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OPTIMIZATION STUDIES FOR THE REMOVAL OF NICKEL FROM AN AQUEOUS SOLUTION ON TO *MADHUCA INDICA* LEAVES POWDER

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ABSTRACT

Industrial evolution, urbanization and agricultural development are highly required for the development of socity and nation. However, untreated or partially treated industrial effluents, sewage waste, domestic waste and agricultural runoffs generate large amounts of pollutants and became major source of water pollution. Wide varieties of organic and inorganic pollutants released into water streams from these sources are highly toxic and pose serious threat to all life forms. Therefore the present work was aimed to evaluate the biosorption potential of *madhuca indica* leaves for the removal of nickel from aqueous solution. Batch experiments such as contact time, pH of the solution, biosorbent dosage, average partical size of the biosorbent, initial metal concentration and temperature of the aqueous solution were conducted for the removal of nickel from aqueous solution using *madhuca indica* leaves as biosorbent. The Central Composite Design (CCD) programming was utilized to outline the analyses and to decide the optimum (68% for 25 min contact time) conditions for the present study.

KEYWORDS: ***Biosorption;Madhuca indica; Central composite design model and Ni.***

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INTRODUCTION

Metal pollution has been a great stress as far back as couple of decades. It is assumed that the wide usage of man made chemicals, anthropogenic lifestyle, and fast industrialization is the huge wellspring of metal harmful quality.[1] Nickel is outstanding as a considerable metal poison, show in effluents of electroplating endeavors, cleaning, and composite gathering, mining, and refining industries.[2] Nickel has been caught as an embryotoxin and teratogen.[3] The higher gathering of nickel causes dermatitis, squeamishness, hurling, behavioral, and respiratory issues despite cyanosis, gastrointestinal inconvenience, and shortcoming.[4] All these natural issue results alert the need of nickel expulsion from the earth and to raise its levels beneath as far as possible from its sources. The established physicochemical strategies are generally utilized for the expulsion of nickel from the mechanical effluents, to be specific, evaporative recuperation, filtration, particle trade, and layer advances. in spite of the fact that they are promising to some degree, however these procedures have high reagent or vitality prerequisites and create lethal slime that requires watchful disposal.[5] Biosorption is a procedure that utilizations economical biomaterials to sequester metals from fluid arrangements and the biomaterials utilized as a part of this procedure are named as biosorbents. The side-effects from agribusiness, nourishment and pharmaceutical businesses give monetarily suitable wellsprings of biosorbent; this makes biosorption a cheap option treatment strategy. Late research on biosorption has demonstrated that biomaterials containing acidic gatherings, for example, hydroxyls and carboxyls were compelling in restricting metal cations.[6] The objective of this study is to determine the optimum conditions for the removal nickel from aqueous solution using *Madhuca indica* leaves powder.

MATERIALS & METHODS

Preparation of the biosorbent

Madhuca indica leaves were collected from a nearby Tenali canal place in Guntur District, Andhra Pradesh, India. The collected leaves were washed with distilled water several times until the dirt particles removed. After through washing with distilled water, biosorbent was sun dried for ten days until they became crispy, cut into small pieces, powdered and sieved.

Preparation of nickel stock solution

Hydrated nickel chloride ($NiCl_2.6H_2O$, Nickel(II) Chloride Hexahydrate) was used as the source of nickel stock solution. All the required solutions were prepared with double distilled water. 1000 ppm of nickel stock solution was prepared by dissolving 8.48 grams of $NiCl_2.6H_2O$ in 1000 mL of double distilled water. From the standard stock solution, working solutions of lower concentrations were prepared to nickel (20ppm,40ppm,80ppm,120ppm and 160ppm), used for batch experimentation.

Statistica

Statistica provides its uses with a vast array of capabilities, some of which cannot easily found elsewhere. Statistica is particularly strong in certain areas. Its primary strengths are in the areas of graphical analysis, data mining, optimization of process variables and process control statistics. In the present work STATISTICA.7 was used for the optimization of nickel from aqueous solution.

Effect of contact time

To determine the effect of contact time, 0.5 g of 53 μm biosorbent was taken in 50 ml of aqueous solution of initial metal concentration of 20 mg/L, at pH=7 and the contents were shaked well for 1 min on an orbital shaker. The experiment was repeated for different time intervals like 2, 3, 4 etc. up to 195 min at constant agitation speed. After each interval of time the sample was filtered and was analyzed for determination of residual metal ion concentration. This gives an opportunity for determination of optimum contact time.

