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ORIGINAL ARTICLE

Kinetic study on adsorption of Cr(VI), Ni(II), Cd(II) and Pb(II) ions from aqueous solutions using activated carbon prepared from *Cucumis melo* peel

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Abstract
The adsorption of Cr(VI), Ni(II), Cd(II) and Pb(II), ions from aqueous solutions by *Cucumis melo* peel-activated carbon was investigated under laboratory conditions to assess its potential in removing metal ions. The adsorption behavior of metal ions onto CMAC was analyzed with Elovich, intra-particle diffusion rate equations and pseudo-first-order model. The rate constant of Elovich and intra-particle diffusion on CMAC increased in the sequence of Cr(VI) > Ni(II) > Cd(II) > Pb(II). According to the regression coefficients, it was observed that the kinetic adsorption data can fit better by the pseudo-first-order model compared to the second-order Lagergren's model with $R^2 > 0.957$. The maximum adsorption of metal ions onto the CMAC was found to be 97.95% for Chromium(VI), 98.78% for Ni(II), 98.55% for Pb(II) and 97.96% for Cd(II) at CMAC dose of 250 mg. The adsorption capacities followed the sequence Ni(II) ≈ Pb(II) > Cr(VI) ≈ Cd(II) and Ni(II) > Pb(II) > Cd(II) > Cr(VI). The optimum adsorption conditions selected were adsorbent dosage of 250 mg, pH of 3.0 for Cr(VI) and 6.0 for Ni(II), Cd(II) and Pb(II), adsorption concentration of 250 mg/L and contact time of 180.

Keywords *Cucumis melo* peel · Heavy metals · Adsorption · Chromium(VI) · Lead(II) · Cadmium(II) · Nickel(II)

Introduction

Heavy metal ions such as Cr(VI), Ni(II), Zn(II), Cd(II) and Pb(II) are among the toxic inorganic pollutants that may cause environmental problem, although at low concentration in surface or subsurface water (Gupta and Nayak 2012; Gupta and saleh 2013; Gupta et al. 2013, 2015; Jain et al. 2003; Vinod et al. 2012; Saleh and Gupta 2012a). The conventional method for removing these toxic metal ions from aqueous solution is by chemical precipitation and the major drawback of this method is the generation of huge amount of chemical sludge (Gupta et al. 1997, 2011a, b, 2012; Mohammadi et al. 2011; Ahmaruzzaman and Gupta 2011). Adsorption is a simple treatment method for removing metal ions in wastewater, and activated carbon is a powerful adsorbent that is commonly used in industrial wastewater treatment plant (Saleh and Gupta 2011; Saleh and Gupta 2012a, b; Karthikeyan et al. 2012; Mittal et al. 2009a, b, 2010, 2012; Wang et al. 2006, 2009). The growing interest in this material is motivated by its favorable surface properties, uniformity of adsorption, and consequently exceptional adsorption effect (Zuorro and Roberto 2010; Saleh and Gupta 2012b; Saleh and Gupta 2014a, b; Li et al. 2016; Devaraj et al. 2016). A wide variety of materials have been investigated for this purpose and they can be classified into three categories: (1) natural materials, (2) agricultural wastes, (3) industrial wastes (Saravanan et al. 2013a, b, c, d, e, f, 2014a, b, c, 2015a, b, c, d; Rajendran et al. 2016; Huang et al. 2015). These materials are generally available at free of cost (Pollard et al. 1992). Various naturally occurring materials having characteristics of an adsorbent are available in large quantities. The abundance of these materials are used in many continents of the world and their low cost make them suitable as adsorbents for the removal of various heavy metals from waste waters such as chitosan, peat, wood, natural coal, bentonite, sawdust, chitin, radish leaves and *Ricinus communis* (Santhi and Manonmani 2009; Hadi Khani et al. 2010).

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Thus, the present research aims to develop inexpensive and effective adsorbents using *Cucumis melo* peel a common agricultural waste, as an alternative to the existing commercial adsorbents. In addition, the present work is aimed to analyze the role of the surface chemistry in creating the activated carbon surface–metallic species interactions that govern the adsorption of Pb(II), Cd(II) and Ni(II) heavy metal ions. The adsorption behavior of heavy metals on CMAC was analyzed with Elovich, intraparticle diffusion model, Lagergren first-order model.

