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Removal of Nickel from Wastewater using Natural Adsorbents through Regression Model

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Abstract

Nickel is one of the heavy metals present in industrial effluents from metal plating, electroplating, fertilizers, pesticide and battery industries which are contaminating land and water due to its disposal. The methods which are used to remove nickel from waste water are adsorption, ion exchange, coagulation / flocculation, membrane separation, electro coagulation, floatation and biological methods. In this study, adsorption method has been used for treating the industrial waste water. The naturally and locally available materials are used as adsorbents to remove nickel. The objective of the paper is to study the removal efficiency of natural materials such as orange peel powder, neem leaf ash, saw dust and coffee husk on synthetic nickel solution. The performance of these adsorbents have been analyzed through batch test and column study. The results of important parameters such as adsorption capacity, concentration of nickel, adsorbent dosage and contact time were discussed using regression analysis.

Keywords: Nickel, orange peel powder, neem leaf ash, saw dust, coffee husk, batch test, column study, regression analysis.

1. INTRODUCTION

Excessive release of heavy metals from the industrial effluents is a major threat to both environment and the health of people [1]. The wastewater coming from industries like metal plating, tanneries, mining operations, electroplating, fertilizers, pesticide and battery industries are contaminated with heavy metals [2]. Nickel is one of the major heavy metal whose increased concentration in water causes various diseases like gastrointestinal problems, blood disorders, giddiness, nausea, vomiting, diarrhea, shortage of breathe, headache, dermatitis, skin irritation, damages to kidneys, liver problems and temporary problems in the eye [3]. Electroplating industry is a major source of nickel. In these industries, concentration of nickel in effluent ranges from 3.4 to 900 mg/l [4]. But, the permissible discharge level of it is only 0.1 mg/l [5]. Therefore, it is necessary to reduce the concentration of nickel from effluent before releasing it to the environment. There are various conventional methods available for reducing it. They are adsorption, ion exchange, coagulation / flocculation, membrane separation, electrocoagulation, floatation and biological methods. Many researchers have studied these methods and have found that the adsorption method is more effective as compared to the other methods. In order to make this method more effective and economical many attempts were made to prepare adsorbents from natural materials. Such materials are orange peel [4,6], durian rind [7], banana peel [6,8], mosambi peel [6], coconut coir [9], coconut shell [10], carrot peel [11], tamarind shell [11], palm shell [12], corn cob [13], maize cob [14], rice husk [15], saw dust [16,17], neem bark charcoal [18], coffee, tamarind and tur dal husk [19], peat [20], moringa oleifera seeds [21] and coconut leaves [22]. The objective of the paper is to perform batch test for orange peel powder, neem leaf ash, saw dust and coffee husk.

2. METHODS OF NICKEL REMOVAL

The various methods for removal of nickel like adsorption, chemical modification, ion exchange and reverse osmosis were reviewed. An extensive list of materials like mosambi fruit peelings, maize cob, rice husk, saw dust, activated clay, tea waste, ceralite IR 120, biomass, polymer sulphonate materials, etc. were used as adsorbents for removal of nickel. It was concluded that the adsorption method was more effective for removal of nickel. The choice of the method to be used is reported which depends on the concentration of the effluent, nature of the effluent, percentage removal and material required. (Sonali R. Dhokpande, et al., 2013).

3. ADSORPTION

Adsorption is a mass transfer operation in which the substances present in the solution gets accumulated on a suitable interface. The molecular species that gets adsorbed on the surface is known as the adsorbate and the surface on which the adsorption occurs is known as adsorbent.

Batch test is a testing procedure in which multiple specimens are prepared and are tested simultaneously. Each batch has number of solutions which are prepared by varying the parameters like adsorption capacity, concentration, dosage and contact time. Then, they are tested for maximum removal efficiency of each of the adsorbents.

4. MATERIALS AND METHODS

4.1 PREPARATION OF BIOSORBENTS

In order to find the adsorbing capacity of the natural adsorbents, they are prepared separately and then mixed with the solutions of known concentration. They are then tested in Atomic Adsorption Spectroscopy (AAS) which gives the concentrations of the unknowns.

Preparation of Orange Peel Powder

Orange peel contains pectin which has the ability to accumulate nickel and hence it helps in reducing the concentration of it in wastewater. Initially, orange peel was collected from the nearby fruit shop. Then, it was dried, powdered and sieved for a size of 1.8mm.