Effect of initial metal ion concentration

To know the effect of initial metal concentration, 0.5 g of biosorbent was added to 50 mL stock solution of initial metal concentration of 20 mg/l by maintaining at optimum contact time. This procedure was repeated with 50 ml of stock solution, for different initial metal concentrations 40, 80, 120 and 160 mg/L keeping the pH value, agitation speed and room temperature constant. Then the samples were filtered from biosorbent and they were analyzed for retained metal ion.

Effect of biosorbent dosage

The effect of biosorbent dosage on the amount of metal adsorbed was obtained by agitating 50 mL of nickel solution of concentration 20 mg/L separately with 0.5, 1, 1.5, 2 and 2.5 g of biosorbent at room temperature for optimum shaking time at a constant agitation speed.

Effect of pH

The pH of the solution was adjusted using 0.1 N NaOH and 0.1 N HCl solutions. In this study, 50 ml of nickel metal solution of concentration 20 mg/Lwas agitated with 0.5g of biosorbent at room temperature with constant agitation speed of 180 rpm for complete one day. Samples were filtered from biosorbent and they were analyzed for concentration of metal ion.

Effect of average particle size of the adsorbent

50 ml of nickel solution of initial concentration varying from 20 to 160 mg/L was added to 0.5 g of biosorbent of average particle size 53 μm, and it was kept for shaking at the constant equilibrium contact time and pH. The sample was filtered from biosorbent and analyzed for concentration of metal ion. This experiment was repeated at constant agitation speed and at room temperature with different average particle sizes of biosorbent from 53 – 150μm.

Effect of solution temperature

To study the effect of temperature,50 ml of nickel solution of initial concentration varying from 20 to 160 mg/L was added to 0.5 g of biosorbent (*madhuca indica*). Then the solutions were agitated with constant speed and pH for the equilibrium contact time at different temperatures from 283K to 323K. Then the solids were separated by filtration and analyzed in the

Atomic Absorption Spectrophotometer for the metal ion content in the test solution after adsorption.

Optimization studies

Responsse surface methodology(RSM) is a systematic methodology designed using mathematical and statistical techniques to develop a mathematical model to relate the process parameters and responses to understand the process with minimal number of experiment runs. The primary objective of RSM is to find out optimum values of operational conditions to maximize the process. A standard experimental design of RSM called Central Composite Design (CCD) for three variables and each with three levels (-1, 0, +1) was used to find out the optimum pH, metal ion concentration and biomass dosage. The design was taken as it fulfills most of the requirement for optimization of the biosorption study. For better accuracy the second – order polynomial was used. The mathematical equation relating three independent process variables and the response function i.e., percentage removal of metal ions has been expressed by the following quadratic model

$$Y = \beta_0 + \beta_1 a + \beta_2 b + \beta_3 c + \beta_{11} a^2 + \beta_{22} b^2 + \beta_{33} c^2 + \beta_{12} ab + \beta_{13} ac + \beta_{23} bc \qquad (i)$$

Where Y is the output of the process, βn is the coefficient associated with factor n, and the letters *a, b, c* are the process variables in the model.

RESULTS AND DISCUSSION

Effect of agitation time

The maximum percentage of biosorption is attained at 25 min of agitation (Fig.1). The percentage removal of nickel becomes constant after 25 min indicating the attainment of the equilibrium. Therefore, all other experiments are conducted at this contact time.[7.]

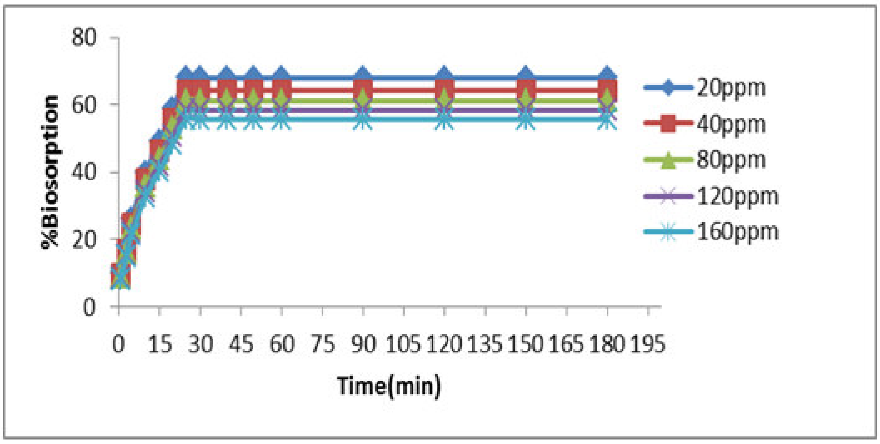

Figure 1
Effect of agitation time on % biosorption of nickel using Madhuca indica at various concentrations (20,40,80,120 &160ppm)