Materials and methods

Adsorbent

Preparation

Cucumis melo peel was collected from in and around pazhamudir nilayam of Coimbatore. The collected peels were cut into small pieces, washed with tap water several times to remove dust and dirt and rinsed with deionised distilled water and then dried. *Cucumis melo* peels were placed in the muffle furnace and carbonization was carried out at 200 °C for 2 h. The activated carbon thus obtained (here after CMAC) was ground well, sieved and the adsorbent of the size 75–125 µm has been used for the present study.

Experimental procedure

The adsorption experiment were performed in a batch mode in a series of beakers equipped with mechanical shaker by agitating 250 mg of the chosen adsorbent with 50 mL of metal ion solution with known previously determined, initial concentration of the considered heavy metal ion and the required initial pH value. The suspension was filtered and the remaining concentration of metal in the aqueous phase was determined. The final pH value was also measured. Batch adsorption experiment were performed by contacting 250 mg of the selected activated samples with 100 mL of the aqueous solution of different initial concentrations (100, 200, 300, 400 mg/L) of natural solution pH. The experiment were performed in a mechanical shaker at controlled temperature (25 + 2 °C) for a known period of time ranging between 10 and 210 min. At the end of the predetermined time, the suspension was filtered; the remaining concentration of Pb(II) in each sample after adsorption at different time intervals was determined by spectrophotometer after filtering the adsorbent with Whatman filter paper to make it carbon free. The batch process was used so that there is no need for volume correction. The Pb(II) concentration retained in the adsorbent phase was calculated according to

$$q_e = (C_i - C_e)V/W$$

where C_i and C_e are the initial and equilibrium concentration (mg/L) of Pb(II) solution, respectively. V is the volume and W is the weight (g) of the adsorbent.

Results and discussion

Adsorption kinetics

The study of adsorption kinetics describes the solute uptake rate, and evidently this rate controls the residence time of the adsorbate uptake at the solid–solution interface, including the diffusion process. The mechanism of adsorption depends on the physical and chemical characteristics of the adsorbents as well as on the mass transfer process. The results obtained from the experiments were used to study the kinetics of the metal ion adsorption. The rate kinetics of the metal ion adsorption on the CMAC was analyzed, using the pseudo-first-order (Lagergren 1898), Elovich and intraparticle diffusion (Weber and Morris 1963) models. The conformity between the experimental data and the model predicted values was expressed by the correlation coefficient (R^2).

Figure 1a–d displays the kinetic curves related to adsorption of Cr(VI), Pb(II), Ni(II) and Cd(II) on *Cucumis melo* peel-activated carbon. Equilibrium contact times from kinetic curves of heavy metals were determined. The optimum agitation time was fixed as 180 for all metal ions. These findings suggest that adsorption kinetic curves of heavy metals fitted the kinetic curve of the first degree.

The greater adsorption rate constant shows that adsorbates are adsorbed faster by the adsorbent.

According to adsorption rate constants, the heavy metal ions used are changing in the order of $Ni^{2+} > Pb^{2+} > Cd^{2+} > Cr^{2+}$. This order was confirmed by equilibrium contact times determined from their kinetic curves.

Adsorption rate constants (K_a) were determined using these curves.

Lagergren rate equation

The Lagergren rate constant for the adsorption of the Cr(VI), Cd(II), Pb(II) and Ni(II) metal ions from aqueous solution and from industrial effluent on the adsorbent CMAC is determined using Lagergren equation (Bhatnagar and Minocha 2006):

$$\log(q_e - q) = \log q_e - (K_a/2.303) \times t$$

where q is the amount of metal adsorbed (mg/g) by the adsorbent at time 't', q_e is the amount of metal adsorbed (mg/g) by the adsorbent at equilibrium time, K_a is the rate