Preparation of Neem Leaf Ash

Neem leaves were procured from the neighbourhood. Then, they were sun dried for three days and oven dried for 8 hours. They were kept in muffle furnace for 2 hrs and made into ash.

Preparation of Saw Dust

Saw dust was collected from the nearby carpenter shop. Then, it was washed with distilled water and sun dried for 2 days. Finally, it was sieved to a size 0.6mm.

Preparation of Coffee Husk

Coffee husk was obtained from a nearby shop. It was obtained as a dried husk which was used as such for performing the test.

4.2 PREPARATION OF ADSORBATE

Preparation of Stock Solution

Synthetic Nickel (II) sample was prepared by dissolving Nickel Nitrate of 4.95g in one litre of double distilled water to make 1000 ppm of stock solution.

Preparation of Synthetic Solution
From 1000ppm solution, 10ml of solution was taken and mixed with 90ml of distilled water in order to obtain 100ppm. The synthetic nickel solution with known concentrations of 2, 4, 6 and 8 ppm were prepared.

4.3 BATCH PREPARATION

In batch test the known dosage of the adsorbent is taken. Then, they are added to the prepared synthetic solutions of nickel nitrate. The solution is then placed in jar test apparatus. In jar test, the rotating rim mixes the solution uniformly for a particular interval of time. The adsorbents present in the solution acts as a coagulant. On uniform mixing, the nickel nitrate present in the solution gets adsorbed by the adsorbent. After this, they are allowed to rest for some time in order to attain uniform settling. At this stage, the coagulants undergo settling process. Finally, the solution is filtered with whatman filter paper. Thus, the nickel nitrate is adsorbed and clean water is obtained. The unknown concentrations of the adsorbents are found using Atomic Adsorption Spectroscopy (AAS).

Batch Preparation for Orange Peel Powder
Orange peel powder of 1.8mm sieve size was used. Dosages of 2, 4, 6, 8 and 10g were added to the synthetic nickel solutions. Then, they were kept in the jar test apparatus and then left undisturbed for settling process. Test was performed for five different contact times such as 30, 60, 90, 120 and 150 minutes.

Batch Preparation for Neem Leaf Ash
Neem leaf ash with dosages of 0.5, 1, 1.5, 2 and 2.5 g were added to the synthetic nickel solutions. Then, they were kept in the jar test apparatus and then left undisturbed for settling process. Test was performed for two different contact times such as 15, 20, 25, 30 and 35 minutes.

Batch Preparation for Saw Dust
Saw dust with dosages of 0.25, 0.5, 0.75, 1 and 1.25g were added to the synthetic nickel solutions. Then, they were kept in the jar test apparatus then left for settling process. Test was performed for five different contact times such as 30, 60, 90, 120 and 150 minutes.

Batch Preparation for Coffee Husk
Coffee husk with dosages of 2, 2.5, 3, 3.5 and 4g were added to the standard solutions. Then, they were kept in the jar test apparatus and then left undisturbed for settling process. Test was performed for five different contact times such as 30, 60, 90, 120 and 150 minutes.

4.4 COLUMN STUDY

Column study was carried out in a glass column of 6 cm internal diameter and 45 cm height. A stopper provided at the bottom end of the column was used to maintain the desired flow rate. The experiment was carried out at room temperature 30±2^{0}C. Effect of the process parameters like concentration and contact time for a constant flow rate and fixed bed depth were investigated. Synthetic solutions were allowed to pass through the column and collected for varying contact time. The supernatant thus collected are tested for finding the adsorption efficiency.

Langmuir Isotherm
The Langmuir isotherm assumes monolayer adsorption onto a surface containing a finite number of adsorption sites. Once a site is filled, no further adsorption can take place at that site. This indicates that the surface reaches a saturation point where the maximum adsorption of the surface is achieved. The isotherm is represented by

$$\frac{C_e}{q_e} = \frac{1}{K_L q_{max}} + \frac{C_e}{q_{max}}$$

Where, the constants K_L and q_{max} are the values obtained from the slope and interception of the plot of C_e/q_e and C_e (Sheel Ratan, et al., 2016)

Freundlich Isotherm
The Freundlich isotherm is an empirical equation describes the removal of metal ions occurs on a heterogenous adsorbent surface and undergoes multilayer adsorption. The linearized equation is given by,

$$\log q_e = \log K_f + \frac{1}{n}\log C_e$$

Where, K_f is Freundlich constant which correspond to adsorption capacity. 1/n is a dimensionless constant which explains the adsorption intensity between solute concentration and the adsorbent respectively. The values of K_f and 1/n are directly obtained from the slope and the intercept of the linear plot of log q_e Vs log C_e (Sheel Ratan, et al., 2016).

