



Removal of Nickel (II) by Polyvinyl Alcohol Coated Carbon Prepared from *Datura stramonium*

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ABSTRACT

Wastewater containing heavy metal pollutants cause direct toxicity, both to human and other living organisms due to their presence beyond specified limits. Industrial effluents containing nickel play an important role in polluting water bodies. The removal of nickel ions from aqueous solution by polyvinyl alcohol coated carbon prepared from *Datura stramonium* as an adsorbent is reported in this paper. The effect of pH, initial metal ion concentration, contact time and adsorbent dose were studied. The adsorption capacity of *Datura stramonium* was dependent on the pH of the nickel solution, maximum nickel removal (78.9 %) was obtained at pH of 6. The adsorption experimental data fit well with the Langmuir and Freundlich adsorption isotherms. The kinetics of the adsorption process follows the pseudo second-order kinetics model. The results indicate that *Datura stramonium* can be employed as a low cost alternative to commercial adsorbents in the removal of nickel (II) from wastewater.

INTRODUCTION

Heavy metals in wastewater arise mainly from industrial sources (Culp & Culp 1974). Toxic metals are often discharged by a number of industrial processes and this can lead in turn to the contamination of freshwater and marine environment (Low et al. 2000). The important toxic metals, i.e., Cd, Zn, Ni, and Pb find their way to water bodies through wastewaters (Ajmal 1998). Nickel is used in many industrial and consumer products, including stainless steel, magnets, coinage, rechargeable batteries, electric guitar strings and special alloys. Nickel salts are commonly used in metal plating and their concentration in industrial wastewater varies between 6 and 12 mg/L, which is above the safe limit (Bansal & Gotal 2005). The major sources of nickel into water bodies are electroplating industries, nickel mining and processing, etc. Dermatitis (Ni-itch) is the most frequent effect of exposure to Ni, such as coins and jewellery. The higher concentration of Ni (II) in ingested water may cause severe damage to lungs, kidneys, gastrointestinal distress, e.g., nausea, vomiting, diarrhoea, pulmonary fibrosis, renal edema, and skin dermatitis (Chen & Lin 1998). Nickel is removed from electroplating wastes by treatment with hydroxide, lime, and/or sulphide to precipitate the metal. Adsorption with activated carbon, activated alumina and iron filings is also used for treating nickel containing wastewater. Ion exchange is also used for nickel removal and recovery. Of these, adsorption has been preferred over other processes because of its cheapness and the high-quality treated effluents it produces.

The present study was undertaken to evaluate the efficiency of polyvinyl alcohol coated carbon prepared from *Datura stramonium* for the removal of Nickel (II) ions in aqueous solutions. In the present work, an attempt is made to analyse the cost of polyvinyl alcohol coated *Datura stramonium*

(PVAC-DS).

MATERIALS AND METHODS

Preparation of polyvinyl alcohol coated carbon of *Datura stramonium*: The naturally dried stems of the plant *Datura stramonium* were obtained locally, and cut into small pieces. The stems were treated with 2% v/v sulphuric acid in 1:1 ratio, kept in an oven at 150°C for 24 hours, and then filtered and washed with distilled water repeatedly to remove sulphuric acid (washings tested with two drops of barium chloride solution) and finally dried and powdered (ACDS). Chemical activation of carbon using sulphuric acid produces a high surface area and high degree of microporosity.

Polyvinyl alcohol is a good hydrophilic polymer and has water-adsorbing capacity. The powdered ACDS was treated with an emulsion of readily available synthetic polymer-PVAC. Nine parts by weight of activated carbon obtained from the stems of *Datura stramonium* were mixed with one part by weight of PVAC to form a semisolid mass. The agglomerated product was dried and ground into fine powder. The adsorbent was sieved to 40-60-mesh size and dried at 110°C for 2 hours. This powder was then used as an adsorbing material.

Adsorbate: Ni (II) stock solution (1000 mg/L) was prepared in double distilled water using nickel sulphate. The working solution was obtained by diluting the stock solution in distilled water.

Batch adsorption studies were carried out to study the effect of pH (3, 4, 5, 6, 7 and 8), contact time (30-150 min), adsorbent dose (3-6 g/L) and initial metal ion concentration (100-250 mg/L) at room temperature using stopper bottles. The initial pH of the solution was adjusted by using 0.1 M sodium hydroxide and/or 0.1 M nitric acid without changing volume of the sample. After agitating the sample for the required contact time, the contents were centrifuged and filtered through Whatman No. 41 filter paper and unreacted nickel in the filtrate was analysed.

