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Comparative Evaluation of Leaching behavior of Valuable Metals from Zinc Plant Residue in Ammonia-Ammonium Carbonate Solution

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ABSTRACT

In Iranian zinc plants, Ni-Cd filtercake is produced as zinc plant residue containing about 51.72% zinc, 12.52% cadmium and 3.03% nickel. In this study, ammonia leaching method for recovery of nickel in ammonium hydroxide and ammonium carbonate solution was investigated. First, pretreatment process was performed by sodium hydroxide on filtercake to reduce the zinc element and concentrate nickel. Then the ammonia leaching was performed on the achieved solid from pretreatment process. The optimum condition for pretreatment process was determined as follows: sodium hydroxide: 8M; liquid to solid ratio: 10:1; temperature: 75 °C; time: 60 minute. Under the mentioned condition, 96.24% of zinc was removed. In the ammonia leaching of nickel the parameters such as concentration of ammonium hydroxide and ammonium carbonate, liquid to solid ratio, stirring speed and time were investigated. The optimum condition for ammonia leaching was determined as follows: temperature 45 °C; ammonium hydroxide 5 mole; ammonium carbonate 1 mole; liquid to solid ratio 8:1; stirring speed 400 rpm and time 90 minute. Under the optimum condition 98.34 % of nickel was recovered.

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INTRODUCTION

The amount and variety of waste materials have increased with the growing of technology and population. Of the priority pollutants, heavy metals cause adverse effects on aquatic ecosystem by entering into the food chain and accumulating in living organisms. The introduction of metal contaminants into the aquatic system has various sources a few of which are atmospheric spread, originating from smelting processes and fuel combustion, industrial leaks and effluents, land application of sewage materials and leaching of garbage. In metal extraction processes, large amounts of various solid wastes including flotation tailings, slags, slimes, filtercakes, flue dusts, etc. are generated. These wastes become activated due to the process applied such as grinding, leaching, roasting, leaching, roasting, smelting, quenching, etc. Exposure of these wastes to atmospheric oxygen and moisture results in solubilization of toxic metals which may seriously affect the water quality and biological life in surface waters. The potential release of toxic heavy metals from such by-products and waste materials to the surface and ground water are of particular concern [1].

In lead and zinc plants located in Zanjan, Iran, a leach-electrolysis process is utilized for zinc production. In this process, great amounts of filtercakes as by-product are generated daily. These wastes are retained for a future zinc and lead recovery and dumped in open stockpiles where they may cause heavy metal pollution problems [2].

Different studies have been carried out throughout the world to recover valuable metals from zinc plant residues (ZPR). For example, Kul and Topkaya studied the recovery of germanium and other valuable metals from ZPR of Cinkur Zinc Plant. In another study, Wang and Zhou recommended a hydrometallurgical process for the production of cobalt oxide after inspecting the recovery of cobalt from ZPR. Raghavan *et al.* described hydrometallurgical processing of lead-bearing materials generated at zinc plants [3].

Among the generated filtercakes, the most toxic of them is Cd-Ni filtercake; certainly for its high cadmium content.

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The high nickel content associated with some of the zinc concentrates represents a metallurgical challenge related to the downstream processing of purification filtercakes since nickel is precipitated with cadmium and copper during zinc powder cementation process used to purify the zinc electrolyte prior to the electrowinning. This poses potential difficulty in subsequent processing of Cd–Ni purification residue. This has given rise to several process strategies such as selective cementation, selective leaching, and selective electrowinning, which have been examined for the separation of cadmium from nickel, all with less success [4-8].

Selective sulfation of zinc plant leach and purification residues with such sulfating reagents as sulfuric acid and elemental sulfur has been extensively reported and used in zinc industry [9]. This process, so called as baking, converts the metal values to water-soluble sulfates at relatively low temperatures. According to the authors' experience, the sulfation of Cd–Ni zinc plant purification residues is readily possible with concentrated sulfuric acid at 300 °C. However, this process offers no selectivity of zinc and cadmium over nickel. The purpose of this research is therefore to evaluate the possibility of selective separation of metals from ZnO–CdO–NiO mixture with the weight ratios similar to those happening in some zinc plant purification residues. For this reason, ammonium chloride baking of the mixture was considered as a possible means for the defined goal [10-11].