5. Batch analysis of nickel removal

Supernatant solutions collected from batch adsorption test were then tested using Atomic Adsorption Spectroscopy (AAS). A reduction in concentration was found due to different adsorbents and the efficiency of each of the biosorbents was also obtained.

The instrument is turned ON. The nickel lamp is chosen followed by switching ON the compressor, gas and acetylene nob. Then, the instrument is calibrated or zero calibration. The standards are then kept and there adsorbance are read on the computer screen. The unknown samples thus obtained from batch test are studied and the reduced concentrations of synthetic nickel solutions were noted. Removal efficiencies of each of the adsorbents are found.

Table 1: Optimized Nickel Removal Efficiency of Different Adsorbents

ADSORBENTS	INITIAL CONC.	FINAL CONC.	DOSAGE	CONTACT TIME	REMOVAL EFFICIENCY
ORANGE PEEL POWDER	8ppm	6.84ppm	10g	60 mins	85.51%
NEEM LEAF ASH	8ppm	7.8ppm	2.5g	35 mins	97.43%
SAW DUST	8ppm	7.846ppm	1g	150 mins	98.08%
COFFEE HUSK	8ppm	7.82ppm	4g	150 mins	97.75%

5.1 Multiple Linear Regression Model Study

Multiple linear regression analysis was carried out with removal efficiency of adsorbent as dependent variable and contact time, concentration and dosage as independent variables. It analyzes the correlation of contact time, concentration and dosage with removal efficiency of adsorbent. The equations were arrived for different adsorbents such as orange peel powder, neem leaf ash, saw dust and coffee husk as follows with correction coefficient

Regression Equation for orange peel powder

$Y = 0.812 - 0.47X_1 + 9.13X_2 + 0.71X_3$

Regression Equation for neem leaf ash

$Y = 0.892 - 0.27X_1 + 11.83X_2 + 0.94X_3$

Regression Equation for saw dust

$Y = 0.904 + 0.86X_1 + 12.73X_2 + 1.14X_3$

Regression Equation for coffee husk

$Y = 0.922 + 0.14X_1 + 15.8X_2 + 1.32X_3$

These equations are used to find the trend and the effects of adsorbents on nickel removal. This multiple linear regression focuses on the strength of the relationship between contact time, and dosage on the percentage of nickel removal of adsorbents.

7. Column analysis of nickel removal

Correlation between concentration and adsorption were obtained using both Langmuir and Freundlich isotherm models. The regression equation is derived for both isotherms and from which the values of slope and intercept were obtained. Freundlich isotherm shows the best fit for all four adsorbents.

Orange peel powder

The values of q_{max}, n and R^2 from the Langmuir and Freundlich isotherm data analysis are as shown in table 2. It shows that Freundlich isotherm is better fit than Langmuir isotherm. The figure 1 shows that the line fit between amounts of metal adsorbed per unit mass (qe) and concentration (Ce).

Table 2: Langmuir and Freundlich isotherm constants for orange peel powder

Langmuir Isotherm			Freundlich Isotherm		
q_{max} (mg/g)	K_L (l/mg)	R^2	n	K_F (mg/g)	R^2
0.0166	0.01295	0.872	1.43	0.463	0.91

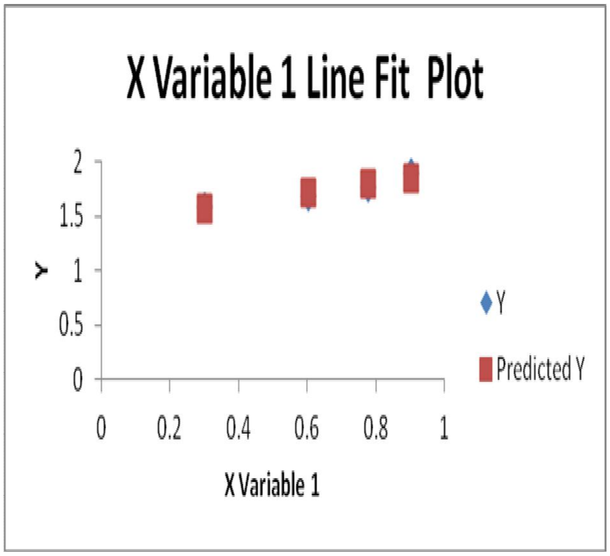

Figure 1: Correlation between log qe (Y-axis) and log Ce (X-axis) for orange peel powder

Neem Leaf Ash

The values of q_{max}, n and R^2 from the Langmuir and Freundlich isotherm data analysis are as shown in table 3. It shows that Freundlich isotherm is better fit than Langmuir isotherm. The figure 2 shows that the line fit between amounts of metal adsorbed per unit mass (qe) and concentration (Ce).

