

# Kinetics of Cr(III) Adsorption on by-pass Cement Kiln Dust

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## ABSTRACT

Kinetic investigations were carried out to evaluate the applicability of bypass Cement Kiln Dust (CKD) as a low cost adsorbent to removal of chromium trivalent from wastewater of tannery effluents. The effects of various kinetic parameters were investigated using a batch process. The results indicated that, the contact time of around 60 min. was required to achieved equilibrium. The adsorption rate increases with increasing initial concentration of Cr(III) in the wastewater stream and processing temperature. The adsorption kinetics follows pseudo-second order rate equation better than the pseudo first order one. The results of the kinetics experiments showed that the adsorption of Cr(III) on CKD is an endothermic process and the activation energy of adsorption is  $1.867 \text{ kJmol}^{-1}$ . This small value of activation energy suggests that the adsorption easily occurs. That refers to the feasibility of the practical use of CKD as a low cost adsorbent for the removal of Cr(III) from aqueous systems.

**Keywords:** Adsorption, Cement Kiln Dust, Kinetic Analysis, Tannery Wastewater.

## 1. INTRODUCTION

Chromium is an important toxic material because it does not undergo biodegradation. This pollutant is introduced into natural waters by a variety of industrial wastewaters including those from textile, leather tanning, electroplating, and metal finishing industries. When accumulated at high levels, chromium can generate serious problems and, when concentration reaches 0.1 mg/g body weight, it can ultimately become lethal (Schneider, et al., 2007). Currently, the most common processes for elimination chromium are adsorption, reverse osmosis and chemical reactions that involve reduction and precipitation. Among them adsorption has been shown as a feasible alternative method for removing traces of chromium from wastewater. Although many different adsorbents were tried to remove chromium from wastewaters (Cooney, 1999; Youssef, 2004; and Schneider, et al., 2007).

Adsorption is by far the most effective and widely used technique for the removal of toxic heavy metals from wastewater. (Selvi et al., 2001). Owing to the high cost and difficult procurement of activated carbon, efforts are being directed towards finding efficient and low cost adsorbent materials. A variety of low cost materials like fly ash (Grover and Narayanswamy, 1982), wood charcoal (Deepak and

Gupta, 1991), bituminous coal (Kannan and Vanangamudi, 1991), bagasse and coconut jute (Chand et al., 1994), rice husk carbon (Srinivasan et al., 1988), peat (Brown and Allen, 2000), red mud (Gupta et al., 2001), Used black tea leaves (Houssain et al., 2005), activated carbon from sugar industrial waste (Fahim et al., 2006) and sugarcane bagasse (Khan and Mohamad, 2007) are have been tried.

As shown recently, research efforts have been directed towards the use of industrial waste as an adsorbent material in an attempt to minimize processing costs and with the protection of the environment and public health. Therefore, we have interested in the use of Cement Kiln Dust (CKD) as low cost adsorbent because of their high removal capacity for Cr(III) (Ibrahim and Abushina, 2008). Earlier investigations were mainly focused on the conformational aspects of their removal or sorption capacities. The kinetics is an important physicochemical factor to evaluate the basic qualities of an adsorbent as well as the application of the adsorption process, whereas there is no report about the kinetic study on the removal of Cr(III) by CKD. The prediction of the adsorption rate for a given system is the most important parameter for adsorber design with adsorbate residence time, ultimately controlled by the kinetics of the adsorption system (Eligwe, et al., 1999).

Various researchers have described the order of adsorptive reaction of metal ions in aqueous system using different kinetic models. The adsorptions of Hg(II), Cd(II) and Pb(II) on lignite, and Fe(II) on low rank brown coal were explained by simple first order kinetics with respect to the solution concentration (Eligwe and Okolue, 1994; Eligwe, et al., 1999). Traditionally, adsorptive reactions are described by pseudo- first order rate equation, based on the concentration of adsorbate in the adsorbent phase (Lagergren, 1898). Numerous studies reported Lagergren kinetics for metal sorption; for example, the sorption of Ni(II) onto Wollastonite and China clay (Sharma, et al., 1990, 1991) and Cr(IV) onto bismuth trioxide in aqueous solutions (Bhutani and Kumari, 1994). On the other hand, a pseudo-second order rate equation was also used to describe the adsorption of metal ions from aqueous solution onto peat (Ho, et al., 1995) and sludge solids (Tien and Huang, 1991).