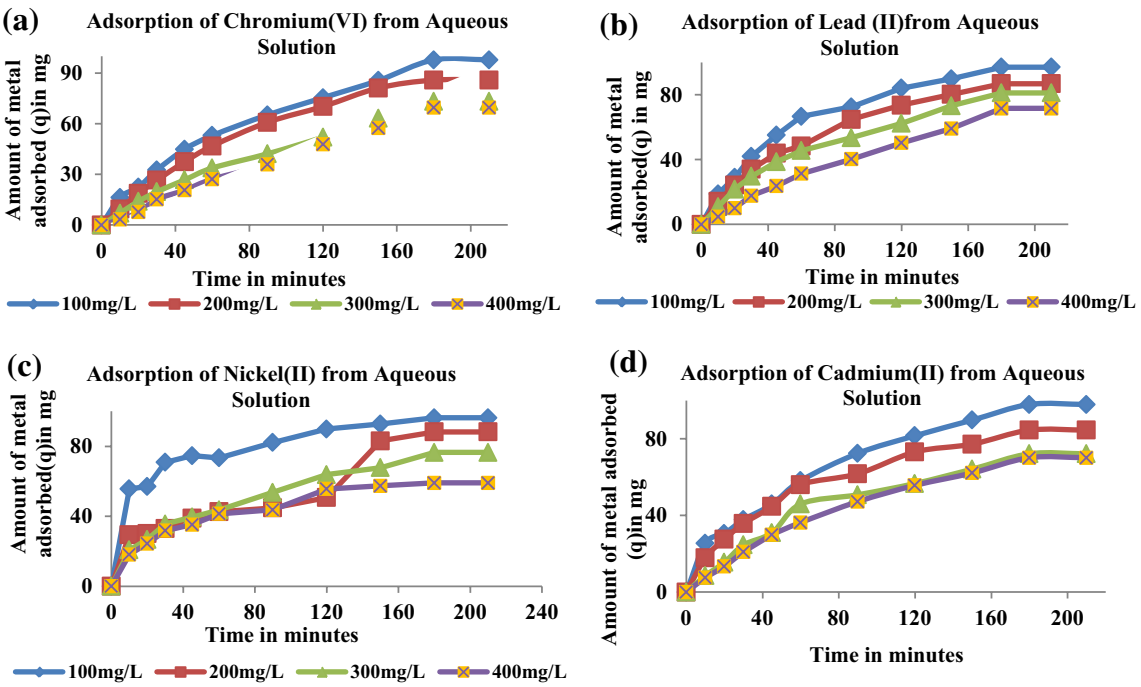

Fig. 1 **a** Kinetic curve related to adsorption of Cr(VI). From aqueous solution. **b** Kinetic curve related to adsorption of Pb(II). From aqueous solution. **c** Kinetic curve related to adsorption of Ni(II). From aqueous solution. **d** Kinetic curve related to adsorption of Cd(II). From aqueous solution

constant of adsorption in time^{-1} and t is the agitation time in minutes.

Experimental data for adsorption capacity of Cr^{6+}, Pb^{2+}, Ni^{2+} and Cd^{2+} from aqueous solution by activated carbon (Tables 1, 2, 3, 4).

The model has been widely used by researchers on kinetic study of metal ions adsorption onto various derived adsorbents (Rengaraj et al. 2003, 2004; Li et al. 2011; Argun and Dursun 2008). Batch mode experiments were carried out by varying the concentration of the metal solutions at 27 °C and at pH 3.0 for Cr(VI) metal ion and at pH 6.0 for Cd(II), Pb(II) and Ni(II) metal ions used in this study. The data obtained from Lagergren equation for the adsorption of the metals in terms of initial concentration of the metal solutions with the adsorbent CMAC. The linear plot obtained shows the applicability of Lagergren rate equation for the adsorption of the metal ions used in this study and suggested the formation of monolayer of metal ions onto the surface of the adsorbent (Madhava Rao et al. 2009). The rate constants K_a are calculated from the slope of the plots of $\log(q_e - q)$ vs t. The values of correlation coefficient (r^2) obtained show good correlation. The reason for this behaviour can be attributed to the high competition for the adsorption surface sites at high concentration which leads to lower adsorption rates (Venkata Subbaiah et al. 2009). From the slope of each line, the rate constants were determined. Figure 2 shows the linear plots of $\log(q_e - q_t)$ against t. Table 5 shows the amount of adsorption equilibrium (q_e), Lagergren's rate constant (K_{ad}) and the correlation coefficient (R^2) which are derived from the Lagergren's equation.