Table 3: Langmuir and Freundlich isotherm constants for neem leaf ash

Langmuir Isotherm			Freundlich Isotherm		
q_{max} (mg/g)	K_L (l/mg)	R^2	n	K_F (mg/g)	R^2
0.032	0.008	0.974	1.48	0.44	0.99

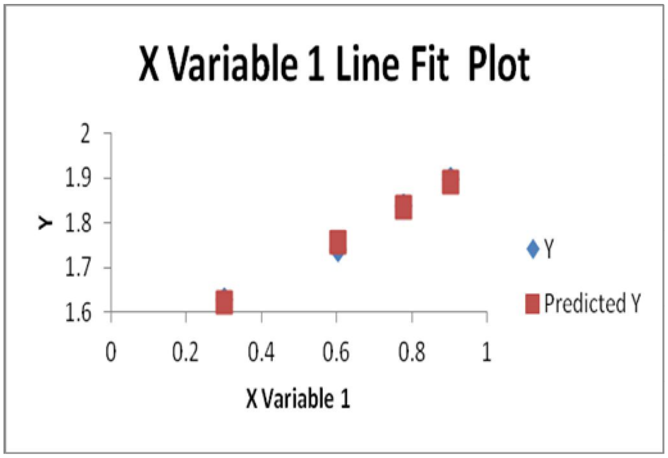

Figure 2: Correlation between log qe (Y-axis) and log Ce (X-axis) for neem leaf ash

Saw Dust

The values of q_{max}, n and R^2 from the Langmuir and Freundlich isotherm data analysis are as shown in table 4. It shows that Freundlich isotherm is better fit than Langmuir isotherm. The figure 3 shows that the line fit between amounts of metal adsorbed per unit mass (qe) and concentration (Ce).

Table 4: Langmuir and Freundlich isotherm constants for saw dust

Langmuir Isotherm			**Freundlich Isotherm**		
q_{max} (mg/g)	**K_L (l/mg)**	**R^2**	**n**	**K_F (mg/g)**	**R^2**
0.045	**0.006**	**0.900**	**1.324**	**0.691**	**0.946**

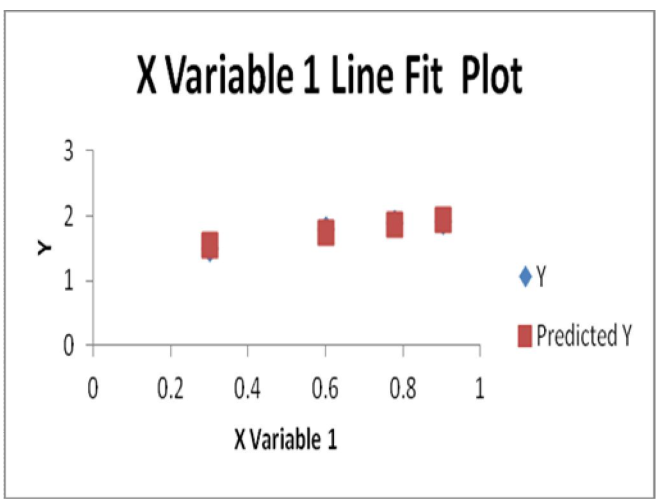

Figure 3: Correlation between log qe (Y-axis) and log Ce (X-axis) for saw dust

Coffee Husk

The values of q_{max}, n and R^2 from the Langmuir and Freundlich isotherm data analysis are as shown in table 5. It shows that Freundlich isotherm is better fit than Langmuir isotherm. The figure 4 shows that the line fit between amounts of metal adsorbed per unit mass (qe) and concentration (Ce).

