

## Experimental studies on removal of nickel using foundry sand

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### Abstract

The objective of the present study was to investigate the removal of nickel (II) by foundry sand. The scope of work included batch mode adsorption, column mode removal and isotherm studies. Basically, batch adsorption study depended on various parameters like optimum time, dosage, pH, particle size and speed. Maximum removal of 29.07% nickel was achieved at 60 min contact time, dosage of 7g, pH 7, and particle size of 500 micron and speed of 100rpm. The Langmuir and Freundlich isotherm study showed favourable condition for adsorption. The use of column adsorption study was investigated at optimum condition to study the feasibility for application in small scale industries. This column adsorption study showed that the results of flow rate of 15 ml 24 hrs<sup>-1</sup> and bed height of 1cm were feasible. The breakthrough curves were constructed for various ratios of adsorbents. The percentage removal of Ni in clay adsorbent, clay with foundry sand (1:20), clay with foundry sand (1:10), and clay with foundry sand (1:5) were found to be 68.11%, 59.28%, 54.28 and 48.30% respectively.

### Key words

Adsorption, Batch, Column, Foundry sand, Isotherm, Nickel removal, Soil contamination

### Introduction

Heavy metal pollution in the environment can originate from natural as well as manmade sources. The natural sources of heavy metal pollution are geological weathering and volcanic activities. Electroplating industries, battery manufacturing units are some of the man-made sources of heavy metal pollution (Yin *et al.*, 2009). Lata *et al.* (2008) illustrated that heavy metals are toxic at low concentrations to humans and other living organism. The presence of highly concentrated toxic metals such as lead, cadmium, copper, nickel, chromium and mercury in industrial waste waters is of great environmental concern since they exert detrimental impacts on the surrounding ecological systems, as well as pose, public health concerns.

The permissible limit of nickel in drinking water is 0.01 mg l<sup>-1</sup> and for discharge in industrial wastewater is 2.0 mg l<sup>-1</sup> in India (Kadirvelu *et al.*, 2001, Lata *et al.*, 2008). Ni (II) in higher concentration causes cancer of lungs, nose and

bone. Dermatitis (Ni itch) is the most frequent effect of exposure to Ni, such as coins and costume jewellery. Ni carbonyl [Ni (CO)<sub>4</sub>] has been estimated as lethal in humans at atmospheric exposures of 30 ppm for 30 min. Nickel poisoning causes headache, dizziness, nausea, vomiting, tightness of chest, dry cough, shortness of breath, rapid respiration, cyanosis and extreme weakness (Lata *et al.*, 2008).

Removal of nickel from industrial wastes can be done by adsorption principles by using sand. Though importance of this treatment is felt, its cost keeps the industrialists away from adopting the same. It is also fact that till date, no material proves to be a better adsorbent than commercial activated carbon (Lakshmi Narayanan *et al.*, 2013). Since the cost of activated carbon and its reactivation cost is high, technology of activated carbon remains at an inaccessible distance and its application does not find a place (Strkalj *et al.*, 2009). However, an attempt was made to employ a low cost material like sand to remove nickel from electroplating industry.

Certain organic compounds in wastewater are resistant to biological degradation and many others are toxic even at low concentration. Low concentration materials are not readily removed by conventional treatment methods. Sand and clay are used for removal of organic contaminant from waste water is wide spread. Its large surface area enhances the effectiveness of sand and clay for removal of organic compounds from wastewater by adsorption.

The widespread concern over cumulative toxicity and environmental impact of heavy metals like nickel, chromium, cadmium have led to extensive research in developing effective, economic and alternative technologies for removal of these potentially damaging substances from effluents and waste water (Rajalakshmi *et al.*, 2009; Yin *et al.*, 2009). Current technologies available for removal of metals from aqueous solutions include chemical precipitation, electrolysis, ion exchange and membrane filtration membrane processes such as reverse osmosis, ultra filtration, precipitation and ion exchange (Alyüz *et al.*, 2009; Ewecharoena *et al.*, 2009; Dabrowski *et al.*, 2004). Capital costs and operation and maintenance commitments are high for these methods. Precipitation processes generate large quantity of sludge, which needs further treatment and disposal. High operational costs, particularly due to high energy consumption associated with electrolytic and membrane processes, limits their use. Secondary wastes generated are sometimes difficult to treat. Although, sorption processes are efficient, they have their application limited by high cost of ion exchange resins and activated carbon, the most used sorbents. One potential strategy is to use low-cost natural materials as sorbents for the contaminants of concern. Due to their low cost these materials can be disposed off directly when their adsorptive capacity gets exhausted, rather than regenerating and reusing them.