The removal efficiency (E) of the adsorbent was defined as:

$$E (\%) = [(C_o - C_e)/C_o] \times 100$$

Where C_o and C_e are the initial and equilibrium concentrations of metal ion solution (mg/L), respectively.

The equilibrium adsorption data were then fitted to Freundlich and Langmuir isotherm equations.

Freundlich adsorption isotherm:

$$\log q_e = \log K_f + 1/n \log C_e$$

Where,

q_e = the amount of the adsorbate adsorbed per unit mass of adsorbent (mg adsorbate/g adsorbent)

K_f = adsorption capacity and n = adsorption intensity are the empirical constants.

C_e = equilibrium concentration of adsorbate (mg/L)

The equilibrium concentration was calculated using following formula:

$$C_e = C_o - (\% \text{ adsorption} \times C_o/100)$$

C_o = initial concentration of adsorbate (mg/L)

The amount of the adsorbate adsorbed per unit mass of adsorbent q_e (mg adsorbate/g adsorbent)

was evaluated by the following formula:

$$q_e = (C_0 - C_e) \times V/m \times 1000$$

Where, V = volume of the aqueous phase (L)

Langmuir Isotherm:

$$C_e/q_e = 1/Q_m b + C_e/Q_m$$

Q_m and b are Langmuir constants related to adsorption capacity (maximum specific uptake corresponding to the site saturation) and energy (intensity) of adsorption (L of adsorbent/mg of adsorbate) respectively. The essential characteristics of the Langmuir isotherm can be expressed in terms of a dimensionless constant, separation factor or equilibrium parameter R_L that is defined as:

$$R_L = 1/(1 + b C_0)$$

Where, b is the Langmuir constant and C_0 is the initial concentration of the metal ions. Values of dimensionless equilibrium parameter R_L show the adsorption to be favourable if ($0 < R_L < 1$).

Kinetics

To describe the adsorption kinetics, a pseudo-second order rate model reported in the literature was applied in the following form.

$$t/q_t = 1/h_0 + 1/(q_e)t$$

Where,

h_0 = the initial adsorption rate (mg/g min)

q_e = the amount of metal ion adsorbed at equilibrium (mg/g)

q_t = the adsorbed at time t (mg/g)

The initial adsorption rate, h_0 , as $t' \rightarrow 0$ is defined as:

$$h = K_2 q_e^2$$

Where, K_2 is the pseudo second order rate constant for the adsorption process (g/mg min). The initial adsorption rate h_0 , the equilibrium adsorption capacity, and the rate constant K_2 were determined from the slope and intercept of the plot of t/q_t against t .

RESULTS AND DISCUSSION

Effect of contact time: In adsorption system, the contact time plays a vital role irrespective of the other experimental parameters, affecting the adsorption kinetics. Fig. 1 shows that increasing contact time increased Ni (II) ion uptake and it remained constant after 120 minutes, indicating that the adsorption sites are well exposed. Thus, the effective contact time (equilibrium time) is taken as 120 min.

Effect of pH: At low pH the adsorption percentages of Ni (II) ion are small (Fig. 2). Between pH values 5-6, the percentage increased sharply up to 78.9 %, and then decreased with increase in pH. The lower the pH, the more H^+ ions compete with Ni (II) ions for adsorption sites, thus, reducing their adsorption. On the other hand, the higher the pH, the less the H^+ ions competing with metal ions for adsorption sites, thus, increasing their adsorption, which explains the obtained results in Fig. 2.

Effect of initial metal ion concentration: The increase in initial metal ion concentration decreased the percent adsorption and increased the amount of metal uptake per unit weight of the PVAC-DS (adsorption capacity Fig. 3). The adsorption efficiency decreased with increasing of the initial con-

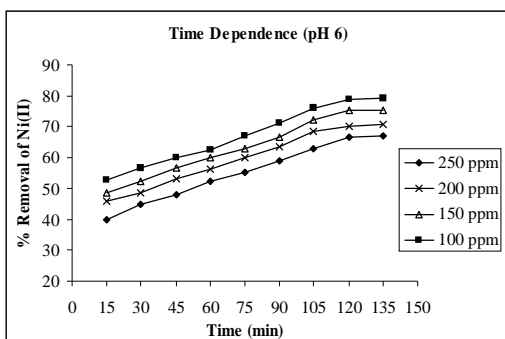


Fig. 1: Effect of contact time on removal of Ni(II) at different concentrations by PVAC-DS at pH 6.

