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Recovery of Lead and Nickel from Rinsing Water of Electroplating Industries

F. E. Farghaly
Department of Chemical & Electrochemical Central Metallurgical, R & D Institute, Cairo, Egypt.

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ABSTRACT

In this study, a hydrometallurgical treatment for valuable metals recovery from rinsing water of electroplating industries is reported. This water contains high contents of Copper, Nickel, Lead, Zinc and Chromium ions. Copper is recovered by leaching in hydrochloric acid solution. The effects of acid concentration, temperature and treatment time were investigated. Lead and Nickel are removed by adsorption onto activated carbon from artificial binary-component aqueous solutions. The effects of pH, time, temperature, and amount of adsorbent were investigated. Maximum Lead adsorption was obtained at pH 5.5-6 and 80°C for 30 min., while for Nickel at pH 7 and room temperature for 60 min.

Introduction

Treatment of lead frame nickel-plating wastewater with newly designed electro-deposition reactor was studied. The electrolyte was circulated rapidly past the anode and cathode at a higher flow rate, allowing for improvement in efficiency and recovery, and then nickel electro-deposited on the surface of cathode. The effect of solution pH and current density were investigated as well. The results showed that the pH decreased during the electro-treatment, which is due to the production of H^+ ions on the anode surface, and the lower current density was accompanied with the higher current efficiency. The optimum pH value was found in the range of 2-2.5. Under the optimum conditions, over 99.9% Nickel would be removed (Chuh-Shun et al., 2011).

Removal of Nickel, Copper, Zinc and Chromium from synthetic and industrial wastewater by electro-coagulation was studied (Konstantinos et al., 2011). Parameters affecting the electro-coagulation process, such as initial pH, current density, initial metal ion concentration, COD and contact time were investigated. Best removal capacity for all studied metals was achieved in the pH range 4.8. Mixed solutions containing the same concentrations of all metals, i.e. 75, 150 and 300 mg/L were tested. Nickel, Copper and Zinc showed similar removal rates. They were completely removed in 20, 40 and 50 minutes respectively, while for the corresponding complete removal of Chromium 40, 60 and 80 minutes were needed (Konstantinos et al., 2011).

Recovery of Nickel ions from electroplating rinse water using Hectorite Clay was studied. Batch mode adsorption experiments showed that the ion-exchange process was spontaneous and exothermic under different temperatures. The maximum adsorption capacity obtained was 62.24 mg/g at pH 7-8, the optimum adsorbent dosage and contact time was found to be 2 g/L within 5 hours (Ramamurthi et al., 2009).

Electrochemical reactor with rotating cylinder electrode for optimum electrochemical recovery of nickel from plating rinsing effluents was studied (Hernández-Tapia, 2013). The pH control around 4 was crucial to yield high purity nickel, and thus prevent the precipitation of hydroxides and oxides. Macro electrolysis experiments were systematically conducted to analyze the impacts of the applied current density in the recovery efficiency and energy consumption, particularly for very diluted effluents (100 and 200 ppm Ni(II)), which present major recovery problems. Promising nickel recoveries in the order of 90 % were found in the former baths using a current density of 3.08 mA cm^{-2}, and with overall profits of 9.64 and 14.69 \$ kg^{-1}, respectively.

Nickel is a toxic heavy metal ion that is widely used in silver refineries, electroplating, Zinc base casting and storage battery industries (Kadirvelu et al., 2001). It is essential to remove Ni^{2+} ions from industrial wastewater before being discharged. Thus, pretreatment such as chemical reduction, ion exchange, reverse osmosis, electro-dialysis, and activated carbon adsorption were carried out. Since the costs of these processes are rather expensive, the use of agricultural residues or industrial by-product of biological activities have been received with considerable attention (Hasar et al., 2000). The major sources of Nickel contamination to water comes from industrial process such as electroplating, batteries manufacturing, mine, metal finishing and forging. Different methods were investigated and applied to remove Nickel ions from waste water such as adsorption, chemical precipitation, ion exchange, filtration, membrane separation, and reverse osmosis. Adsorption is widely used because it is cost-effective and simple such as seaweeds (Vijayaraghavan et al., 2005), crab shell (Vijayaraghavan et al., 2004), dried aerobic activated sludge (Aksu et al., 2000).