Adsorption has gained relatively wide application due to the fact that there are several types of adsorbents that are commonly used. The maximum acceptable concentrations of nickel in drinking water standards varied from  $0.2\mu\text{g l}^{-1}$  to  $20\mu\text{g l}^{-1}$  (EPA, 2009; IS10500, 2004).

In view of the above the objective of the present study was to evaluate reuse of foundry sand as waste product for removal of nickel and to conduct batch and column adsorption studies using foundry sand and clay soil to remove Nickel.

### Materials and Methods

Foundry sand of FS-1 grade (without clay) was collected from Harihar Alloy factory, Tiruchirappalli, India. Double distilled water was used to prepare synthetic solution.  $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$  salt used was analytical grade. Chemicals such as HCl and NaOH were added to adjust the pH. The column diameter and height were 2 cm and 40cm respectively. The

weight of the column was 105gm. Glass wool was fed in the bottom column for 2cm.

**Batch study :** The Nickel solution was prepared at 80ppm by diluting 3.58gm of Nickel (II) sulphate salt in 1 litre of double distilled water. A batch study was conducted with varying the dosage of foundry sand are 1g, 2g, 3g, 4g, 5g, 6g, 7g and 8g in 100 ml of synthetic solution and for the batch periods of 15, 30, 45, 60, 75 and 90 min similarly, pH optimization was done by varying the pH from 1 to 9. Size adsorption study was performed by varying sieve size were 150, 250, 500 between 500-710, 710 and 1000 microns in 100 ml of synthetic solution. Another batch experiment was carried out to optimize the speed of shaker by varying the speed of 20, 40, 60, 80, 100, 120, 140 and 160 rpm. Finally, the Kinetics study was carried out for different initial concentrations from 10 to 80 ppm. The samples were analysed in UV spectroscopy.

**Column study :** The nickel solution was prepared at 100 ppm by adding 4.75 g of Nickel (II) sulphate salt to 1 l of double distilled water. A column study was conducted for different ratios of adsorbents Clay, clay with foundry sand (1:20), clay with foundry sand (1:10), clay with foundry sand (1:5). The bed height and depth of the synthetic solution was 1 cm and 10cm (15 ml) respectively. The flow rate was maintained for 15ml per 24hrs. The samples were collected and tested each day and this experiment was carried out for 8 days. The samples were analysed in UV spectroscopy.

After completion of the test contaminated clay materials sliced and analysed. The thickness of each sub layer was approximately controlled to 0.5 cm. Approximately, 2 g of soil was placed in 100 ml distilled water was added to conical flasks. The samples were placed in optical shaker speed of 100 rpm. The sample was withdrawn from shaker after 1 hr and filtered through centrifuge apparatus at speed of 6000 rpm run for 3 min. The samples were analysed in UV spectroscopy.

### Results and Discussion

Adsorption increases with increasing time and equilibrium is attained after shaking for 60 min. Therefore, in each experiment, shaking time was set for 60 min. Fig.1 shows that percent adsorption of Ni(II) ion increased with time as well as, adsorbent dosage. Due to availability of more and more adsorption sites for complexation of nickel (II) ions (Sprynsky *et al.*, 2006; Lata *et al.*, 2008,)

When adsorption of outer surface reached diffusion, nickel ions entered into the pores of foundry sand particles and were adsorbed by the interior surface of the particle. The range between optimum time was 30 to 60 min (Hasar 2003; Venkatesan *et al.*, 2014). The optimum removal of nickel was 25.43% n at 60 min.