Effect of biosorbent size

It was observed from fig. 2 that the percentage biosorption of nickel as a function of biosorbent size is decreases with increasing particle size due to lack of specific surface area of biosorbent that cannot accommadte much more biosorbate available in the solution[8]

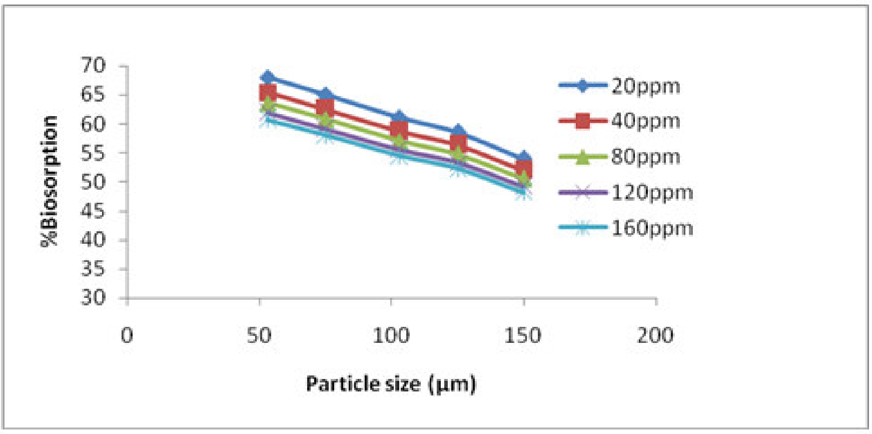

Figure 2
Measurement of % removal of nickel for biosorbent size

Effect of pH of the aqueous solution

The effect of pH on the percentage removal of nickel was shown in figure 5 under various other fixed operating conditions. The initial pH of adsorption medium is one of the most important parameters affecting the adsorption process. It can be seen (fig.3) here that the percentage biosorption was increased from pH from 2 to 5.[9]

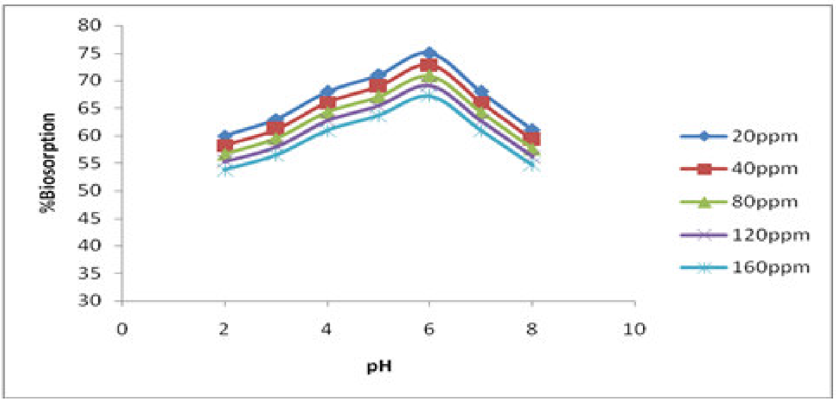

Figure 3
Dependence of % biosorption on pH of aqueous solution using Madhuca indica

Effect of initial concentration of nickel in the aqueous solution

It was noted from the fig. 4 that the % removal decreases with increase in metal concentration due to the adsorbent cannot accommodate much more adsorbate available in the solution.[10]