The Nickel ions removal by activated carbon from aqueous solution has been investigated under several conditions such as pH, carbonization temperature of husk, initial concentration of metal ions, contact time, and adsorbent concentration (Halil H, 2003).

The adsorption capacity for Nickel ions from aqueous solutions increased significantly onto the surface of the oxidized carbon nano-tubes CNTs compared to that on the as-produced CNTs. The effects of adsorption time, pH and initial Nickel ions concentration on the adsorption uptake of Nickel ions for both the as-produced and oxidized CNTs were investigated at room temperature (Munther et al., 2007).

Tele:
E-mail addresses: femam66@yahoo.com

Nickel waste tailings are produced during the hydrometallurgical leaching of nickel-cobalt ore slurry with concentrated sulfuric acid. For reducing the heavy loading of leachable Nickel that apparently engenders long-term stability, the tailings are practically leached with 2% sulfuric acid to recover valuable Nickel at specified conditions. Approximately 5.98 - 7.69 g/L of Nickel was recovered in the final acidic solution at a purity of above 70 % (Zhuang et al., 2006).

The ion exchange is widely used for the recovery and removal of metals from process and waste streams in chemical process industries. A set of mass balance equations that take into account possible aqueous complexation reactions was used to establish the pH diagram of Ni^{2+} ions species in the presence of anionic ligand citrate or phosphate (Juang et al., 2006).

Nickel was recovered from pickling acid solutions of crude ore using boric acid as catalyst by ultrasound assisted electrolysis. A method of removing Nickel from Nickel picking acid wastewater comprising treating said pickling acid wastewater metal in an electrolytic cell, which cell comprises a support of a non-electrically conducting material which is provided with a cathode comprising a carbon material, a current feeder for the cathode, an anode spaced from said cathode and a current feeder for the anode, where in the treating comprises introducing said picking acid wastewater into the electrolysis cell from which said Nickel is to be removed along with a catalyst and subjecting said electrolysis vessel to ultrasonic waves by placing said electrolysis chamber inside another chamber capable of producing ultrasound of varying frequency ultrasound (Robina F., 2011).

The removal of Nickel from aqueous solutions under different experimental conditions using a Ceralite IR 120 cationic exchange resin (CXR) as an adsorbent was investigated by batch adsorption techniques. Batch studies indicated that the adsorption decreased with increasing initial concentration of Ni^{2+}. Maximum Ni^{2+} ions removal was observed near a pH of 5. The adsorption process obeys the Langmuir and Freundlich adsorption isotherms (Senthil et al., 2010).

Materials and Methods

Sample preparation and characterization

Banha Company, Egypt, supplied 20 L of the spent rinsing water of electro-plating industries, used in this study. The sample was characterized by means of X-ray Diffraction (XRD), X-ray Fluorescence (XRF) and Inductive Coupled Plasma (ICP).

Leaching tests

According to the selected experimental conditions (Zhuang et al., 2006) a pure copper chloride was obtained by adjustment pH at 6.8 with 1 M HCl.

Adsorption active carbon

Pure activated carbon was used for the adsorption of Lead and Nickel ions from waste solution. The activated carbon was boiled twice with de-ionized water and dried in an oven at 110˚C for 6hrs prior to use. The pH of Lead and Nickel ions solution was adjusted by HCl solution. Adsorption of the heavy metal ions was carried out by agitation of 1 gm of the activated carbon with 50 ml of Lead and Nickel ions solution in 100 ml round bottom flask for 30 min. at room temperature. All experiments were carried out at agitation speed of 100 rpm.

The pH was kept less than 7.7 throughout the experiments to prevent precipitation of metal ions as hydroxides. The pH value of solution was measured using pH meter (Cobrscan 510 Model). The heavy metal ions concentration was determined using complexmetry technique with EDTA.

Results and Discussion

Table 1. The chemical composition of the spent solution

Copper	120	g/L
Lead	1.675	g/L
Zinc	1.024	g/L
Nickel	10	mg/L
Chromium	1.9	mg/L

Fig. 1 shows the effect of volume ratio of 1M HCl on the pH of spent solution at room temperature. The pH increases from 9.2 passing through a maximum of 9.5 with addition of equivalent volume of 1M acid. Addition of extra volume of acid decreases the pH towards the acidic range of 6.8 after addition of five times of the acid. Precipitation of basic copper chloride salt takes place in this pH range. Fig.2. shows the XRD of the solution after evaporation process. Table 2, shows the chemical analysis by XRF to remain solution after precipitate copper chloride.

