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Nickel (Ni^{+2}) Adsorption on Borax Production Waste from Industrial Wastewater

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Abstract

Borax sludge was selected as the adsorbent due to its dolomite content to purify the industrial wastewater. Ni^{+2} concentration in the industrial wastewater was decreased with the adsorption on the borax production waste. To clarify the adsorption mechanism, various pH values, initial concentrations, and contact times were applied. The adsorption percentage increased with the increasing pH value because of the precipitation of metal ion. In the isothermal analyses, the best-fitted isotherms were in the order of Temkin> Freundlich> Langmuir> Harkins-Jura isotherms. The adsorption heat (B_T) was determined as 277 kJ/mole in Temkin isotherm and n value of Freundlich was estimated as 1.3. In the kinetic analyses of Ni^{+2} adsorption, the pseudo-second-order was the best fitted kinetic method. The adsorbed amount value of Ni^{+2} per unit mass (q_e) varied between 8.3682 and 40 mg/g. The estimated isothermal and kinetic results indicated that the sludge of borax production waste can be preferred as an alternative for the preparation of low-cost adsorbents.

Keywords: Adsorption kinetics, Borax waste, Nickel, Isotherms, wastewater, water treatment

1. INTRODUCTION

Waste formation is one of the world's major problems. The concentration and composition of wastes in water resources have increased over the last years with the expansion of population and industrial activities. Industrial wastewater includes heavy metals at various types and concentrations. Metal pollution in water supplies exhibits harmful effects on both the environment and human health [1 – 3]. As a heavy metal, nickel and its components are commonly seen in nickel refining, welding and electroplating. In the exposure of nickel, the target organs are the lung and the skin. The significant health effects because of occupational exposure to nickel can be listed as lung cancer, skin allergies and lung fibrosis [4].

As a separation method, adsorption is a process for the heavy metals removal from contaminated water that receives attention due to its advantages of high efficiency, being easy and economic. To obtain an effective adsorption process, the adsorption mechanism should be clarified by using the isothermal and kinetic analyses. With this purpose, different adsorbents have been studied for heavy metal removal. Farmaki et al., experimented the adsorption of Ni^{+2} and Pb^{+2} on tailings of limestone and dolomite as a mine waste [5]. Dada et al., investigated the isothermal analyses of Zn^{+2} on the modified risk husk, as an agricultural adsorbent [6]. Meng et al., synthesized the zeolite A and used as an adsorbent for the removal of heavy metals from the industrial wastewater [7].

The borax production waste is also known as borax sludge, generally contains the clay and borax and/or tincalconite. Annually, more than 120.000 tonnes of borax waste are produced in Turkey. Generally, they can be preferred as an extender in the wail tile or briquette production [8]. Due to its clay component, it can be used as an alternative to the low-cost adsorbents.

In this study, the borax production waste was selected to purified nickel content of the industrial wastewater. The Ni^{+2} adsorption on borax production waste was studied and the adsorption mechanism was clarified by using the isothermal and kinetic methods. The effects of the pH, initial concentration and contact time on to the adsorption were determined.

2. EXPERIMENTAL PROCEDURE

2.1. Materials

The borax sludge was supplied from Bandırma Boron and Acid Factory Operations of Eti Mine Works in Balikesir, Turkey. The sludge was dried at 105°C in Ecocell oven for 2 hours to decrease its moisture content. The dried waste was powdered and sieved to decrease particle size lower than 90

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µm. for sieving process, the vibrating screen of Fritsch was used. The dried adsorbent was identified as the mixture of tincalconite (pdf. no: 00-008-0049; $Na_2B_4O_7 \cdot 5H_2O$) and dolomite ($Ca\text{-}Mg(CO_3)_2$; pdf. no: 00-005-0622) phases. The BET surface area of borax sludge was 5.54 m^2/g [9].

The industrial wastewater was provided from the inlet water line of a wastewater treatment facility in the region of the Ikitelli Organized Industrial Zone where wastewater from metal coating industries' was collected. Initial pH of wastewater was 3. For the pH adjustment, NaOH was purchased from Merck Chemicals. The wastewater mainly included the heavy metals of Ni, Cu, Cr, Zn, Al and Fe. From these heavy metals, Ni was selected due to its high concentration. The initial Ni concentration of the sample was 133.67 ppm.

2.2. Adsorption Experiments

The 0.2 g of dried adsorbent was added 100 ml of wastewater and stirred at 200 rpm. For the batch adsorption study, the experimental conditions were summarized in Table 1. The adsorption experiments were applied at optimum conditions of each parameter. The solution 10 M NaOH was used to adjust the pH of the adsorption medium.

Table 1. The parameters of the batch adsorption experiments

Parameter	Values
pH	3, 5, 7, 9
Dilution ratio	0.25, 0.33, 0.5, 1
Contact time (min)	15, 30, 45, 60, 90,120

The samples were filtered after the batch adsorption and a Perkin Elmer Optima DV 2100 model of Inductively coupled plasma - optical emission spectrometer (ICP-OES) was performed to analyse the filtrates.