In the present study, the kinetics of Cr(III) adsorption on CKD is discussed through batch experiments under the various conditions: initial concentration of Cr(III), and processing temperature. The factors affecting the rate constant and the adsorption capacity of this adsorption process were investigated with different kinetic models.

## **2. EXPERIMENTAL**

### **2.1 MATERIAL**

The experiment was carried out to remove chromium (III) from tanning wastewater effluents by using by-pass cement kiln dust (CKD) as an adsorbent. The cement kiln dust was collected from the (Al-Mergheb Portland Cement Factory, Al-Khoms city).

The analysis of the sample of CKD was made by X-ray fluorescence, the composition of the constituents is shown in Table.1.

**Table.1 The chemical composition of CKD**

Constituents	Composition*, (wt%)
$SiO_2$	19.58
$Al_2O_3$	3.6
$Fe_2O_3$	2.97
$CaO$	54.89
$MgO$	2.12
$K_2O$	2.32
$SO_3$	0.83
$Cl$	0.54
$Na_2O$	0.109
Other residues	13.041

\* Average values for four different batch samples.

The wastewater obtained from national tanneries effluents, this wastewater obtained after the tanning operation was filtered to obtain a tanning solution, which is to be further treated for the chromium (III) removal.

## 2.2 Methods

The most of chromium is discharged into aqueous solution from tannery effluents as Cr(III) and small content of Cr(VI). Also, CKD had no effect on the removal of hexavalent chromium, while it adsorbs trivalent chromium from solutions. Thus, before the addition of CKD to tanning solution, reduction of hexavalent chromium was carried out by using sodium bisulfite as a reducing agent and sulfuric acid (0.1 N). The sodium bisulfite was added in different amounts 0.5, 1 and 1.5 g to 50 ml of the tanning solutions sample, while sulfuric acid was gradually added, (Ayres, et al., 1994). The addition of sulfuric acid was continued until the reduction was completed as detected by oxidation reduction potential which was used as a sensor in the device titroprocessor (Type: Metrohm 682).

The experimental was carried out by batch process for the tanning solution obtained from tanneries effluents. The Cr(III) content in the tanning solution was determined using UV visible spectrophotometer (Unicam 8700), at a wavelength of  $\lambda = 540$  nm. Each sample was digested by adding 5 ml of nitric acid to 50 ml of the sample before measuring its chromium content by the spectrophotometer. Then the solution was evaporated on a hot plate to its volume becomes 15 ml. Further, 5 ml of concentrate nitric acid and 10 ml of concentrate sulfuric acid (95% wt.) were added and the solution was evaporated until dense white fumes of  $SO_3$  appeared. The solution was cooled and diluted to about 50 ml with distilled water. Then, the solution was heated top almost boiling to dissolve gradually salts and then cooled and diluted with distilled water to 100 ml after that, the solution was ready for chromium (III) measurement.

The initial concentrations of Cr(III) in tanning solutions were treated in an experimental were 2336 and 4320 mg/l respectively. These two tanneries batches are taking from two different national tanneries effluents wastewater.

In the determination of equilibrium adsorption isotherm 3 g CKD and 250 ml of the stock solution of Cr(III) (tanning solutions prepared above) were transferred a conical flask and agitated using an agitator operation at 1200 rpm for 60 min (the time required for equilibrium to be reached between trivalent chromium adsorbed and trivalent chromium in solution) at room temperature ( $25 \pm 2$  °C) and pH value 0.5. These conditions was found to be the optimum for chromium trivalent adsorption onto CKD in the previous experiments (Ibrahim and Abushina, 2008). The pH of the test solutions was adjusted using reagent grade dilute sulfuric acid. A pH meter (Model: HI 8417, HANNA Instrument) was used to measure the pH of solutions.