Fig.3. shows the removal rates of Pb^{2+} & Ni^{2+} ions as function of solution pH measured after adsorption equilibrium was attained. The resulting data for metal removal at pH around 7 were accumulated in a narrow pH range in the adsorption equilibrium state; metal removal was enhanced from pH 5-7 in which Pb was always adsorbed onto the activated carbon to greater extent than Ni for metal concentrations. The metal adsorption rate does not change much even if the pH drifts between 5.5 - 7 as long as HCl is not added to the solution from our previous study (Juang et al., 2006). The effect of pH on the adsorption was attributed to interactions between ions in solution and complexes formed at the adsorbent surface. Ni^{2+} in aqueous solution can form different species whose presence depends on the solution PH is well documented. The predominant ionic species is Ni^{2+} for $pH < 7$, whereas Ni (II) is present mainly as Ni^{2+} and $Ni(OH)_2$, and in lesser quantities as $Ni(OH)^+$ for pH between 8 and 9. Since all the experiments were carried out at a maximum pH of 7 because at higher pH means higher OH^- anions in the solution that can bind with Ni^{2+} ions to hydroxide complexes form as the Nickel hydroxide.

Fig.4. shows changes in removal rates of Pb^{2+} & Ni^{2+} ions as function of temperature measured after adsorption equilibrium were attained. Increasing temperature, increases adsorption of Pb^{2+} up to 80% but decreases the adsorption of Ni^{2+}. The isotherm of Ni^{2+} onto activated carbon either was independent of temperature or the effect of temperature was so minimal as to be masked by experimental error. Fig. 5 and 6, show the effect of time onto adsorption of Pb^{2+} at 80°C and Ni^{2+} at room temperature. Increasing time increases the adsorption of Pb^{2+} and Ni^{2+} up to 80% at 30 and 60 min, respectively.

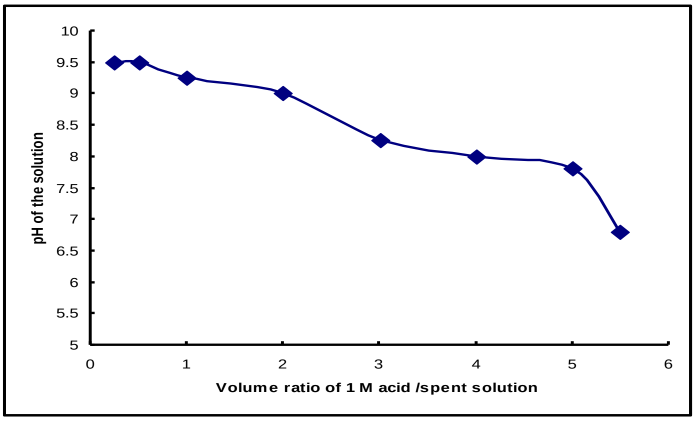

Fig 1. Effect of volume ratio of 1M HCl on the pH of spent solution at room temperature

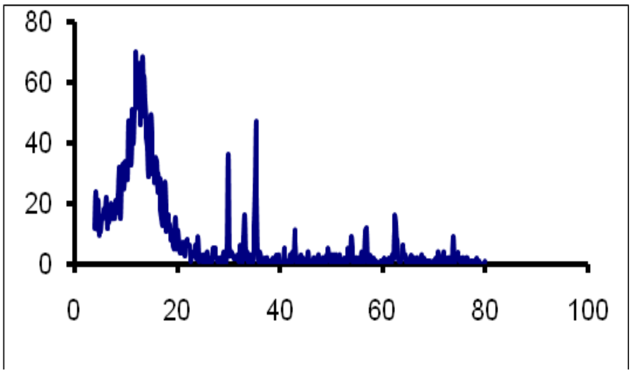

Fig 2. XRD of the remaining solution after the evaporation process

Table 2. The chemical analysis of remain solution

Compound Formula	Concentration (%)
Na_2O	0.162
MgO	0.044
Al_2O_3	0.092
SiO_2	0.128
P_2O_5	0.007
SO_3	0.143
CaO	0.104
MnO	0.083
Fe_2O_3	0.062
NiO	10.049
CuO	0.676
ZnO	0.022
Rb_2O	0.040
PbO	37.683
Cl	50.693
Br	0.012