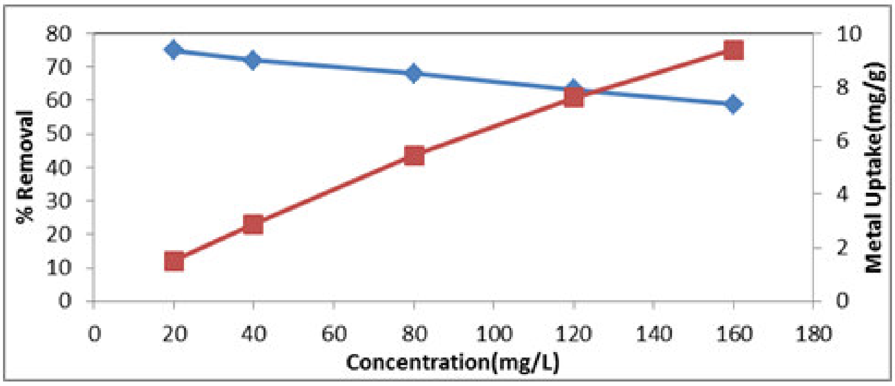

Figure 4
% biosorption as a function of initial concentration of nickel

Effect of biosorbent dosage

The percentage removal of nickel is drawn against biosorbent dosage for biosorbent size 53μ and shown in figure.5. It is evident from the plots that the percentage removal of nickel metal from the aqueous phase increases with increase in the biosorbent dosage due to availability of surface area.[11]

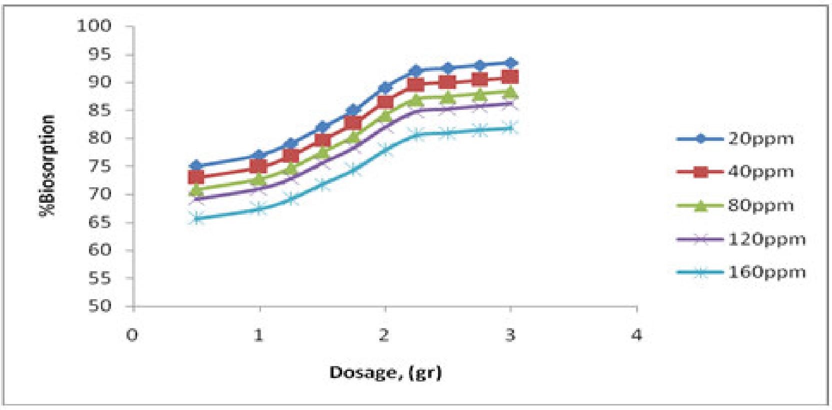

Figure 5
Dependence of % biosorption of nickel on biosorbent dosage

Effect of Temperature
The effect of changes in the temperature on the nickel removal is shown in fig. 6. It was noted that the % biosorption increases with increase in temperature up to 310K thereafter there is mild increase in removal of nickel.[12]

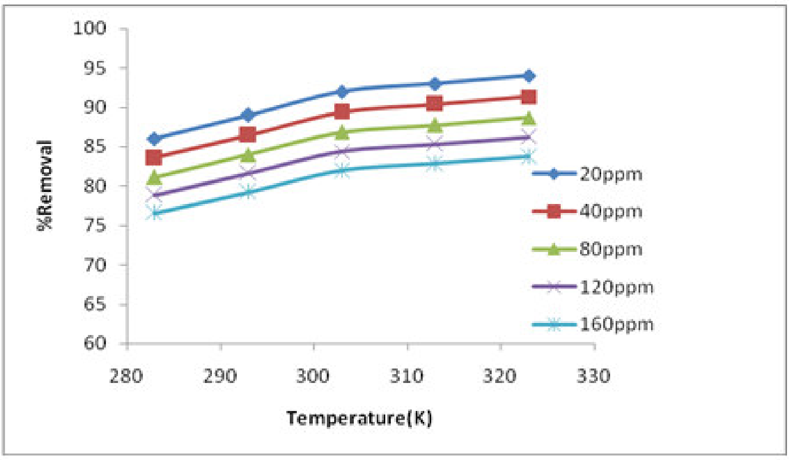

Figure 6
Variations of % biosorption of nickel on temperature

Optimization using Response Surface Methodology (RSM)
Optimization using CCD
The number of tests required for the CCD includes the standard 2n factorial with its origin at the center,2n points fixed axially. For four variables, the recommended number of tests at the center is six. Hence the total number (N) of tests required for the four independent variable is

$$N=2^n+2n+6=2^4+2\times4+6=30$$

The regression equation for % biosorption of Nickel (Y) is function of pH (X_1), C_o (X_2), w (X_3) and T (X_4). The variations in the corresponding coded values of four parameters and response are presented in table 1.

