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Study of Pb Adsorption by Carbon Nanofibers Grown on Powdered Activated Carbon

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Abstract: The sorption of lead (Pb) from aqueous solutions by using carbon nanofibers (CNFs) grown on nickel impregnated Powdered Activated Carbon (PAC) was studied. In this study, we investigated the affection of the lead initial concentration on the sorption of the heavy metal from water. The isotherm of the sorption of the heavy metal onto the nanocomposite was also studied. Firstly, the optimum pH for the sorption of the lead ions was determined. The maximum sorption capacity of the heavy metal onto the adsorbent was achieved at initial pH of 5.5. The effects of initial lead ions concentration were examined and the results showed that the adsorption capacities of the carbon nanofibers to uptake the heavy metal increased from about 16 to 89 mg g⁻¹ with increasing the initial lead concentration from 5 to 70 mg L⁻¹. The process sorption could be best fitted by the Langmuir isotherm. The equilibrium sorption capacities of lead ion were determined and found to be 100 mg g⁻¹.

Key words: Nanocomposite, impregnation, lead, carbon, Langmuir, Freundlich

INTRODUCTION

The development of industries such as coating, electric battery manufacturing, paint, lead smelting, internal combustion engines, generates large quantity of wastewaters containing large quantity of lead (Gueu *et al.*, 2007). This metal is potentially toxic as reported by Goyer and Chisholm (1972) and Assenato *et al.* (1986). So, its removal from industrial wastewaters remains an important challenge. For instance, in similar works, Duran *et al.* (2006) showed that a vermicompost allows to uptake 123.5 µg g⁻¹ of lead.

Sekar *et al.* (2004) used an activated carbon from coconut shell to adsorb 26.5 mg g⁻¹ of lead from aqueous solution and in another study, Gueu *et al.* (2006) also used activated carbon from coconut shell and seed shell of the palm tree to remove Pb from aqueous solution. The study showed that the maximum of lead cations uptake were at pH 4.

Abu-El-Halawa *et al.* (2003) used different dry plant leaves for the removal of lead from aqueous solutions and a removal of 88% of the heavy metal was achieved after 168 h.

Increasingly, stringent standard on the quality of drinking water has stimulated a growing effort on the

exploiter of new high efficient adsorbents. For potential environmental applications as superior sorbents, carbon nanomaterials have been studied for removal of metals (Li *et al.*, 2003).

With the emergence of nano science and technology in the last decade, research has been initiated to exploit the unusual and unique properties of carbon nanomaterials (CNMs). CNMs may exist in several forms, such as, single-walled carbon nanotubes (SWCNTs), multi-walled carbon nanotubes (MWCNTs), carbon beads, carbon nanofibres and nanoporous carbon (Ruparelia *et al.*, 2008). CNMs have been studied widely for potential applications in catalyst supports, optical devices, quantum computer and biochips. However, their sorption potential has not been studied extensively. CNMs are engineered materials targeted to exhibit unique surface morphologies, hence, they may prove to be good sorbents (Kurniawan *et al.*, 2006). It was reported that CNFs have high adsorption capacity and can be used as an adsorbent for removal of arsenic from water (Mamun *et al.*, 2009).

In this study, we report the use of CNFs and examined the properties of lead adsorption from aqueous solution in terms of initial pH and concentration.

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MATERIALS AND METHODS

The palm kernel shell granular activated carbon (obtained from Effigen Carbon Sdn Bhd, Malaysia) was grinded by using commercial grinder and the PAC of size between 100-250 μm was collected by sieving for the production of CNFs. The impregnation of PAC and the growth of CNFs were described elsewhere (Ahmed *et al.*, 2009). The prepared material was used as adsorbent. In each experiment, a 50 mL of the lead ion solution were mixed with a definite amount of the adsorbent. The mixture of the test solution and each of the adsorbents was shaken on JEIO mechanical shaker model SK-600 at 200 rpm. At the end of each experiment, adsorbents were filtered from the solution and the remaining lead concentration was analyzed using Atomic Absorption Spectrophotometer AAS (PerkinElmer HGA900). All experiments were carried out at room temperature. The effect of pH on adsorption of lead was studied at room temperature by varying the pH of lead solution from 2 to 10.5. The pH was adjusted with sodium hydroxide and HCl solutions.