### 3. RESULTS AND DISCUSSION

#### 3.1 Adsorption rate

The amount of Cr(III) adsorbed,  $q_t$  (mg/g), was computed as follows:

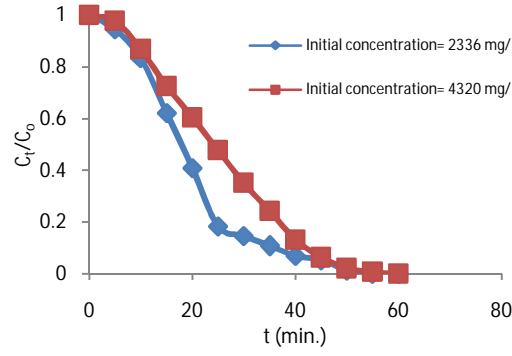
$$q_t = \frac{(C_o - C_t)V}{m_s} \quad (1)$$

where  $C_o$  and  $C_t$  are the concentrations of Cr(III) in the solution at  $t=0$  and  $t=t$ , (mg/L) respectively,  $V$  the volume of the solution (L) and  $m_s$  the weight of CKD used (g). Each experiment was performed twice at least under identical conditions. Reproducibility of the measurements was mostly within 2%.

In the experiments of batch kinetic adsorption, 250 ml of the chosen desired concentration of the stock solution of Cr(III) were placed in a conical flask together with 3 g CKD and agitation by agitator at temperature ( $25 \pm 2$  °C) and pH value 0.5. At predicted intervals of time, samples were taken, and their concentrations were determined.

The concentration of Cr(III) for different initial concentrations rapidly decreases during the first 50 min. as shown in Figure(1). The figure also shows that the equilibrium amount adsorbed depends on the initial concentration of Cr(III). The initial adsorption rate were calculated from  $q_t$  vs.  $t$  plot according to Eq.(2). The values increase with an increase in initial concentration of Cr(III) as shown in Table.1.

$$\left. \frac{dq_t}{dt} \right|_{t=0} = - \frac{V}{m_s} \left. \frac{dC_t}{dt} \right|_{t=0} \quad (2)$$



**Figure 1: Concentrations change of Cr(III) in the solution with time for different initial concentrations (3 g/250 ml dose, 25 °C temperature and pH 0.5)**

**Table 1.: The effect of initial concentration of Cr(III) on the initial rate of adsorption on CKD**

<i>Initial concentration</i> (mg/L)	<i>Initial adsorption rate</i> (mg/g)
2336	194.6667
4320	360.01

It is apparent that two different diffusion mechanisms played respective roles during this adsorption process. The first is a simple film diffusion driven by a large concentration difference between the bulk solution and the surface of the CKD, and the second is the surface diffusion of Cr(III) to the active sites. Thus, this behavior was tried to describe from the kinetic aspects based on the concentration of the bulk solution and also on the concentration in the adsorbent phase. Various kinetic evaluations have been developed for adsorption study. Here we attempted to apply a simple first order kinetic model (Eligwe et al., 1999) for changing the bulk concentration, and Lagergren's pseudo-first order rate equation (Lagergren, 1898) and Ritchie's pseudo-second order rate equation (Ho and McKay, 2000) for the adsorbent phase concentration, to describe the kinetic behavior of the system as follows:

### 3.1.1 Simple First Order Model

The sorption kinetics may be described by a simple first order equation (Hossain et al., 2005; and Eligwe et al., 1999). The change in bulk concentration of the system using the following simple first order rate Equation (3):

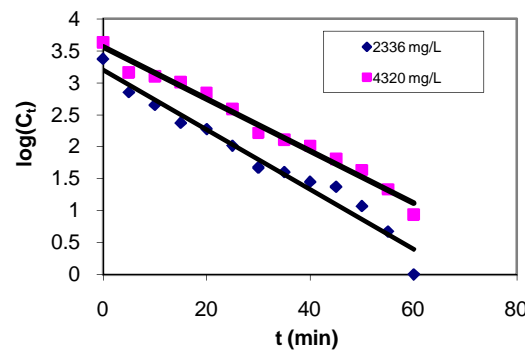
$$C_t = C_o e^{-k_1 t} \quad (3)$$

Equation (3) can be rearranged to obtain a linear form:

$$\log C_t = \frac{k_1}{2.303} t + \log C_o \quad (4)$$

Where  $C_t$  and  $C_o$  are the concentrations of chromium at time  $t$  and initially (mg/L), respectively, and  $k_1$  is the first order rate constant, (1/min.).

The experimental results showed that the  $\log C_t$  versus  $\log t$  (Figure.2) for different initial concentrations of Cr(III) were deviated considerably from the theoretical data. A comparison of the results with the correlation coefficient is shown in Table.2, which indicates the failure in expressing this adsorption process by the simple first order kinetics. On the other hand, the literature review (Sparks, 1989) has proposed that the simple kinetic models such as first- or second order rate equations are not applicable to the adsorption system with solid surfaces, which are rarely homogeneous like our CKD, because the effect of transport phenomena and chemical reactions are often experimentally inseparable. This trend was investigated also by (Hossain et al., 2005).