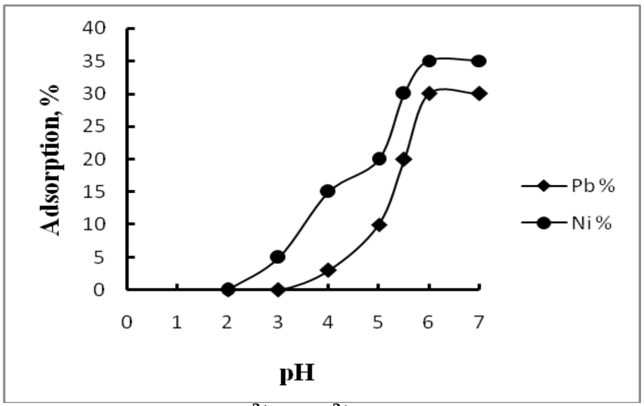

Fig 3. Adsorption of Pb^{2+} & Ni^{2+} ions onto activated carbon as a function of pH

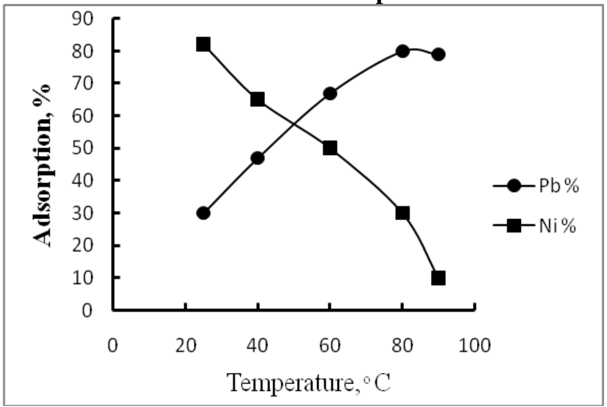

Fig 4. Adsorption of Pb^{2+} & Ni^{2+} onto activated carbon as a function of temperature.

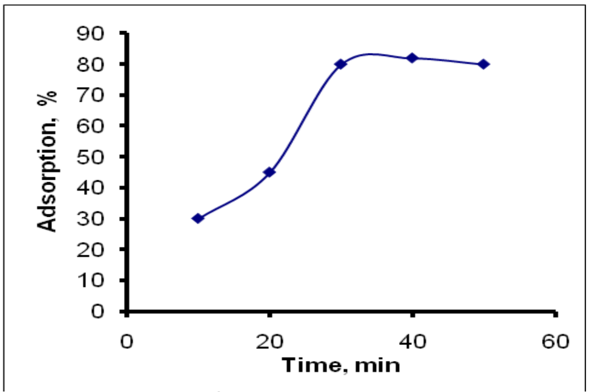

Fig 5. Adsorption of Pb^{2+} onto activated carbon as a function of time at 80°C

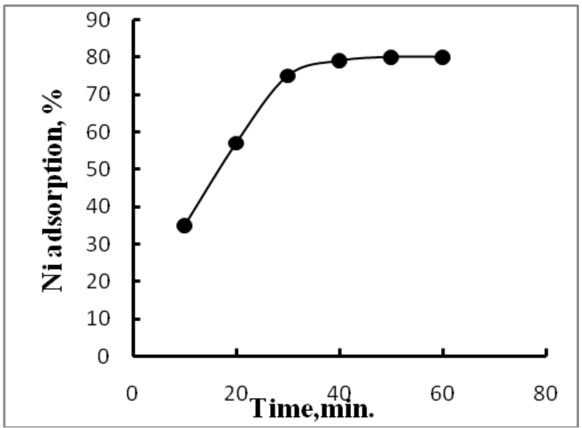

Fig 6. Adsorption of Ni^{2+} onto activated carbon as a function of time at room temperature.