Metal solutions of varying concentrations of lead ranging from 5 to 70 mg L⁻¹ adjusted to optimum pH 5.5 were treated with 0.005 g of CNF for 2 h at room temperature.

The equilibrium sorption isotherms express the relationship between the concentration of the sorbate and its degree of accumulation onto sorbent surface at constant temperature. The Langmuir isotherm (Langmuir, 1918) can be explained in the following linearized form Eq. 1:

$$\frac{C_e}{q_e} = \frac{1}{q_{max} \times b} + \frac{C_e}{q_{max}} \quad (1)$$

where, q_{max} is the monolayer sorption capacity and b (L mg⁻¹) is the Langmuir constant related to the free energy of sorption, independent of temperature.

The Freundlich sorption isotherm assumes that the uptake of sorbate occurs on a heterogeneous surface by multilayer sorption. This model can be adopted in the following linearized form Eq. 2:

$$\text{Log } q_e = \text{log } K_F + 1/n \text{ log } C_e \quad (2)$$

where q_e is the amount of Pb adsorbed per unit weight of the adsorbent (mg g⁻¹), C_e is the equilibrium concentration of solute in the bulk solution (mg L⁻¹), K_F is the multilayer sorption capacity and $1/n$ is the characteristic constant indicative of the intensity of the adsorption. The slope of the linear plot of log q_e against log C_e gives the value of $1/n$ and the intercept yields the value of K_F .

RESULTS AND DISCUSSION

The effect of pH is presented in Fig. 1 lead removal recorded its minimum values at pH 2. This can be justified on the basis that at lower pH values, the H⁺ ions compete with the metal cation for the adsorption sites in the system. This fact was approved by Gundogdu *et al.* (2009) when Pb ion was removed by pine bark from aqueous solutions.

Maximum removal was obtained at pH 5.5 and remained constant at higher pH where the removal occurs due to precipitation at pH higher than 6 (Nadeem *et al.*, 2009). Various studies showed that Pb⁺² is the predominant lead species at pH < 6 and PbCO₃ at pH between 6-11 (Smolyakov *et al.*, 2000; Herrera-Urbina and Fuerstenau, 1995).

Experimental data points obtained for the Pb (II) sorption by CNF at several lead initial concentrations are presented in Fig. 2.

The Fig. 2 shows that specific metal uptake increased with an increase in the sorbate concentration. The highest uptake of CNFs was around 89 mg g⁻¹ at the initial Pb concentration of 70 mg L⁻¹ with lowest sorption capacity of 15.77 (mg g⁻¹) at initial concentration of 5 mg L⁻¹. The enhancement in metal sorption could be due to an increase in electrostatic interactions involving the sites of progressively lower affinity for metal ions (Garni, 2005). However, the increase of Pb(II) concentration, a

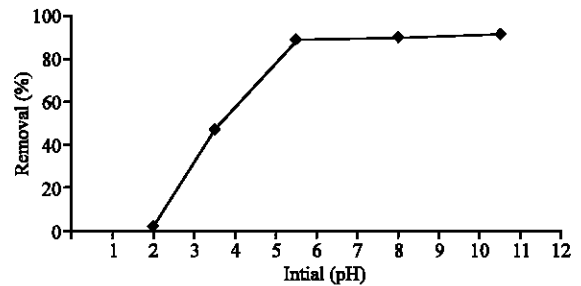


Fig. 1: Effect of pH on removal of Pb by CNFs

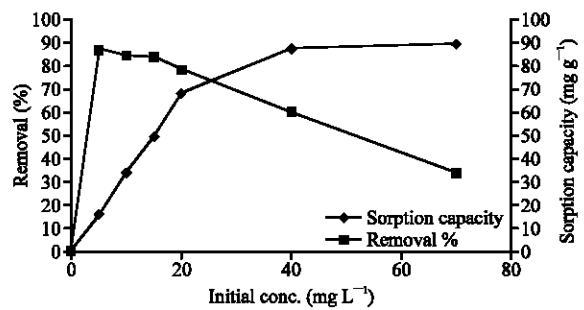


Fig. 2: Effect of initial Pb concentration on sorption capacity and removal (%)