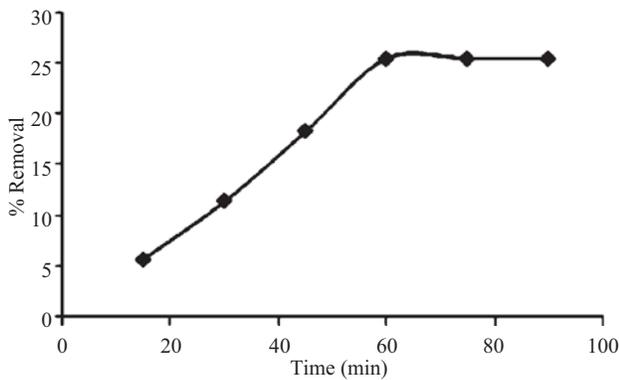


Fig. 1 : Effect of time on nickel removal for sand

The increasing trend of adsorbed nearly 29.07% removal of Ni at 7gm of adsorbent are shown in Fig. 2a. After reaching optimum dosage, decreasing tendency was noted upto 10 gm of adsorbent. It indicated that a direct proportion between dosage and percentage removal. The increasing amount of dosage could increase percentage removal. This could be recognized to the increase in the surface area of the adsorbent creating more reaction take places in molecules (Venkatesan *et al.*, 2014).

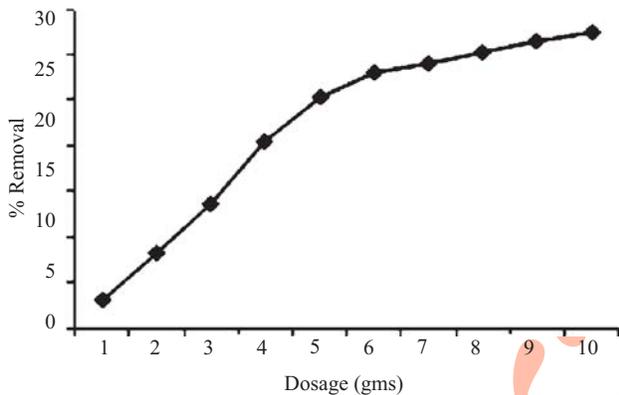


Fig. 2 (a) : Effect of dosage on nickel removal at 60minutes contact time for sand

A fixed volume (100ml) of synthetic solution was treated with 7 g of adsorbent with varying initial concentration up to 80 ppm. The test was carried out for 60 min contact time. The graph is plotted as shown in Fig. 2b with initial concentration against percent removal and defined that the initial concentration has a clear effect on the adsorption possible. The percentage of removal was high at initial concentration (60ppm) and then decreased. Percentage removal was found to be 31.92%.

Optimum particle size is one of the important factors in adsorption studies which controls the capital and operation cost. The synthetic solution is treated for various size of the adsorbent. The results indicated that the percentage removal increased with increase in sieve size and reached maximum removal at 500 micron (Fig. 2c). The contact time and dosages were found to be 60 min and 7 g respectively. The percentage of nickel removal was 34.72%.

Speed is also one of the parameters which influences adsorption. Fixed volume of synthetic solution was treated for various speeds (rpm) with 7g of adsorbent at 60 min contact time. Fig 2d reveals that adsorption was high at

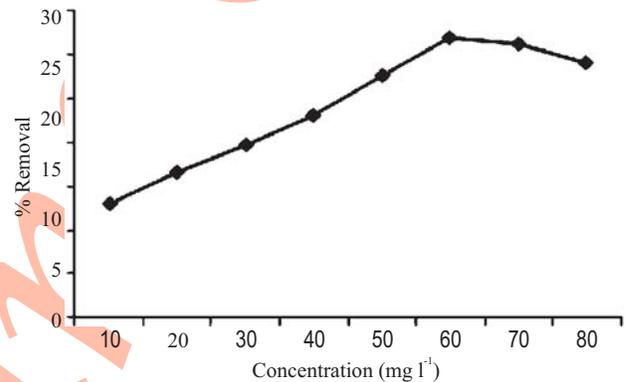


Fig. 2 (b) : Effect of initial concentration on nickel removal at 60 min contacts time for sand