Table 1 Kinetic modelling for adsorption of chromium(VI) metal from aqueous solution using Lagergren's equation (initial concentration of chromium(VI) solution)

Time in minutes	100 mg/L		
	q_e	$(q_e - q)$	$\log(q_e - q)$
10	16.33	81.63	1.9118
20	22.45	75.51	1.878
30	32.65	65.31	1.815
45	44.9	53.06	1.724
60	53.06	44.9	1.652
90	65.31	32.65	1.513
120	75.51	22.45	1.351
150	85.71	12.25	1.008
180	97.96	–	–
210	97.96	–	–
Intercept (log q_e)	1.986024		
q_e	7.286505		
Slope (K_a/2.303)	0.00574		
K_a in min^{-1} × 10^{-2}	1.322		
Correlation coefficient (r^2)	0.99577		

Table 2 Kinetic modelling for adsorption of lead(II) metal from aqueous solution using Lagergren's equation onto CMAC (initial concentration of lead(II) solution)

Time in minutes	100 mg/L		
	q_e	$(q_e - q)$	$\log(q_e - q)$
10	23.19	75.36	1.877
20	44.93	53.62	1.729
30	69.57	28.98	1.462
45	73.91	24.64	1.392
60	82.61	15.94	1.202
90	86.96	11.59	1.064
120	98.55	–	–
150	98.55	–	–
180	98.55	–	–
210	98.55	–	–
Intercept (log q_e)	1.881811		
q_e	6.565387		
Slope (K_a/2.303)	0.01006		
K_a in $min^{-1} \times 10^{-2}$	2.316		
Correlation coefficient (r^2)	0.95655		

Table 3 Kinetic modelling for adsorption of cadmium(II) from aqueous solution using Lagergren's equation (initial concentration of cadmium(II) solution)

Time in minutes	100 mg/L		
	q_e	$(q_e - q)$	$\log(q_e - q)$
10	25.51	72.45	1.860
20	30.61	67.35	1.828
30	37.76	60.2	1.779
45	45.92	52.04	1.716
60	58.16	39.8	1.599
90	72.45	25.51	1.407
120	81.63	16.33	1.213
150	89.8	8.16	0.912
180	97.96	–	–
Intercept (log q_e)	1.977797		
q_e	7.226807		
Slope (K_a/2.303)	0.00668		
K_a in $min^{-1} \times 10^{-2}$	1.566		
Correlation coefficient (r^2)	0.99294		

As shown in Table 5, the values of R^2 for the metal ion adsorption onto activated sludge are larger than 0.85 and higher compared to the adsorption of the metal ions onto dried sludge. This shows that the adsorption of Cr(VI), Ni(II), Pb(II) and Cd(II) onto CMAC was well fitted with the first-order Lagergren's model.

Table 4 Kinetic modelling for adsorption of nickel(II) metal from aqueous solution using Lagergren's equation (initial concentration of nickel(II) solution)

Time in minutes	100 mg/L		
	q_e	$(q_e - q)$	$\log(q_e - q)$
10	55.7	43.03	1.634
20	56.96	41.77	1.621
30	70.88	27.85	1.445
45	74.68	24.05	1.381
60	73.47	25.31	1.403
90	87.22	16.51	1.218
120	89.87	8.86	0.947
150	98.73	–	–
180	98.73	–	–
210	98.73	–	–
Intercept (log q_e)	1.691917		
q_e	5.429878		
Slope (K_a/2.303)	0.00585		
K_a in $min^{-1} \times 10^{-2}$	1.347		
Correlation coefficient (r^2)	0.97332		