Table 1
Levels of different process variables in coded and un-coded form for % biosorption of Nickel using madhuca indica leaves powder

Variable	Name	Range and levels				
		-2	-1	0	1	2
X_1	pH of aqueous solution	4	5	6	7	8
X_2	Initial Ni concentration, C_o, mg/L	10	15	20	25	30
X_3	Biosorbent dosage, w, g/L	35	40	45	50	55
X_4	Temperature, T, K	283	293	303	313	323

The following equation represents multiple regression analysis of the experimental data for the biosorption of Nickel:

$$Y = -949.624 + 22.269\,X_1 + 2.386\,X_2 + 6.203\,X_3 + 5.278\,X_4 - 1.822\,X_1^2 - 0.073\,X_2^2 - 0.072\,X_3^2 - 0.009\,X_4^2 - 0.082\,X_1X_2 + 0.036\,X_1X_3 - 0.001\,X_1X_4 + 0.013\,X_2X_3 + 0.001\,X_2X_4 + 0.000\,X_3X_4 \quad \text{(ii)}$$

Table 2,3&4 represents the results obtained in CCD.

Table 2
Results from CCD for Nickel biosorption by madhuca indica leaves powder

Run no.	X_1	X_2	X_3	X_4	% biosorption of Nickel	
					Experimental	Predicted
1	-1	-1	-1	-1	89.28	89.31
2	-1	-1	-1	1	90.92	90.96
3	-1	-1	1	-1	90.42	90.43
4	-1	-1	1	1	92.12	92.10
5	-1	1	-1	-1	87.54	87.53
6	-1	1	-1	1	89.36	89.38
7	-1	1	1	-1	89.92	89.91
8	-1	1	1	1	91.72	91.78
9	1	-1	-1	-1	90.06	90.03

10	1	-1	-1	1	91.62	91.64
11	1	-1	1	-1	91.88	91.87
12	1	-1	1	1	93.48	93.51
13	1	1	-1	-1	86.58	86.61
14	1	1	-1	1	88.40	88.42
15	1	1	1	-1	89.72	89.71
16	1	1	1	1	91.56	91.54
17	-2	0	0	0	89.12	89.09
18	2	0	0	0	89.58	89.58
19	0	-2	0	0	91.16	91.14
20	0	2	0	0	87.42	87.40
21	0	0	-2	0	87.32	87.28
22	0	0	2	0	91.52	91.52
23	0	0	0	-2	91.42	91.44
24	0	0	0	2	94.98	94.92
25	0	0	0	0	96.62	96.62
26	0	0	0	0	96.62	96.62
27	0	0	0	0	96.62	96.62
28	0	0	0	0	96.62	96.62
29	0	0	0	0	96.62	96.62
30	0	0	0	0	96.62	96.62

Experimental conditions [Coded Values] and observed response values of central composite design with 2^4 factorial runs, 6- central points and 8- axial points. Agitation time fixed at 25 min and biosorbent size at 53 µm Response obtained from regression in eq.(ii) in the form of ANOVA is presented.[13]

$$Y = -949.624 + 22.269\, X_1 + 2.386\, X_2 + 6.203\, X_3 + 5.278\, X_4 - 1.822\, X_1^2 - 0.073\, X_2^2 - 0.072\, X_3^2 - 0.009\, X_4^2 - 0.082\, X_1X_2 + 0.036\, X_1X_3 - 0.001\, X_1X_4 + 0.013\, X_2X_3 + 0.001\, X_2X_4 \quad \text{(iii)}$$

Table 3
ANOVA of nickel biosorption for entire quadratic model

Source of variation	SS	Df	Mean square(MS)	F-value
Model	285.6234	14	20.40167	17192
Error	0.0178	15	0.001186	
Total	285.6412			

df- degree of freedom; SS- sum of squares; F- factor F; P- probability R^2*=0.99996;* R^2 *(adj):0.99988:*

Table 4
Regression coefficients for the Nickel biosorption onto madhuca indica leaves powder