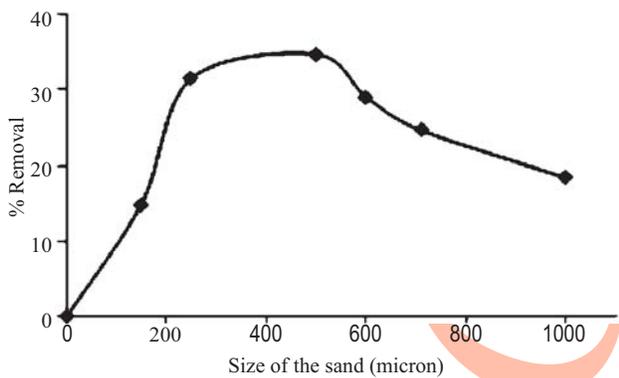


Fig. 2 (c) : Effect of particle size on nickel removal at 60 minutes contact time for sand

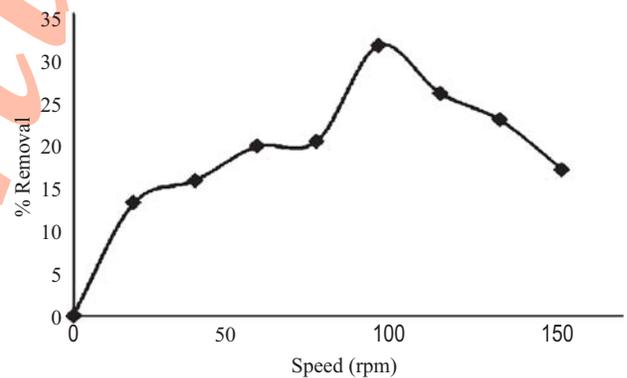
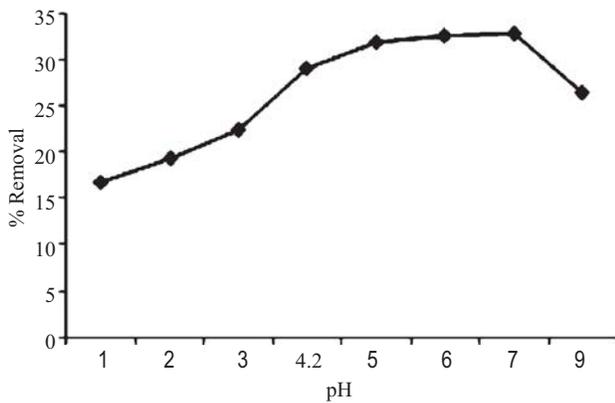
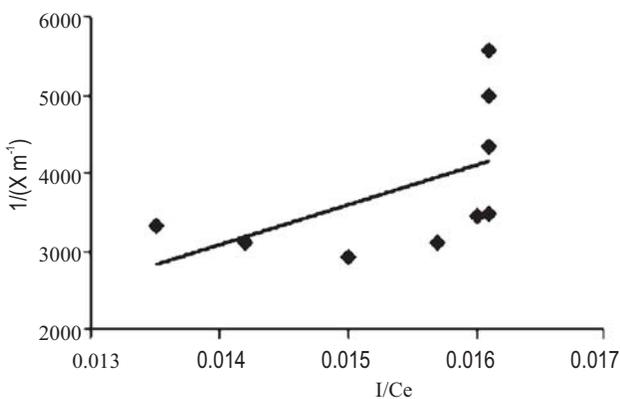


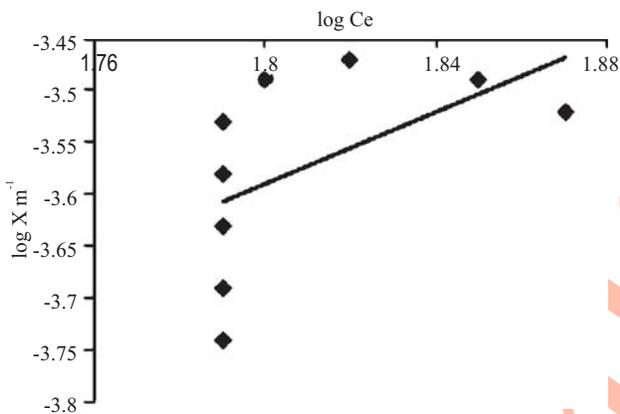
Fig. 2 (d) : Effect of speed on nickel removal at 60 min contact time for sand



**Fig. 3 :** Effect of pH on nickel removal with 7g of dosage at 60 min contact time



**Fig. 3(a) :** Langmuir isotherm for foundry sand



**Fig. 3(b) :** Freundlich isotherm for foundry sand

moderate speed. But in case of low and high speed, adsorption rate was low. The percentage of nickel removal was 31.3%.