2.3. Isothermal and Kinetic Analyses

The obtained batch adsorption data were applied to different isotherm models: Langmuir, Freundlich, Temkin and Harkins-Jura. The Langmuir and Freundlich isotherms explained the adsorption process in the monomolecular and multimolecular layer states, respectively. The Langmuir and Freundlich isotherm models were expressed in the Equations of (1) and (2).

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

$$q_e = K_F.C_e^{1/n} \quad (2)$$

where q_e was the adsorbed Ni^{+2} amount for per unit mass in mg/g, q_{max} was the maximum adsorption capacity in mg/g, C_e was the Ni^{+2} concentration of solution at equilibrium state in mg/L, V was volume in L, K_L and K_F were the coefficients of Langmuir and Freundlich related to the affinity of the binding sites. The separation factor of R_L is preferred to indicate the required properties of the Langmuir isotherm. When R_L is between 0 and 1, it indicates that adsorption is favourable Equation (3).

$$R_L = \frac{1}{1 + K_L.C_e} \quad (3)$$

The Temkin isotherm (Equation (4) and (5)) assumed that the adsorption energy would decrease with the filling of the active pores on the adsorbent surface. B_T was the ratio of RT/b_T. A_T, was the equilibrium binding constant of Temkin isotherm, B_T was the adsorption heat and b_T was the coefficient of the adsorption heat.

$$q_e = B_T.lnA_T + B_T.lnC_e \quad (4)$$

$$B_T = R.T/b_T \quad (5)$$

The Harkins-Jura isotherm model expresses the possibility of multi-layer adsorption due to the existence of heterogeneous pore distribution. The Harkins-Jura isotherm model can be expressed by Equation (6). A_H and B_H were the Harkins-Jura isotherm model constants.

$$q_e = \sqrt{\frac{A_H}{logC_e + B_H}} \quad (6)$$

The kinetic parameters of the adsorption process were estimated by using the adsorption kinetic models of Lagergren pseudo-first-order, pseudo-second-order, and intraparticle diffusion. The related equations of Lagergren pseudo and second-order were given in (7) and (8), respectively. The terms of k_I and k_{II} were rate coefficients for pseudo-first-order and second-order, respectively.

$$\frac{dq_t}{dt} = k_I(q_e - q_t) \quad (7)$$

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

The function of the intraparticle diffusion kinetic model was seen in equation (9). The value of k_{III} was the rate coefficient of the related kinetic model.

$$dq_t = k_{III}t^{0.5} + A \quad (9)$$

3. RESULTS AND DISCUSSION

3.1. Adsorption Results

The percentage of absorbed Ni^{+2} from the wastewater was presented in Figure 1 at different pH values. The initial pH value of wastewater was 3. The alkalinity of the solution was a major factor for the Ni^{+2} adsorption from the wastewater. The higher adsorption percentages can be explained with the precipitation of metal ions in alkali mediums [10]. According to the Figure 1, the acidic conditions would decrease the precipitation rate of nickel precipitates. The removal rate was low due to the insufficient precipitation. The optimum pH value was selected as 7 in the experimental setup to obtain treated wastewater in an environmentally friendly way.

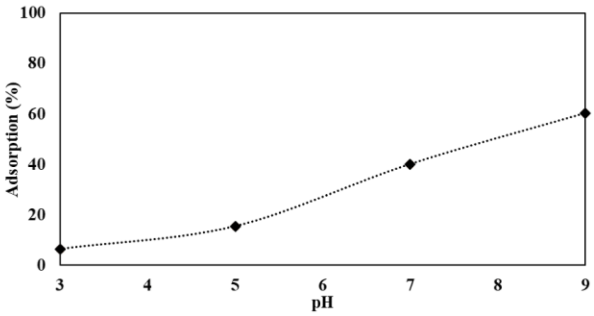

Figure 1. pH effect on Ni^{+2} adsorption

The wastewater was diluted at the ratios of 0.25, 0.33, 0.5 and 1 to obtain different initial concentrations. The Ni^{+2} adsorption at different contact times for each dilution ratio was presented in Figure 2. Ni^{+2} adsorption rapidly increased in the first 30 minutes. The minor changes were seen after the 30 minutes.