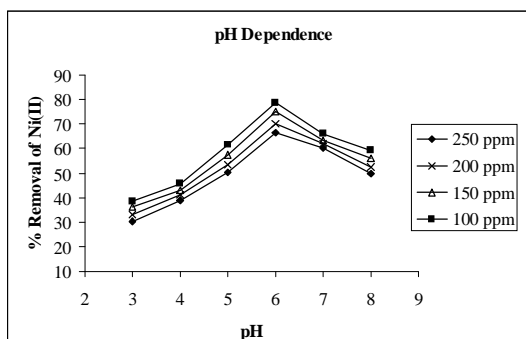


Fig. 2: Effect of pH on removal of Ni (II) at different concentrations by 5 g/L of PVAC-DS at constant contact time of 120 min.

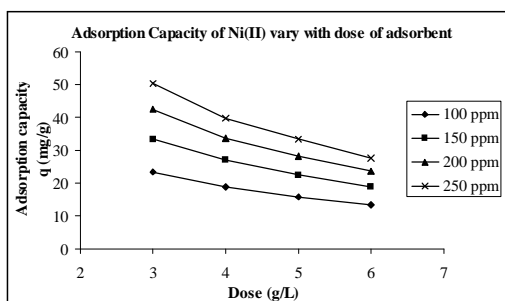


Fig. 3: Effect of dose of adsorbent on adsorption capacity at equilibrium contact time of 120min and effective pH 6.

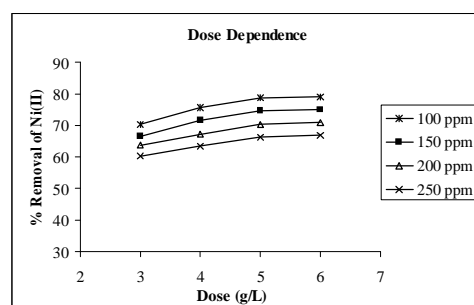


Fig. 4: Effect of PVAC-DS dose on percent removal of Ni(II) at equilibrium contact time 120 min and at effective pH 6.

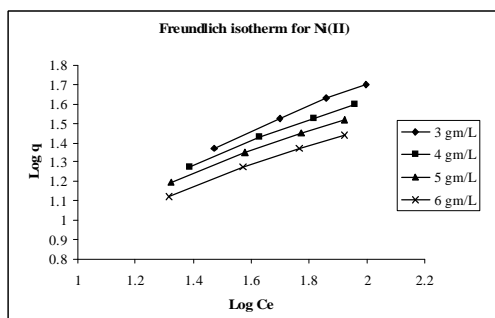


Fig. 5: Freundlich isotherm plot for Ni(II) adsorption by PVAC-DS at optimum conditions.

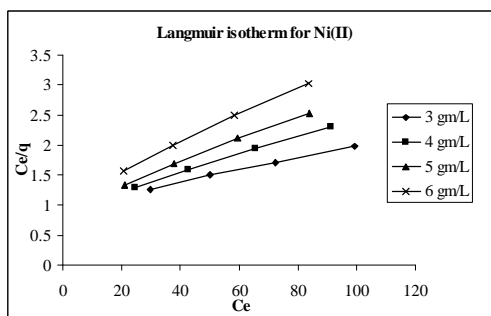


Fig. 6: Langmuir isotherm plot for Ni(II) adsorption by PVAC-DS at optimum conditions.

centration of the metal ions. These results may be explained on the basis that the increase in the number of ions competing for the available binding sites and also because of the lack of active sites on the adsorbent at higher concentrations. Therefore, more metal ions were left unadsorbed in solution at higher concentration levels.