Table 5: Langmuir and Freundlich isotherm constants for coffee husk

Langmuir Isotherm			**Freundlich Isotherm**		
q_{max} (mg/g)	**K_L (l/mg)**	**R^2**	**n**	**K_F (mg/g)**	**R^2**
0.039	**0.008**	**0.926**	**1.431**	**0.507**	**0.972**

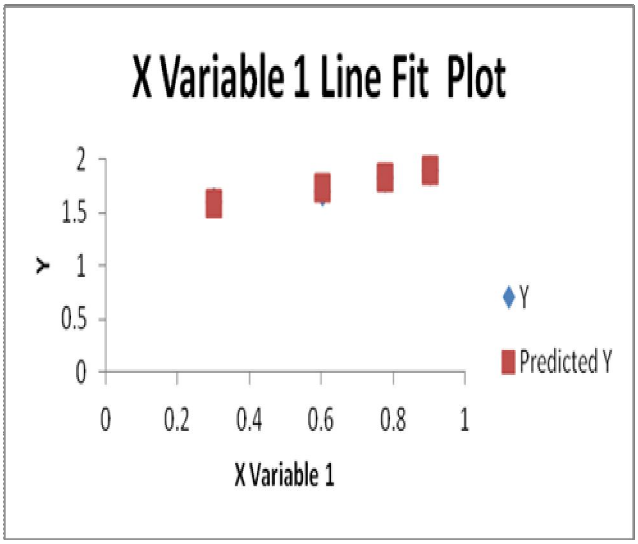

Figure 4: Correlation between log qe(Y-axis) and log Ce(X-axis) for coffee husk

8. RESULT

The adsorption capacity of orange peel powder, neem leaf ash, saw dust and coffee husk were studied for the removal of nickel by varying the different parameters such as initial concentration, dosage of the adsorbent and contact time. The removal efficiency of the adsorbents depends on concentration, dosage and contact time. From batch test, the maximum efficiency was obtained for saw dust with 98.08% followed by coffee husk with 97.75%, neem leaf ash with 97.43% and orange peel powder with 85.51%. The relation between removal efficiency and various parameters such as contact time, concentration and dosage were obtained using multiple linear regression analysis. From column study, it was inferred that all four adsorbents follow the Freundlichmodel and thus show multilayer adsorption. Since, the adsorbents used (orange peel powder, neem leaf ash, saw dust and coffee husk) are efficient in removing nickel from its synthetic solutions, they can be used as an alternative for commercially activated carbon. They are both available at low cost and are eco – friendly.

9. REFERENCES

1. Gopal Rathor, Neelam Chopra and Tapan Adhikari (2014), “Nickel as a pollutant and its management” in International Research Journal of Environmental Sciences, vol. 3(10), pp. 94-98, October 2014.
2. Sonali R. Dhokpande, Dr.Jayant P. Kaware, Sunil J. Kulkarni (2013), “Research for removal of nickel from wastewater: A Review” in International Journal of Science, Engineering and Technology Research (IJSETR), vol. 2, issue 12, December 2013.