**Figure 2. Plot of the simple first order adsorption kinetics of Cr(III) on CKD at different initial concentration.**

### 3.1.2 Pseudo First-Order Model

The sorption kinetics may be described by a pseudo first order equation (Hossain et al., 2005; Özacar, 2003; Annadurai et al., 2002; Özacar and Şengil, 2002; Wu et al., 2001a-c; and Ho and Chiang, 2001). The differential equation is the following:

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (5)$$

After integration by applying the initial conditions  $q_t = 0$  at  $t = 0$  and  $q_t = q_t$  at  $t = t$ , Equation (5) becomes:

$$\log\left(\frac{q_e}{q_e - q_t}\right) = \frac{k_1}{2.303}t \quad (6)$$

Equation (6) can be rearrangement to obtain a linear form:

$$\log(q_e - q) = \log q_e - \frac{k_1}{2.303}t \quad (7)$$

where  $q_e$  and  $q_t$  are amounts of chromium adsorbed at equilibrium and at time  $t$  (mg/g), respectively, and  $k_1$  is the equilibrium rate constant of pseudo first-order adsorption, (1/min).

Figure (3) shows a plot of linearization form of pseudo first-order model at all concentrations studied. The slopes and intercepts of plots of  $\log(q_e - q_t)$  versus  $t$  were used to determine the pseudo first-order constant  $k_1$  and equilibrium adsorption density  $q_e$ . However, the experimental data deviated considerably from the theoretical data. A comparison of the results with the correlation coefficients is shown in Table 2. The correlation coefficients for the pseudo first order kinetic model obtained at all the studies concentrations were low. Also the theoretical  $q_e$  values found from the pseudo first-order kinetic model did not give reasonable values. This suggests that this adsorption system is not a pseudo first-order reaction. Lagergren's equation is suitable for homogeneous surface (Hossain et al., 2005), but the surfaces of CKD are not so homogeneous.

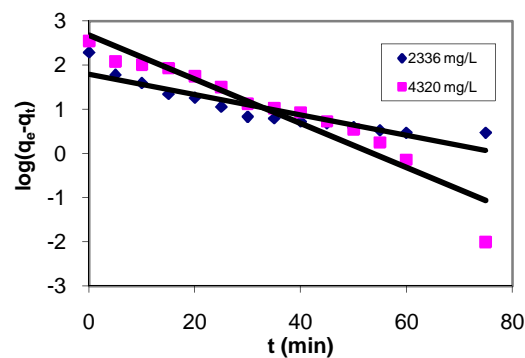


Figure 3: Plot of the pseudo first-order adsorption kinetics of Cr(III) on CKD at different initial concentration.

### 3.1.3 Pseudo Second-Order Model

The adsorption kinetics may also be described by a pseudo second-order equation (Özacar, 2003; Chiou and Li, 2002; Wu et al., 2001a-c; Ho and Chiang, 2001; and Ho and Mckay, 1999b). The differential equation is the following:

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (8)$$

Integrating Equation (8) and applying the boundary conditions, gives:

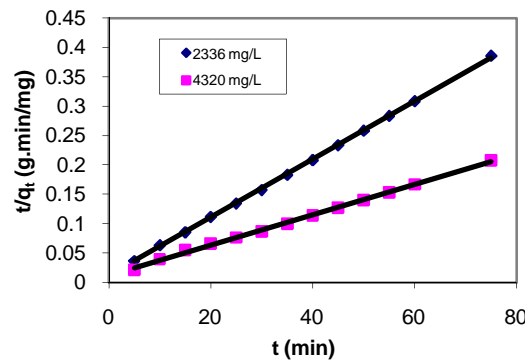
$$\frac{1}{(q_e - q_t)} = \frac{1}{q_e} + k_2 t \quad (9)$$

Equation (9) can be rearranged to obtain a linear form:

$$\frac{1}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (10)$$

where  $k_2$  is the equilibrium rate constant of pseudo second-order adsorption (g/mg.min).