The pH of the aqueous solution is an important parameter in the adsorption process. The role of hydrogen ion concentration varied from 1 to 9. Nickel removal percentage gradually increased with increasing pH and showed maximum removal at pH 5 to 7 (Fig. 3). The surface of the

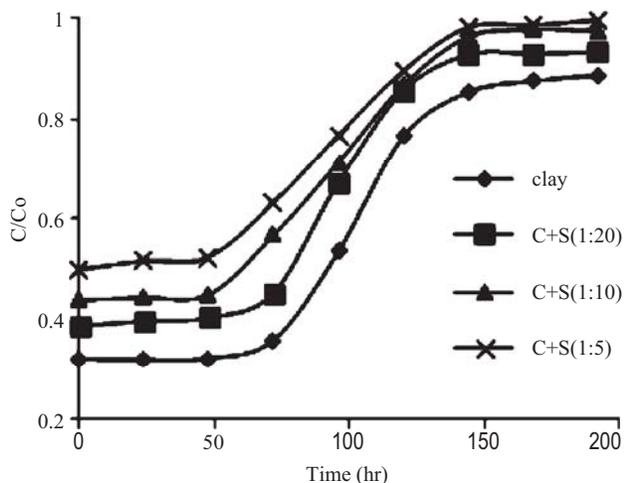
adsorbent bond to the proton at low pH. This would hold back the access, by repulsion forces to the surface functional groups by the metals. In case of high pH, more functional groups on the surface of the adsorbent was available for binding with the metals. It was observed that adsorption increased with increasing pH, where metal ions precipitated at optimum pH (5 to 7). The results showed that (Fig. 3) maximum retrieval was only possible between pH 5 to 7. It is found to be 32.7% of the removal. Same trend was observed by Pandey *et al.* (2007); Lata *et al.* (2008); Ewecharoena *et al.* (2009), respectively.

Langmuir adsorption isotherm is based on valence gap exist on the surface of the adsorbent and these gap is a capable of adsorbing one molecule. The presence of adsorbed molecules at one site did not affect the adsorption of molecules at adjacent site. A graph between  $1/C_e$  and  $1/(X/m)$  was plotted and it follow a linear path as shown in Fig.3.a. It showed that Langmuir isotherm efficiency of nickel adsorption by foundry sand. The intensity of adsorption ranged between 0 and 10, which implied stronger interaction between foundry sand and nickel (II) ions (Rengaraj *et al.*, 2007; Moodley *et al.*, 2013). It showed that favourable adsorption condition. Disposal of founder sand after use can be accomplished by secured and filling.

A graph between  $1/C_e$  and  $1/(X/m)$  was plotted and it followed a linear path as shown in Fig.3.b. Langmuir isotherm showed the efficiency of nickel adsorption for foundry sand  $q_m = 0.00014$ ,  $K_A = 0.143$ ,  $R_L = 0.08$ . As value  $R_L$  ranged between 0 and 1 it indicated favourable adsorption for nickel uptake.

The column study was conducted to predict nickle removal of nickle from clay and foundry sand. Initial concentration of nickle was taken as  $100\text{mg l}^{-1}$  and weight of adsorbent was taken as 5gm. A breakthrough curve was constructed between time (t), residual concentration (C) by initial concentration ( $C_0$ ) as shown in Fig.4. Breakthrough time and nickle removal was 72 hrs ( $3^{\text{rd}}$  day) and 68.11%, respectively. The weight of adsorbents (4.75 g of clay and 0.25 g of foundry sand) was found to be 5g. The breakthrough time and nickle removal was 72 hrs ( $3^{\text{rd}}$  day) and 59.28% respectively. The weight of the adsorbents (4.5 g of clay and 0.5 g of foundry sand) was 5 g. The breakthrough time and nickle removal were found to be 48 hrs ( $2^{\text{nd}}$  day) and 54.28%, respectively. The weight of adsorbents (4 g of clay and 1 g of foundry sand) was 5 g. The breakthrough time and nickle removal were found to be 48 hrs ( $2^{\text{nd}}$  day) and 43.30%, respectively.