Effect of dose: From Fig. 4, it can easily be inferred that the percent removal of metal ions increases with increasing weight of PVAC-DS. The present study indicated that the amount of Ni(II) adsorbed

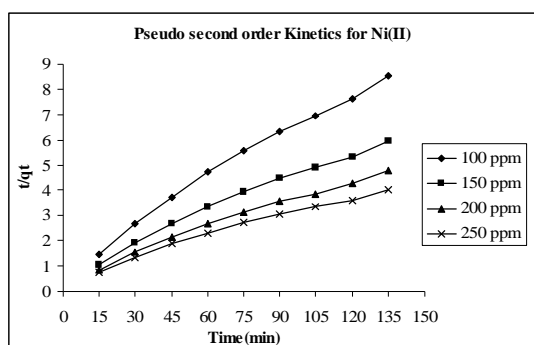


Fig. 7: Pseudo 2nd order kinetic model plot for Ni(II) by PVAC-DS.

calculated from the graph are summarized in Table 1 for Ni(II) adsorption by PVAC-DS. Value of n for Ni (II), was 1.8598 by PVAC-DS at effective doses and contact time, indicates good adsorption potential of the adsorbent. Values of R_L and R^2 show the adsorption to be more favourable. The value of R_L was 0.6273 for Ni (II) solution by PVAC-DS.

Kinetic study: Based on linear regression (R^2) values, the kinetics of Ni (II) adsorption onto PVAC-DS for 100 ppm solution can be described well by pseudo second order ($R^2 = 0.9888$) equation (Fig. 7). The values of K_2 for Ni (II) are 3.2×10^{-3} , 2.0×10^{-3} , 1.6×10^{-3} and 1.2×10^{-3} for 100, 150, 200 and 250 ppm respectively. The results clearly indicate that this model fit progressively well with increasing adsorbate concentration (Table 2).

Cost Analysis

The cost for the preparation of the adsorbent is calculated based on the breakup cost of each step (including physical and chemical activation procedure). The total cost for the preparation of 1 kg of PVAC-DS adsorbent is Rs. 31.87. The commercially available adsorbent (activated carbon) is purchased for Rs. 500.00 per kg. Based on the adsorption capacity (15.78 mg/g) obtained in batch

Table 1: Values of langmuir and freundlich constants for adsorption of nickel (II) by PVAC-DS.

Adsorbent dose (g/L)	Langmuir constants				Freundlich constants		
	Q_m (mg/g)	b (L/mg)	R_L	R^2	K_f	n	R^2
3	99.01	0.0104	0.7626	0.9981	2.7568	1.5748	0.9977
4	66.23	0.0161	0.6738	0.9985	3.1805	1.7778	0.9967
5	52.63	0.0198	0.6273	0.9972	3.1046	1.8598	0.9973
6	43.48	0.0207	0.6170	0.9985	2.5416	1.8328	0.9976

Table 2: Pseudo second order parameters for Ni (II) at pH 6.

Parameters	100 ppm	150 ppm	200 ppm	250 ppm
K_2	3.2×10^{-3}	2.0×10^{-3}	1.6×10^{-3}	1.2×10^{-3}
R^2	0.9888	0.9856	0.9875	0.9843
q_e	17.57	25.31	31.85	38.31
h	0.9836	1.3058	1.6327	1.6898

on PVAC-DS increase with increase in dose up to 5 g/L, but thereafter with further increase in dose, the increase in removal was very small. Thus, the effective dose is taken as 5 g/L at pH 6. This is due to the greater availability of the exchangeable sites or surface area at higher dose of the adsorbent. These results are in agreement with previous studies on many other adsorbents (Yu et al. 2001, Dakiky et al. 2002).

Results of adsorption isotherms: Langmuir and Freundlich isotherms for Ni(II) by PVAC-DS were found to be linear showing the applicability of the isotherms (Figs. 5 and 6). The values of Langmuir and Freundlich constants

study for the adsorbent, the cost of adsorbent is calculated for the removal of 1 g of Ni (II) from wastewaters. The cost of adsorbent for removal of 1 g of the metal ion comes to Rs. 2.02. *Datura stramonium* is the most economical adsorbent, and much cheaper than activated carbon.

CONCLUSION

From the present study, it may be concluded that the removal of Ni (II) from synthetic wastewater by adsorption on adsorbent PVAC-DS is useful. The removal of Ni (II) achieved was 78.9 % on PVAC-DS. The adsorption of metal ion on PVAC-DS reached equilibrium in 120 min, pH 6 and initial concentration 100 ppm. The Freundlich isotherm is followed better than the Langmuir isotherm with $R^2 0.9973$. *Datura stramonium*, which is available in abundance locally, is the most efficient adsorbent, and much cheaper than activated carbon.

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