Terms	Regression coefficient	Standard error of the coefficient	t-value	*P*-value
Mean/Intercept	-707.576	31.82765	-22.2315	0.000000
Dosage, w, g/L (L)	0.255	0.13553	1.8785	0.079888[a]
Dosage, w, g/L (Q)	-0.010	0.00033	-30.7070	0.000000
Conc, Co, mg/L (L)	2.062	0.27235	7.5715	0.000002
Conc, Co, mg/L (Q)	-0.031	0.00133	-22.9898	0.000000
pH (L)	10.560	1.36176	7.7549	0.000001
pH (Q)	-1.752	0.03320	-52.7670	0.000000
Temperature, T, K (L)	4.996	0.20317	24.5892	0.000000
Temperature, T, K (Q)	-0.008	0.00033	-24.7967	0.000000
1L by 2L	0.002	0.00087	2.3001	0.036214
1L by 3L	0.011	0.00435	2.4727	0.025863
1L by 4L	0.001	0.00043	2.4439	0.027367
2L by 3L	0.019	0.00870	2.1564	0.047694
2L by 4L	-0.003	0.00087	-3.9102	0.001392
3L by 4L	0.010	0.00435	2.2426	0.040456

[a]*insignificant (P ≥ 0.05)*

From the fig 8 and fig 9 it was observed that the biosorption is significant for 4 factors.

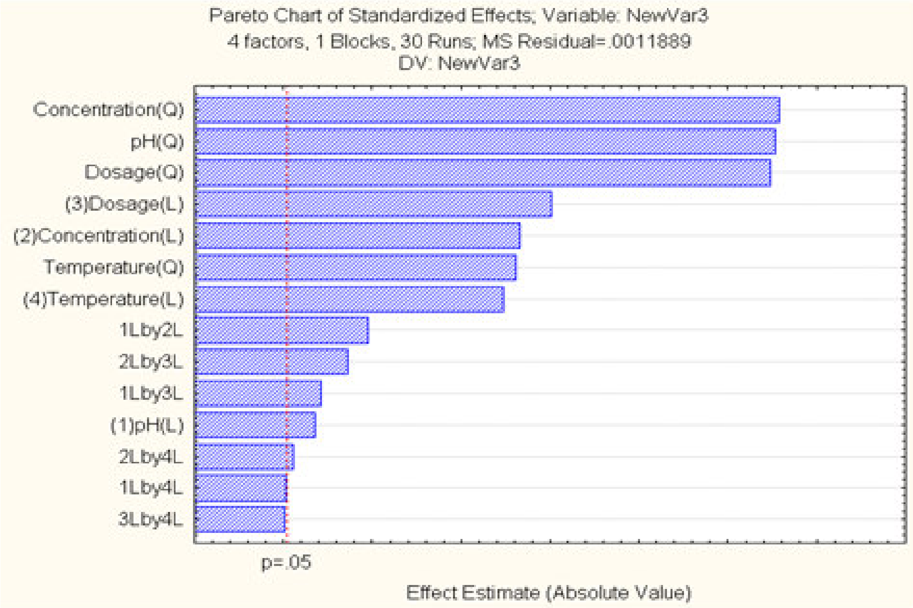

Figure 8
Pareto Chart

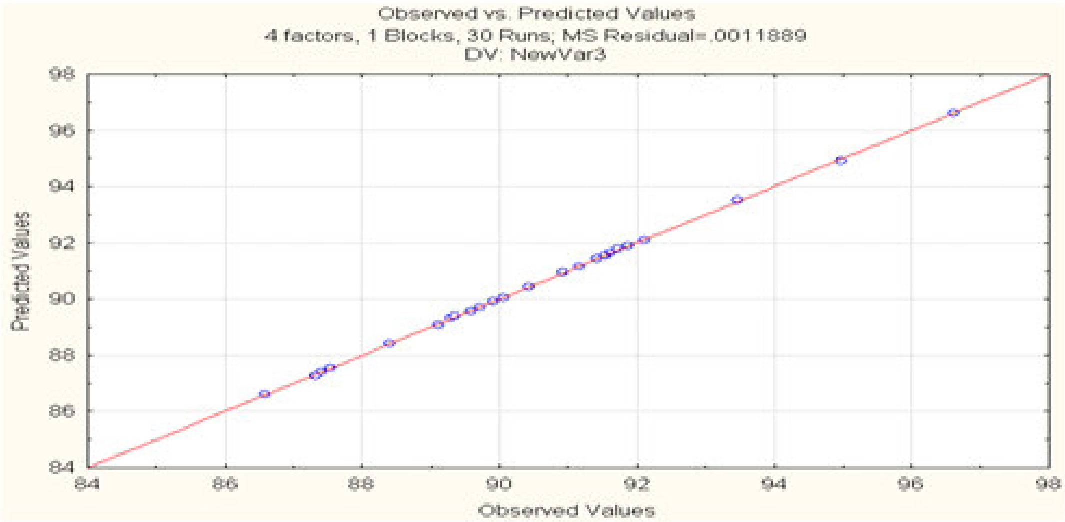

Figure 9
Normal probability plot for % biosorption of Nickel

Interaction effects of biosorption variables
The three-dimensional view of response surface contour plots [Fig.10 (a) to (f)] show % biosorption as a function of four various combinations of independent variables. The % biosorption of Nickel is strongly influenced as evident from figs. 10 (a) to (f). The optimal sets of conditions obtained with CCD and experimental are shown in table-5 along with experimental values.