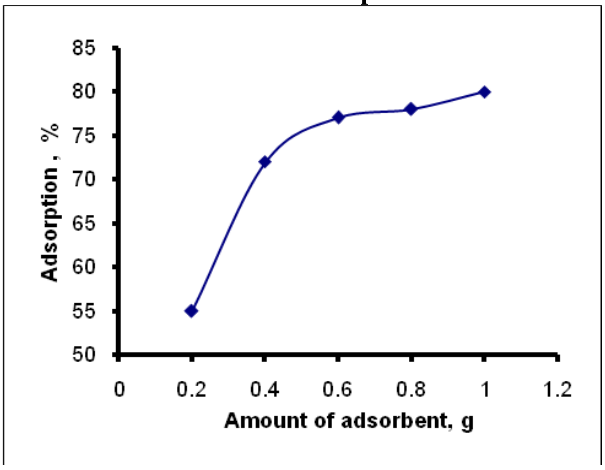

Fig 7. Effect of adsorbent doses on adsorption process

The adsorption of Pb^{2+} at different doses of carbon active is shown in Fig. 7. The adsorption increases as the adsorbent dose is increased. It may be concluded that by increasing the adsorbent dose, the removal efficiency of carbon increases and that results in the increase of removal efficiency.

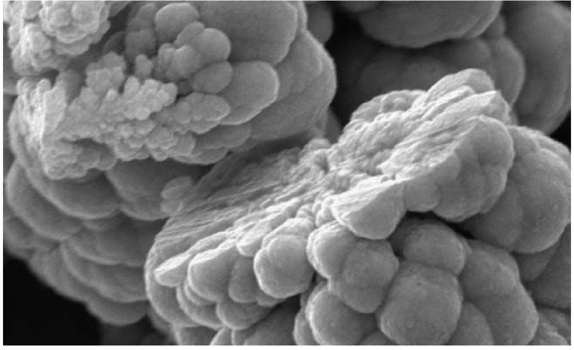

Fig 8. SEM micrograph of Ni (10μm)

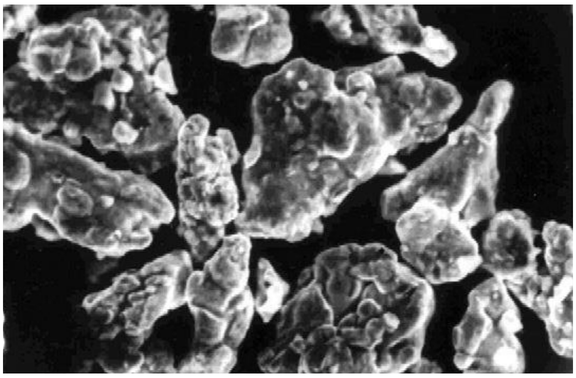

Fig 9. SEM picture of nickel powder. (10µm)

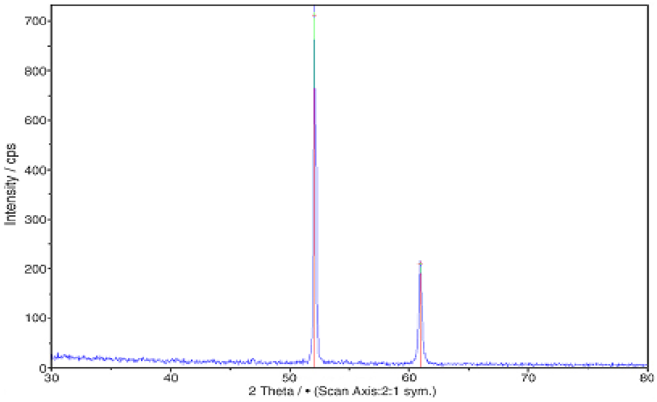

Fig 10. XRD of nickel powder

Conclusions

The simultaneous adsorption of Pb^{2+} & Ni^{2+} ions onto activated carbon was examined. The following conclusions were obtained from the study.

1- The activated carbon is covered by monolayer of Pb^{2+} & Ni^{2+} ions, and the number of maximum adsorption site for Pb^{2+} on the activated carbon is as great as that of Ni^{2+}.

2- The solution pH plays a very important role for Pb^{2+} & Ni^{2+} adsorption onto activated carbon.

3- At pH < 2, Pb^{2+} & Ni^{2+} ions did not adsorb. The adsorption isotherm increased drastically as the pH was increase from 2 to 7 and maximum adsorption uptake was achieved at pH 6 to adsorbed Pb^{2+}, and at pH 7 to adsorbed Ni^{2+}.

4- The effect of pH on the isotherm is attributed to interactions between ions in solution and complexes formed on the carbon surface.

5- The adsorption isotherm of Ni^{2+} onto activated carbon was not dependent on temperature while on Pb^{2+} adsorption temperature changes produced an unusual adsorption response, first increasing then remaining stable as temperature increased.

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