Intraparticle diffusion rate equation

There is a possibility of transport of metal ion molecules from the bulk into pores of the adsorbent as well as adsorption at the outer surface of the adsorbent. The rate-limiting step in the adsorption may be either film diffusion or intraparticle diffusion. As they act in series, the slower of the two will be the rate-determining step. The possibility of the metal ion species to diffuse into the interior sites of the particles of adsorbent was tested with Weber–Morris equation given as follows (Lalitha et al. 2009):

$$q = K_p t^{1/2}$$

where 'q' is the amount of metal adsorbed (mg/g) by the adsorbent at time 't', K_p is the intraparticle diffusion rate constant and 't' is the time (agitation time) in minutes.

To study the diffusion process, batch mode experiments are carried out with the adsorbent CMAC at 27 °C and at pH 3 for Cr(VI) and at pH 6 for Pb(II), Cd(II) and Ni(II) by varying the initial concentration of the metal solutions used in this study.

The rate constant for intraparticle diffusion K_p for initial concentrations of the metal ion is determined from the slope of the linear equation drawn between square root of time ($t^{1/2}$) and the amount of adsorbate adsorbed (q).

If the intraparticle diffusion is the rate-controlling step, the plot should be linear and pass through the origin. It can be noticed from Fig. 2, the plots are linear but not passing through the origin and this deviation from the origin or near

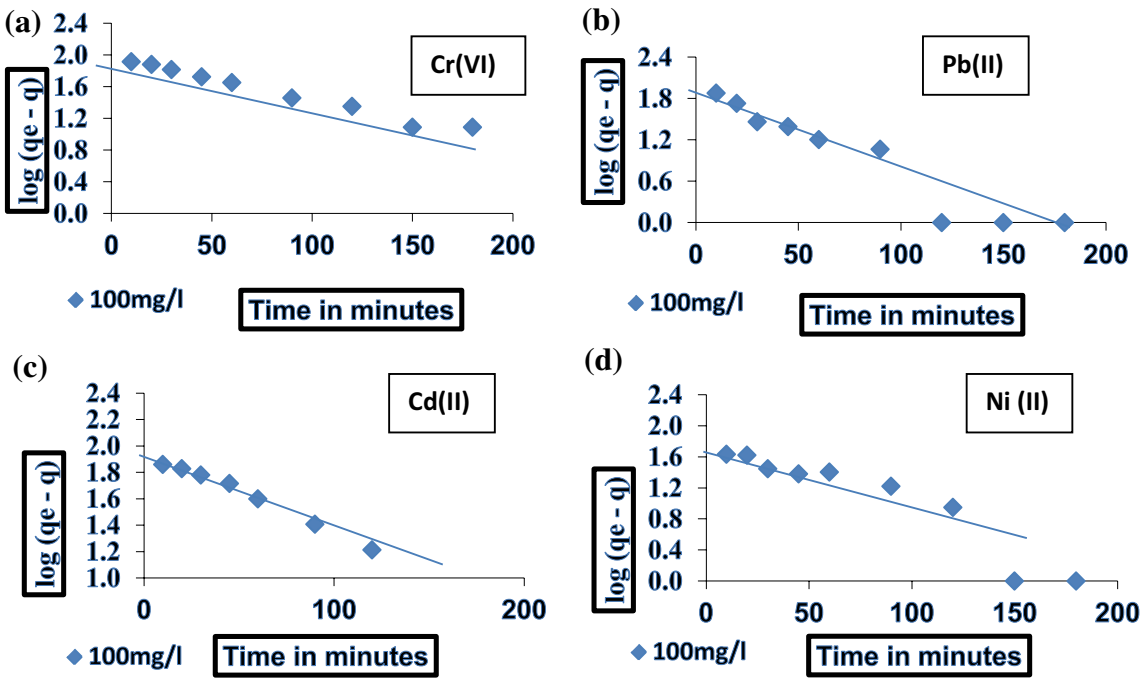

Fig. 2 First-order Lagergren (**a–d**) plots for CMAC on heavy metals adsorption

Table 5 Lagergren's rate equation

Adsorbent	Heavy metal	q_e (mg/g)	K_{ad} ($\times 10^{-2}$ min^{-1})	R^2
CMAC	Cr(VI)	7.29	1.32	0.996
	Pb(II)	6.56	2.31	0.957
	Ni(II)	5.43	1.35	0.973
	Cd(II)	7.23	1.57	0.993