The results showed uptake of clay, ratio of 1:20, 1.10 and 1: 5, respectively, for initial concentration of  $100\text{ mg l}^{-1}$  (Fig. 4). It illustrates the resulting breakthrough curves for Ni at inlet concentrations. The breakthrough curves showed



**Fig. 4 :** Effect of column adsorption for removal nickel using clay and foundry sand clay ,1:5,1:10 and 1:20 ratios

**Table 1 :** Breakthrough time and % removal of clay and foundry sand ratios

Adsorbents	Breakthrough time	%removal
Clay	72 hours(3 <sup>rd</sup> day)	68.11%,
Clay+Foundry sand (1:20)	72 hours(3 <sup>rd</sup> day)	59.28%,
Clay+ Foundry Sand(1:10)	48 hours (2 <sup>nd</sup> day)	54.28,

**Table 2 :** Layer analysis for columnar study

Adsorbents	Amount of the nickel adsorbed (after 8 days) ppm
Clay (upper)	198.28
Clay (lower)	134.14
Clay + Foundry Sand(1:20) upper	153.06
Clay + Foundry Sand (1:20) lower	102.72
Clay + Foundry Sand (1:10) upper	104.89
Clay + Foundry Sand (1:20) lower	66.67
Clay + Foundry Sand (1:5) upper	96.51
Clay + Foundry Sand (1:5) lower	58.17

typical “S” shape profile indicating adsorption for adsorbate of smaller molecular weight and simple structure as same trend as observed (Yin *et al.*, 2009). This is due to the formation of the mass transfer zone in the column. Once the solution containing the heavy metal is exposed to fresh layer of biomass, metal ions are sequestered by the adsorbent until the retained amount is in equilibrium with the influent concentration. At this time, the adsorbent is loaded to full capacity and that portion of the adsorbent becomes exhausted. The progressing in the direction of the flow, adsorption is occurring and the metal ion is being actively transferred from the liquid onto the adsorbent (Reddy *et al.*, 2013; Venkatesan *et al.*, 2014). Mass transfer zone will move

up through the column until it reaches the effluent port, whereupon heavy metal concentration in effluent begins to rise.

The breakthrough curves showed (Fig. 4) typical “S” shape profile indicating fixed-bed adsorption for adsorbate of smaller molecular weight and simple structure (Quek *et al.*, 2007, Yin *et al.*, 2009). Adsorption capacity of the adsorbent increased with increasing initial concentrations. Which might be due to the fact that driving force for adsorption was difference in nickel concentration between solution and adsorbent. Thus, high driving force due to high nickel concentration resulted in better column performance.

The breakthrough time was observed to be higher in case of clay. It suggested that clay adsorb nickel at faster rate and combination of clay and foundry sand also adsorbed nickel effectively, which might be due to high affinity of clay to nickel than clay and foundry mixtures. But more service period was required for nickel removal in case of foundry and clay mixtures. It is suggested that foundry sand also acted as a good adsorbent.

After completion of the test, the contaminated clay materials were sliced and analysed, and washed with distilled water by centrifugation. Further, the supernatant samples were analysed in UV spectroscopy. The contaminated clay and foundry sand layers after taking eight days for analysis are shown in Table.2. It is observed that the bottom of column adsorbed is less in top of column.

In the present study, clay and foundry sand composites were used to evaluate the of nickel removal from wastewater. Langmuir and freundlich adsorption isotherms were best fit for the experimental data and as conclusion the adsorbent selected for this study proved to be good adsorbent for removal of nickel. The adsorption affinity of nickel was reflected in both batch and column study. It is concluded that foundry sand was good adsorbent for nickel removal in both batch and column study. Nickel can be removed readily either from various types of electroplating industry or from solution of nickle salts. It is suggested that the adsorbent was cheap and readily available. Moreover, after the treatment it could be disposed off as a landfill.

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