron-donor group in the biomass or protons in the aqueous phase, the chromium bound on the biomass can remain in hexavalent state. Therefore, a portion of mechanisms I

Table 1: Removal percent of Cr(VI) by calcium alginate at different runs according L16 orthogonal array

Run	Air Velocity (m/s)	Alginate (g/L)	pH	Temperature ( $^{\circ}\text{C}$ )	$\text{Cr}^{6+}$ ( $\mu\text{g}/\text{m}^3$ )	$\text{Cr}^{6+}$ removal (%)
1	1.12	10	5	20	<50	36.90
2	1.12	10	5	35	50.500	32.35
3	1.12	10	6	20	50.500	41.67
4	1.12	10	6	35	<50	39.94
5	1.12	20	5	20	50.500	39.64
6	1.12	20	5	35	<50	35.92
7	1.12	20	6	20	<50	50.35
8	1.12	20	6	35	50.500	44.71
9	2.24	10	5	20	50.500	42.57
10	2.24	10	5	35	<50	44.00
11	2.24	10	6	20	<50	34.00
12	2.24	10	6	35	50.500	54.09
13	2.24	20	5	20	<50	30.74
14	2.24	20	5	35	50.500	36.63
15	2.24	20	6	20	50.500	34.60
16	2.24	20	6	35	<50	29.66



and II is dependent on the biosorption system, such as solution pH, temperature, biomass and Cr (VI) concentrations (Park *et al.*, 2006).

According to the speciation diagram of Cr (III) complexes in aqueous solutions, from Leyva-Ramos *et al.* (1995), at pH 2 the molar percentage of  $\text{Cr}^{3+}$  is 98 % of the total amount of chromium in solution, while at pH 4 this value decreases to about 40 % and almost 60 %

is in the ionic form  $\text{CrOH}^{2+}$  (and a trace amount of  $\text{Cr}(\text{OH})_4^{5+}$ ) (Araujo and Teixeira, 1997).

Pandey *et al.* (2003) reported, adsorptions of chromium by the calcium alginate beads suggest that the functional group present in calcium alginate are the carboxyl groups of gulucronate and mannurate residues (Pandey *et al.*, 2003). When the pH is higher than 3-4, the carboxyl groups are deprotonated and

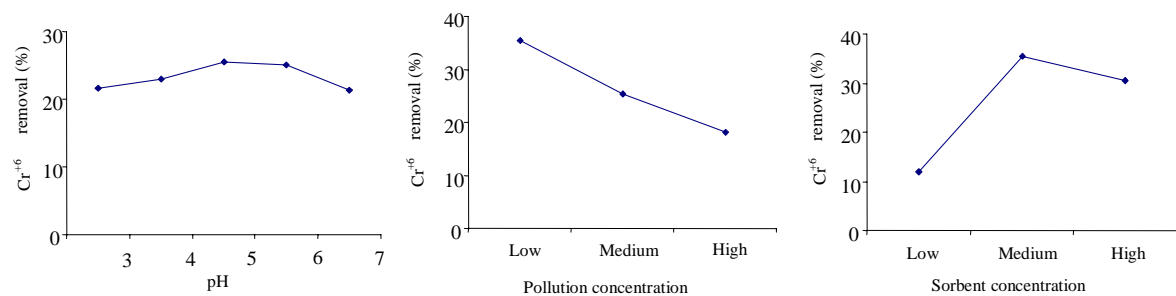


Fig. 3 Screening test results as a function of pH, pollution, and sorbent concentrations at 20°C and air velocity of 2.24 m/s

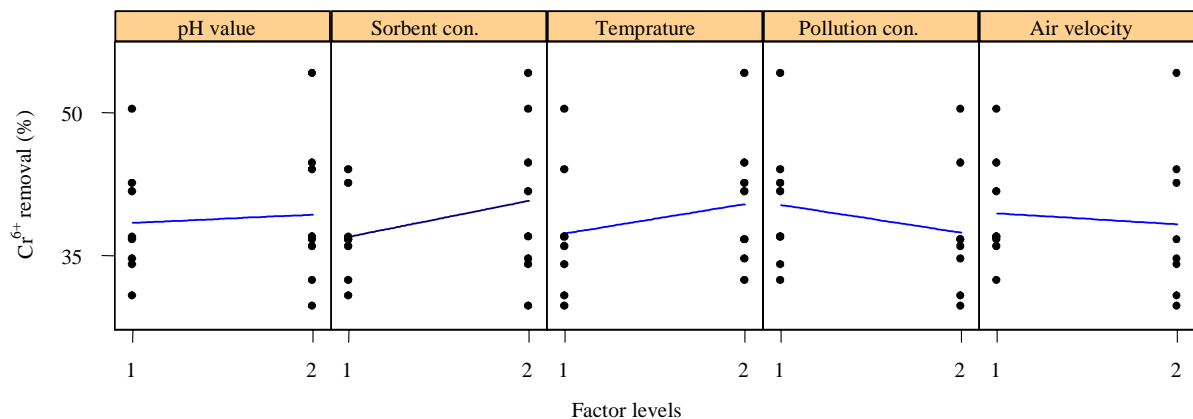


Fig. 4: Effect of air velocity, solution pH, sorbent concentration, pollution concentration, and temperature levels on Cr (VI) removal by alginate beads