The slopes and intercepts of plots  $t/q_t$  versus  $t$  were used to calculate the pseudo second-order rate constants  $k_2$  and  $q_e$ . The straight lines in plot of  $t/q_t$  versus  $t$  Figure (4) show good agreement of experimental data with the pseudo second-order kinetic model for different initial chromium concentrations. Table 2 lists the computed results obtained from the pseudo second-order kinetic model. The correlation coefficients for pseudo second-order kinetic model obtained were greater than 0.998 for all concentrations. The calculated  $q_e$  values also agree very well with the experimental data. These indicate that the adsorption system studied belongs to the second order kinetic model. The similar phenomena are also observed in adsorption of dye RR189 on cross-linked chitosan beads (Chiou and Li, 2002), in adsorption of dye BB69 and DR227 on activated clay (Wu et al., 2001b) and in adsorption of AB9 on mixed sorbents (activated clay and activated carbon) (Ho and Chiang, 2001) and in adsorption of Chromium hexavalent on used black tea leaves (Hossain, et al., 2005).



**Figure 4: Plot of the pseudo second-order adsorption kinetics of Cr(III) on CKD at different initial concentration.**

**Table 2. Comparison of the simple first order, pseudo first- and second-order adsorption constants, and calculated  $q_e$  values for different initial Cr(III) concentrations**

$C_o$ (mg/L)	$q_{e \text{ exp.}}$ (mg/g)	Simple 1 <sup>st</sup> order		Pseudo 1 <sup>st</sup> order			Pseudo 2 <sup>nd</sup> order		
		$k_1$ (1/min)	$r^2$	$k_1$ (1/min)	$q_{e \text{ cal.}}$ (mg/g)	$r^2$	$k_2$ (g/mg.min)	$q_{e \text{ cal.}}$ (mg/g)	$r^2$
2336	194.6667	0.09925	0.976	0.05297	63.1865	0.848	$2.12 \times 10^{-3}$	204.08	0.999
4320	360.01	0.0935	0.982	0.11492	483.931	0.914	$5.87 \times 10^{-3}$	384.615	0.998

$r^2$  is an regression coefficient



### 3.2 Effect of temperature

The effect of the processing temperature on the rate constant is important to determine the thermodynamic parameters to predict the adsorption behavior. Batch experiments were carried out at different temperatures specified in the range of 298 to 363 K with a constant concentration of Cr(III), 4320 mg/L, the amount of CKD 3g/ 250ml, and pH =0.5. Figure (5) shown that the obtained rate constant increases almost linearly with an increase in temperature. These experimental facts suggest that the adsorption process is controlled by chemical reaction, which is general for most of the chemically controlled adsorption processes. The activation energy of adsorption calculated from the Arrhenius' plot of the data is  $1.867 \text{ kJmol}^{-1}$  suggesting the endothermic nature of this adsorption. This small value of activation energy suggests that the adsorption easily occurs.

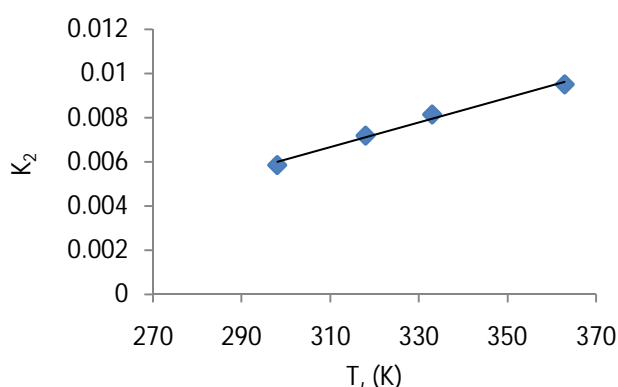


Figure 5 : The effect of temperature on the pseudo-second order rate constant ( $C_o=4320 \text{ mg/L}$ ,  $m_s=3\text{g}/250\text{ml}$  and  $\text{pH}=0.5$ )

### 4. CONCLUSIONS

The adsorption of Cr(III) on the by-pass cement kiln dust following the pseudo-second order kinetics is better than the pseudo-first order one. The adsorption rate increases with increase initial concentration of Cr(III). The rate constant increases linearly with an increase in temperature, suggesting that the endothermic process. The Arrhenius' plot of the rate constants gives a small value of activation energy, indicating that Cr(III) is easily adsorbed on the CKD.

The present study shows the feasibility of the practical use of by-pass cement kiln dust as a low cost adsorbent for the removal of Cr(III) from wastewater tanneries effluents.

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