Table 6 Intraparticle diffusion rate equation

Adsorbent	Heavy metal	K_p	R^2
CMAC	Cr(VI)	7.95	0.998
	Pb(II)	7.58	0.986
	Ni(II)	4.01	0.978
	Cd(II)	7.45	0.996

saturation might be due to the difference in mass transfer rate in the initial and final stages of adsorption (Renugadevi et al. 2011). It also indicates that there is an initial boundary layer resistance and intraparticle diffusion is not the sole rate-controlling step, but other kinetic models may simultaneously control the adsorption rate (Anoopkrishnan et al. 2011).

Table 6 shows intraparticle diffusion rate constant (K_{ad}) and the correlation coefficient (R^2) which are derived from the intraparticle diffusion rate equation. As shown in Fig. 3, the plot of intra-particle diffusion model given a straight line to the data obtained with R^2 greater than 0.9. This shows the adsorption of Cr(VI), Pb(II), Ni(II) and Cd(II) ions onto the CMAC was through the intra-particle diffusion process.

Elovich rate equation

The Elovich equation was developed to describe the kinetics of chemisorption of gases onto solids and it is generally expressed as:

$$dq_t/dt = \alpha \exp(-\beta q_t)$$

where 'q_t' is the amount of metal adsorbed (mg/g) by the adsorbent at time 't', 'α' is the initial adsorption rate (mg/g min) and 'β' is the desorption constant (g/mg) during any experiment.

Assuming the initial boundary condition, $q = 0$ at $t = 0$, the above equation on integration become:

$$1/q_t = 1/\beta \ln(1 + \alpha\beta t).$$

To simplify the Elovich's equation, Chien and Clayton (1980) assumed $\alpha\beta \gg 1$ and applying the boundary