Table 2: Descriptive data for 64 samples collected under Taguchi experimental design and its statistical test result

Factor	Levels	Cr (VI) removal (%)	SD	P-Value
pH	1	38.43	6.20	0.83
	2	39.29	7.89	
Temperature	1	37.31	6.86	0.46
	2	40.41	6.96	
Sorbent concentration	1	36.97	4.51	0.36
	2	40.75	8.51	
Pollution concentration	1	40.32	6.93	0.49
	2	37.41	6.93	
Air velocity	1	39.44	5.79	0.78
	2	38.28	8.17	



left with negative charge. Therefore, at pHs above 3–4, the negatively charged carboxylate group may attract the positively charged chromium ions consequently binding and removing the Cr (VI) ions from solution. At pHs lower than 3–4, the carboxyl groups become protonated and no longer attract the positively charged chrome ions (Gardea-Torresdey *et al.*, 1996).

Araujo and Teixeira studied the effect of pH on metal adsorption at 2–4 and reported that there was a higher trivalent chromium adsorbed at pH 4 (Araujo and Teixeira, 1997).

Gardea-Torresdey *et al.* (2002) reported the same result. They showed that the unmodified alfalfa biomass (this biomaterial binds predominantly through carboxyl ligands) and carboxyl ion exchange resin (WTO1S) displayed similar pH dependent trends and the optimal binding pH for these biosorbents was between 5 and 6 (Gardea-Torresdey *et al.*, 2002) (Fig. 4).

#### *Effect of sorbent concentration*

As Table 2 and Fig. 4 show, increasing alginate concentration between 3.3 to 20 mg/mL enhanced Cr (VI) removal, however, there is no significant difference between the chromium removal in concentrations 10 and 20 mg/mL ( $P > 0.05$ ). Chand *et al.* reported that, higher sorbent concentration gave the greater Cr (VI) removal percent. However, up to a certain level and beyond that, more or less constant removal was observed (Chand *et al.*, 1994).

Results in general, have revealed that the Cr (VI) removal from air is not strongly affected by the sorbent concentration at the range of 10 and 20 mg/mL.

#### *Effect of temperature*

The temperature of the adsorption medium could be important for energy demands mechanisms in metal biosorption. Energy-independent mechanisms are less likely to be affected by temperature since the processes responsible for biosorption are largely physico-chemical in nature (Kacer *et al.*, 2002).

As it shown in Table 2 and Fig. 4, maximum binding capacity values were mostly at 35 °C but the ANOVA test result shows no statistical significant difference between the two levels ( $40.41 \pm 6.96$  vs.  $37.31 \pm 6.86$ ) ( $P > 0.05$ ). Thus, the biosorption of Cr (VI) by alginate appeared to be temperature independent in the range of 20 to 35 °C.

Araujo and Teixeira reported that the higher temperature (in the range of 10 to 27) favors the sorption

of Cr (III) on calcium alginate beads (Araujo and Teixeira, 1997), while our results and the results reported by Kacar *et al.* (2002) indicated that adsorption of Cr(VI) ions on calcium alginate beads was not significantly dependent on temperature at range 15–35 °C (Kacer *et al.*, 2002). In nature, biosorption is largely physico-chemical and an energy-independent mechanism, therefore, it is less likely affected by temperature.

#### *Effect of air pollution concentration*

Airborne Cr (VI) with different concentrations ( $< 50$  and  $50\text{--}500 \mu\text{g}/\text{m}^3$ ) were treated to evaluate the influence of pollution concentration on sorption behavior of alginate beads.

As depicted in Fig. 4, although an increase in Cr (VI) concentration had no drastic effect on removal efficiency ( $40.32 \pm 6.93$  vs.  $37.41 \pm 6.93$ ), higher adsorption yields were observed at lower concentration of metal ions. Similar trend has been reported by other researchers (Chand *et al.*, 1994; Kacer *et al.*, 2002; Ahalya *et al.*, 2005; Tirgar *et al.*, 2006b).

#### *Effect of air velocity*

In order to investigate the effect of contact time and agitation rate on Cr (VI) removal, polluted air samples were passed through the sorbent medium with different air velocity. Fig. 4 and Table 2 show that Cr (VI) uptake decrease with the increase in air velocity, however, there is no statistical significant difference between Cr (VI) uptake at 1.12 and 2.24 m/s ( $P > 0.05$ ). Consequently, it can be concluded that, however agitation facilitates a proper contact between the metal ions in solution and biomass binding sites and thereby it promotes effective transfer of sorbate ions to the sorbent sites (Ahalya *et al.*, 2005), higher air velocity leads to reduction of contact time (passing faster through sorbent medium) and less adsorption.

## CONCLUSION

It can be concluded that, biosorption is an efficient process for the removal of Cr (VI) mist in a wide range of pollution concentration. Obviously, higher removal efficiency could be obtainable, using suitable alginate producer biomass or alginate derivatives.

Based on our experiments, the optimum condition with maximum yield for removal of Cr (VI) by alginate beads are as follows:

Lower levels of air velocity, and pollution concentration,





Higher levels of pH sorbent concentration and temperature.

Also, it was shown that proposed airborne Cr (VI) removal system can work in broad range of working conditions that makes it more feasible at industrial scale.

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