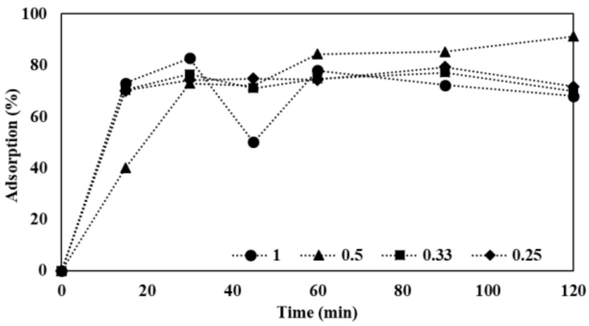

Figure 2. Ni^{+2} adsorption at different contact times

The probable adsorption mechanism for Ni^{+2} by the borax sludge may be explained in three steps:
(i) the dissolution of tincalconite and dolomite phases in borax sludge, and the formation of Na^+, Ca^{+2}, and Mg^{+2} cations;
(ii) the precipitation of $Ni(OH)_2$;
(iii) the exchange of cations on the adsorbent (Na^+, Ca^{+2}, and Mg^{+2}) with Ni^{+2}.
The adsorption results indicate the successful progress of the waste dissolution, $Ni(OH)_2$ precipitation and/or cation exchange with increasing the pH values. This proved that the adsorption mechanism proceeds well in alkaline environments.

3.2. Results of Isothermal Analyses

Isotherm parameters of the Ni^{+2} adsorption on borax waste were given in Table 2. According to the correlation constants in isothermal analysis results, the best-fitted isotherms were in the following order: Temkin> Freundlich> Langmuir> Harkins-Jura. The highest correlation constant of Temkin isotherm model signified that the adsorption heat in the layer decreases in a linear way as a result of the increase in surface coverage. The n value of Freundlich isotherm can be interpreted with the increase in chemical concentration would decrease the relative adsorption.

Table 2. Isotherm parameters of the Ni^{+2} adsorption on borax waste

Isotherms	Plot	Parameter	Result
Langmuir	$1/q_e$ vs $1/C_e$	q_{max}	49.7512
		K_L	0.0185
		R_L	0.4187
		R^2	0.9905
Freundlich	$\log(q_e)$ vs $\log(C_e)$	K_F	1.1637
		n	1.3
		R^2	0.9918
Temkin	q_e vs $\ln(C_e)$	A_T	0.2282
		B_T	8.9329
		b_T	277.3536
		R^2	0.9983
Harkins-Jura	$1/q_e^2$ vs $\log(C_e)$	A_H	-38.9105
		B_H	-1.5486
		R^2	0.9568

The adsorption capacities (q_{max}) of various adsorbents were compared in Table 3. According to the given table, adsorption capacity was affected from the coupling of adsorbent – heavy metal. The results indicated the suitability of borax production waste for the Ni adsorption than the Cr. Also, borax sludge can be an alternative adsorbent in stead of modified ores or some types of plant peels.

Table 3. Comparison of the adsorption capacity (q_{max}) of different adsorbents

Table 3(cont.). Comparison of the adsorption capacity (q_{max}) of different adsorbents

Adsorbent	Heavy metal	q_{max}	Ref.
Borax production waste	Ni	49.7512	This study
Borax production waste	Cr(III)	24.096	[9]
Wheat straw-rice husk	Ni	0.427	[10]
Chitosan-Vermiculite Composite	Pb	0.154	[11]
Modified sepiolite	As(III)	46.7	[12]
Brassica Napus straw	Cu	9.37	[13]
Modified orange peel	Ni	80.0	[14]

3.3. Results of Kinetic Analyses

Kinetic parameters of the Ni^{+2} adsorption on borax waste were given in Table 4. Due to the highest correlation constants (between 0.8436 and 0.9939), the pseudo-second-order kinetic method was determined as the best-fitted model. For this kinetic model, the q_e values were in the range of 8.3682 – 40 mg/g. the adsorption rate coefficients were varied between 3.2×10^{-5} and 0.0363.

Table 4. Kinetic parameters of the Ni^{+2} adsorption on borax waste

Kinetic Method	Concentration	%25	%33	%50	%100
Lagergren Pseudo first order	q_e	0.3950	0.2496	25.650	42.934
	K	-0.0175	-0.0269	0.0090	0.0387
	R^2	0.7601	0.4367	0.8613	0.9159
Pseudo second order	q_e	8.3682	9.3023	98.039	40.00
	K	0.0363	0.0279	3.2×10^{-5}	0.0015
	R^2	0.9939	0.9403	0.8436	0.8991
Intraparticle diffusion	K_{ID}	-0.3195	-0.2197	2.9278	1.3312
	A	9.6961	10.188	-8.3721	19.558
	R^2	0.6923	0.1398	0.8726	0.3327

4. CONCLUSION

The heavy metal adsorption from the industrial wastewater was achieved by using industrial waste. The pH of the adsorption medium was a notable factor to provide the metal ions precipitation as hydroxides. The adsorption mechanism was studied by various isothermal and kinetic models at different initial concentrations and contact times. The best-fitted model function was determined as Temkin isotherm and Pseudo-second-order kinetic method. The adsorption results promoted the possible use of borax sludge, which is readily available in Turkey, as a low-cost adsorbent for the Ni^{+2} removal from industrial wastewater.

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