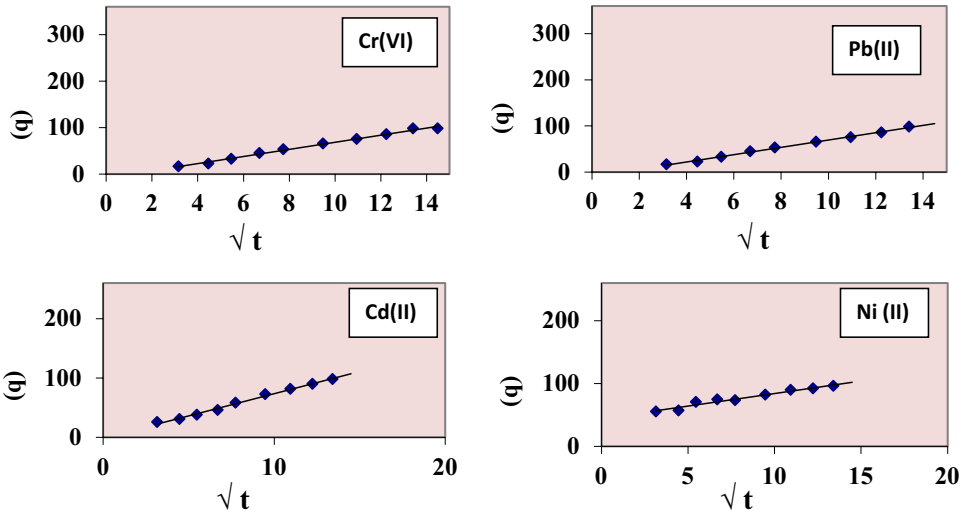

Fig. 3 Intra-particle diffusion plots for CMAC on heavy metal adsorption

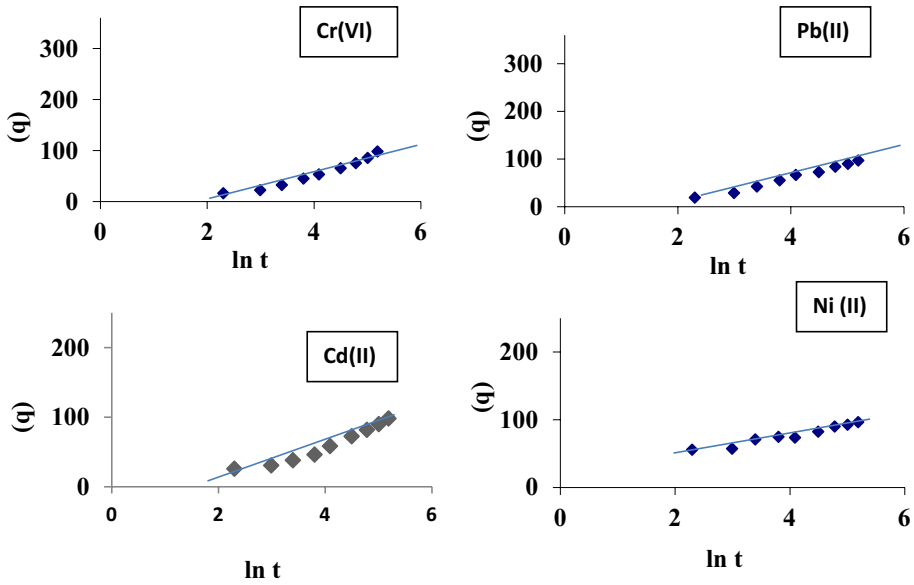

Fig. 4 Elovich's rate equation plots for CMAC on heavy metals adsorption

conditions $q_t = 0$ at $t = 0$ and $q_t = q_t$ at $t = t$, equation becomes (Patil et al. 2011):

$$q_t = 1/\beta \ln(\alpha\beta) + 1/\beta \ln t.$$

This equation is commonly used for the chemisorptions, which is probably the mechanism that controls the rate of adsorption. This model can be applied with success in liquid solution (Khaoya and Pancharoe 2012). Figure 4 shows the Elovich plots of ln t vs q (amount of metal adsorbed (mg/g) by the adsorbent at time 't'). It gives a linear relationship with a slope of $1/\beta$ and an intercept of $1/\beta \ln(\alpha\beta)$. The Elovich equation data obtained in this study for the adsorption of Cr(VI), Pb(II), Cd(II) and Ni(II) from aqueous solution onto CMAC. The high correlation coefficient (r^2) shows the successfulness of the Elovich model.

Table 7 Elovich's rate equation

Adsorbent	Heavy metal	β	α ($\times 10^2$)	R^2
CMAC	Cr(VI)	0.035	2.297	0.978
	Pb(II)	0.035	1.673	0.994
	Ni(II)	0.068	0.509	0.976
	Cd(II)	0.038	1.560	0.973

Table 7 shows desorption constant (β), initial adsorption rate constant (α) and the correlation coefficient (R^2) which are derived from the Elovich rate equation.

The higher value of initial adsorption rate (α) may be due to the greater surface area of the adsorbent CMAC, for the

immediate adsorption of metal ions from aqueous solution and also from industrial effluent. The decrease in the value of desorption constant (β) with the increase of the initial concentration of the metal solutions (Patil et al. 2011).

Conclusion

The adsorption behavior of Cr(VI), Pb(II), Ni(II) and Cd(II) ions onto the *Cucumis melo* peel-activated carbon was analyzed with Elovich rate equation, intra-particle diffusion model and Lagergren's first-order model. The kinetic adsorption data can fit better by the pseudo-first-order Lagergren's model and Elovich's rate equation indicating the rate-limiting step in the adsorption of Cr(VI), Pb(II), Ni(II) and Cd(II) ions could be ascribed to the chemical interaction between the metal ions and the functional groups at the surface of activated carbon. The Elovich's equation data obtained in this study for the adsorption of Cr(VI), Pb(II), Cd(II) and Ni(II) from aqueous solution onto CMAC. The high correlation coefficient (r^2) shows the successfulness of the Elovich's model.

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