Table 5
Comparison between optimum values from CCD and experimentation

Variable	CCD	Experimental value
pH of aqueous solution	6.075	6
Initial nickel concentration, mg/L	18.8380	20
Biosorption dosage, w, g/L	46.3910	45
Temperature, K	308.0048	303
% biosorption	97.09866	94

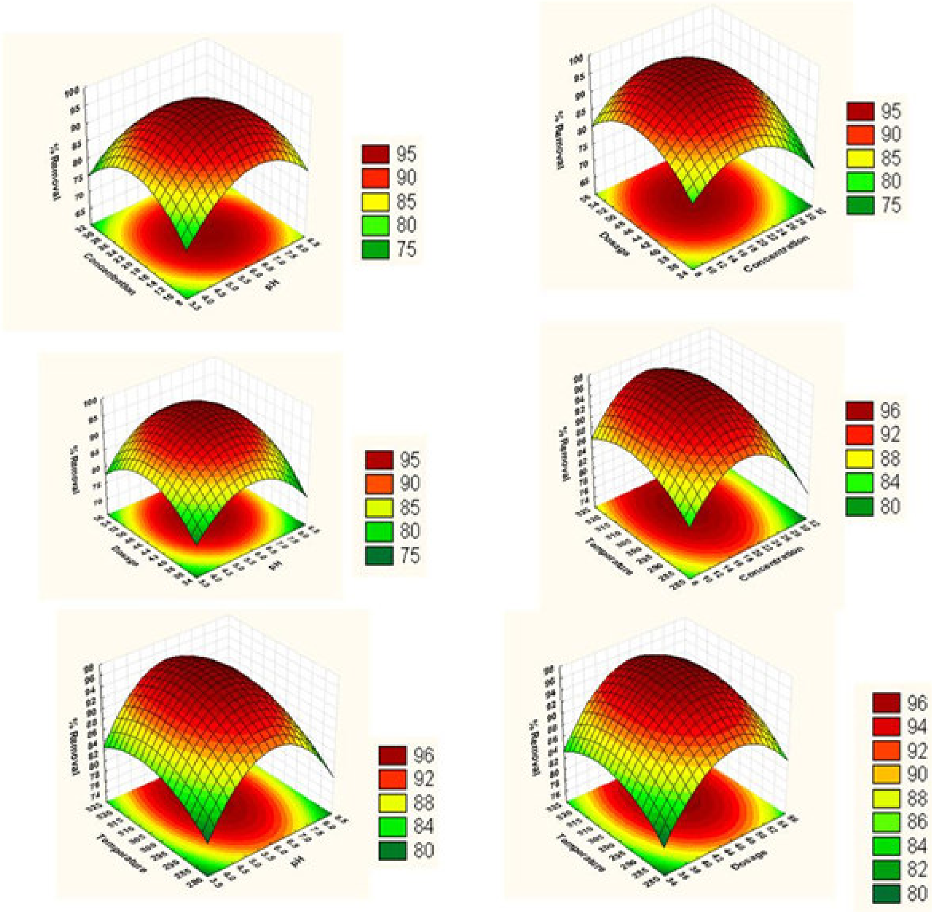

Figure 10(a-f)
Effect of biosorption on contour plots

CONCLUSIONS

The experimental results were analytically discussed and the following conclusions could be drawn from the study of biosorption of nickel from aqueous solution using biosorption technique.

1. The equilibrium agitation time for biosorption of nickel is 25 min.
2. The optimum dosage is 45 g/L .
3. % biosorption is increased upto pH = 6.
4. The CCD (Statistica.7) optimized values are:

Biosorbent dosage,w = 46.39 g/L, pH of the solution = 6.07,
Initial metal concentration,C_o = 18.83 mg/L,
Temperature,T = 308 K and extent of biosorption = 97.09 %.

CONFLICT OF INTEREST

Conflict of interest declared none.

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