3. M. Cempal, G. Nikel (2005), “Nickel: A review of its sources and environmental toxicology” in Polish J. of Envi. Stud., vol. 15, no. 3, pp. 375-382
4. Ferda Gonen and D. Selen Serin (2012), “Adsorption study on orange peel: Removal of Ni(II) ions from aqueous solution” in African journal of biotechnology, vol. 11(5), pp. 1250-1258, January 2012.
5. Jyoti Shah, Sanjeev Kumar, Sudha Sharma and Rajeev Sharma, Rapinder Sharma (2016), “Removal of nickel from aqueous solution using low cost adsorbents: A Review”, International Journal of Scientific Engineering and Applied Sciences (IJSEAS), vol. 2, issue 7, July 2016.
6. Sonali R. Dhokpande, Jayant P. Kaware (2017), “Biosorptive nickel removal by fruit peel waste: Batch, Kinetic and Isotherm Studies” in International Journal of Research Chemical Environment, vol. 7, issue 1(1 – 6), January 2017.
7. Syakirah Afiza Mohammed, Nor Wahidatul Azura Zainon Najib and Vishnuvarthan Muniandi (2012), “Durian Rind as a low cost adsorbent” in International Journal of Civil and Environmental Engineering (IJCEE-IJENS), vol.12, no. 04, December 2012.
8. Zahra Abbasi, Mohammad Alikarami, Eshagh Rezee Nezhad, Farzaneh Moradi, Vahid Moradi (2013), “Adsorptive removal of Co2+ and Ni2+ by peels of banana from aqueous solution” in University journal of chemistry, vol. 1(3), pp. 90-95, January 2013.
9. Sheel Ratan, Indu Singh, Joy Sarkar, and Naik RM (2016), “The Removal of Nickel from Wastewater by Modified Coconut Coir Pith” in Chemistry Sciences Journal, ISSN2150-3494, 7:3.
10. Hamidi A Aziza, Mohd N Adlana, Chieng S Huia, M S M Zaharia & B H Hameedb (2005), “Removal of Ni, Cd, Pb, Zn and colour from aqueous solution using potential low cost adsorbent” in Indian Journal of Engineering & Materials Sciences, vol. 12, pp. 248-258, March 2005.
11. Ashwin Shenoy, Jagadish H Patil, Sudhanva M Desai (2005), “Biosorption of nickel in aqueous solution using tamarind shell and carrot peel” in International Journal of Advance Research in Engineering, Science and Technology (IJAREST), ISSN(O):2393:9877, vol. 2, Issue 5, May 2015.
12. Y. B. Onnundi, A. A. Mamun, M. F. Al Khatib, Y. M. Ahmed (2010), “Adsorption of copper, nickel and lead ions from synthetic semiconductor industrial wastewater by palm shell activated carbon” in International Journal of Environment, Science Technology, 7(4), 751-758, Autumn 2010.
13. Arunkumar, C, Perumal, R, Lakshmi Narayanan, S and Arunkumar (2014), J, “Use of corn cob as low cost adsorbent for the removal of nickel (II) from aqueous solution” in International Journal of Advanced Biotechnology and Research (IJBR), ISSN 0976 – 2612, Vol. 5, issue 3, pp. 325-330, July 2014.
14. Muthusamy P., Murugan S. and Manothi Smitha (2012), “Removal of Nickel ion from industrial waste water using maize cob” in ISCA journal of biological sciences, vol. 1(2), pp. 7-11, June 2012.
15. Manjeet Bansal, Diwan Singh, V. K. Garg and Pawan Rose (2009), “Use of Agricultural Waste for the Removal of Nickel Ions from Aqueous Solutions: Equilibrium and Kinetic studies”, World Academy of Science, Engineering and Technology in International Journal of Civil, Environmental, Structural, Construction and Architectral Engineering, vol. 3, no. 3, 2009.
16. Tajun Meera Begum KM, Alhaji NMI, Ayeshamariam A, Vidhya VS and Jayachandran M (2016), “Removal of Ni (II) ions on to polymer Loaded Sawdust (PLSD) – Batch Adsorption Studies” in Fluid mechanics: Open Access, 3:2, 2016.
17. Lahieb Faisal Muhaisen (2017), “Nickel ions removal from aqueous solutions using sawdust as adsorbent: Equilibrium, Kinetic and Thermodynamic studies”, vol. 21, no. 03, ISSN 2520 – 0917, May 2017.
18. L Lisy, L Subha(2016), “Adsorption isotherm and kinetic studies of nickel (II) ions removal using neem bark charcoal and commercially activated carbon: A comparitive study” in International Journal of Advanced Science and Research, Volume 1, Issue 12, December 2016.
19. Dhananjay Chaturvedi and Omprakash Sahu (2014), “Adsorption of Heavy Metal Ions from Wastewater” in Global Journal of Environmental Science and Technology, ISSN–2360-7955, vol. 2(3), pp 020 – 028, March 2014.
20. Przemyslaw Bartezak, Malgorzata Norman, Lukasz Klapiszewski, Natalia Karwanska, Malgorzata Kawalec, Monika Baezynska, Marcin Wysokowski, Jakub Zdarta, Filip Ciesielezyk, Teofil Jesionowski (2015), “Removal of nickel(II) and lead(II) ions from aqueous solution using peat as a low-cost adsorbent: A kinetic and equilibrium study” in Arabian journal of chemistry, 2015.
21. Marques TL, Alves VN, Coelho LM, Coelho NM (2012), “Removal of Ni(II) from aqueous solution using Moringa oleifera seeds as a bioadsorbent” in Water Science Technology. 2012; 65(8):1435-40.
22. Rudre Gowda, A.G. Nataraj and N. Manamohan Rao (2012), “Coconut leaves as a low cost adsorbent for the removal of Nickel from Electroplating effluents” in International Journal of Scientific & Engineering Research (IJSER), vol. 2, Issue 12, ISSN 2229-5518, March 2012.