Table 1: Comparison of the coefficients isotherm parameters for Pb(II) sorption on CNFs

Equilibrium model	Parameter	Value	q_{exp}
Langmuir isotherm	q_{max}	100 (mg g ⁻¹)	89.3
	K_L	0.4 (L mg ⁻¹)	
	R^2	0.998	
Freundlich isotherm	K_F	27.5 (mg g ⁻¹)	
	R^2	0.808	

Table 2: Comparison of various adsorbents and its percentage uptake of Pb(II)

Adsorbent	pH	Initial conc. (mg L ⁻¹)	Condition Agitation speed (rpm)	Dosage (g L ⁻¹)	Sorption % (mg g ⁻¹)*	Reference
Activated carbon (Tamarind wood)	6.5	20	120	2	97.7	Acharya <i>et al.</i> (2009)
Activated carbon (hazelnut husk)	5.7	200		12	95	Imamoglu and Tekir (2008)
Activated carbon (spartina alterniflora)	5.6	50		1	46.9*	Li and Wang (2009)
CNT	5.0	10		0.5	11.2*	Li <i>et al.</i> (2002)
CNT	5.0	40	50	0.04	96	Kabbashi <i>et al.</i> (2009)
CNFs	5.0	20	150	2.5	89.3*	This study

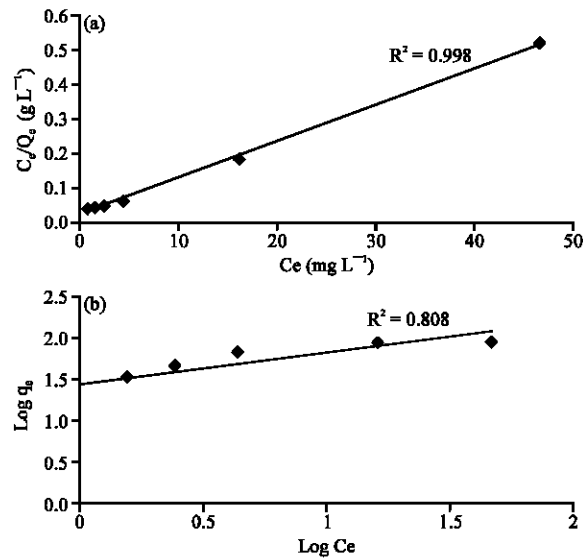


Fig. 3: Pb(II) equilibrium modeling of sorption data using (a) Langmuir and (b) Freundlich

corresponding decrease in percentage sorption was observed suggesting limiting number sorption sites available for sorption at higher Pb(II) concentration. This effect may also be interpreted that at low sorbate ion/sorbent ratios, sorbate ion sorption takes into account the higher energy sites. As the sorbate ion/sorbent ratio increases, the higher energy sites are saturated and sorption begins on lower energy sites, resulting in a decrease in removal efficiency (Sivaraj *et al.*, 2001).

Among several existing isotherms, sorption data was analyzed by two isotherms, i.e., Langmuir and Freundlich to evaluate the maximum saturation capacity of CNFs. Langmuir and Freundlich sorption isotherms of Pb(II) sorption onto CNFs are presented in Fig. 3a and b.

The Langmuir capacity, q_{max} , which is a measure of the maximum sorption capacity corresponding to complete monolayer coverage showed that CNFs had a mass

capacity of 100 mg g⁻¹ for Pb(II) cation, which is close to the experimental value 89.3 mg g⁻¹. Moreover, when R^2 values of the models are compared, indicate, Langmuir model better fits the Pb(II) sorption process onto CNF than Freundlich due to high correlation coefficient (0.998 and 0.808 for Langmuir and Freundlich, respectively). The two isotherms' constants along with correlation coefficients are enlisted in Table 1.

The results here showed the applicability of monolayer coverage of Pb(II) on the homogeneous surface of the adsorbent which is supported for a study done by Kabbashi *et al.* (2009) where CNT was used for the removal of Pb while previous study (Li *et al.*, 2002) showed that both of Langmuir and Freundlich isotherms were fitted to the sorption of Pb by CNT. Other contributions were done by using activated carbon as an adsorbent for Pb from aqueous solutions and Table 2 shows the comparison with other studies and it indicates high capacity for the adsorbent.

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