In this study, the leaching conditions of nickel and cadmium from Cd–Ni filtercake with ammonia-ammonium carbonate is investigated and the leaching behavior of nickel and cadmium is studied.

MATERIALS AND METHODS

Material:

The investigation was carried out on cold filter cake (CFC) Residue sample from Iranian Zinc Mines Development Company (IZMDC). The filter cake was dried at 110 °C for 24 h and then was crushed to –106 µm size using a ball mill. Sample was analyzed by chemical method and examined by X-ray powder diffraction (XRD). Chemical composition of CFC was determined by the atomic absorption. The data is tabulated in Table (1).

Table 1: Chemical composition of filter cake produced in Ni–Cd purification step.

Elements	Zn	Cd	Ni	Fe	Mn	Cu	Pb
Wt, %	51.72	12.52	3.03	0.038	0.11	1.92	1.26

Mineralogical analysis performed using Philips PW-1800 model X-ray diffractometer, indicated that NiO, CdO and ZnO were the major mineralogical phases in the filter cake (Fig.1).

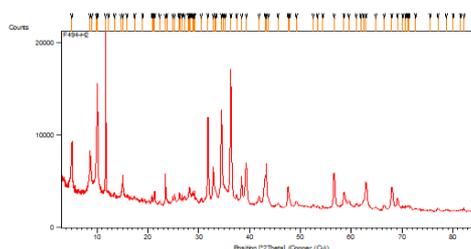


Fig. 1: X-ray diffraction analysis of cold filter cake (CFC).

Experimental Procedure:

Pretreatment of filtercake:

Cold filter cake was ground into certain size (100 µm) and then used in the research. The selective separation of zinc was carried out using sodium hydroxide. The tests were carried out at laboratory scale in a thermostatically controlled stirrer (1 liter beaker). When the solution temperature reached the desired value, CFC powder was added to the solution with an initial volume of 500 ml. Samples of known volume were removed from pulp at timed intervals. The samples were immediately vacuumed filtered, diluted and analyzed for zinc and other elements.

Ammonia- ammonium carbonate leaching tests:

Ammonia-ammonium carbonate leaching tests were carried out in a thermostatically controlled water bath, equipped with a digitally controlled thermometer. Affecting factors, such as ammonia and ammonium carbonate concentration (M), temperature (°C), liquid: solid ratio (L/S), and time (minute) were studied. Leaching of CFC was carried out so that the desired amount of residue achieved from pretreatment step was added to specific volume of solution, setting the temperature and stirring rate at the desired and required values. The range and

values of employed parameters for leaching tests are given in Table 2.

Table 2: The parameter values for ammonia-ammonium carbonate leaching of residue produced from pretreatment stage.

Parameter	Value
Ammonia concentration (M)	3, 5, 7
Ammonium carbonate concentration (M)	0, 0.3, 0.5, 1
Temperature (°C)	25, 45, 60, 75, 90
Phase ratio (L/S)	2, 4, 6, 8
Time(minute)	15, 30, 45, 60, 90, 120, 150, 180

RESULT AND DISCUSSION

Pretreatment of CFC for recovery of zinc:

Alkaline leaching with sodium hydroxide can be exploited in separating cations, since only certain metal oxides such as zinc oxide and aluminum oxide show amphoteric behavior and dissolve in it. The best examples of amphoteric compounds are found with metal hydroxides such as zinc hydroxide $[Zn(OH)_2]$ and aluminum hydroxide $[Al(OH)_3]$. Therefore, in this study sodium hydroxide was chosen as the selective alkaline agent since it is easy for Zn to be leached because of non-metallic and amphoteric character, but Ni and Cd couldn't be leached out [12]. Pretreatment process was performed under the optimum condition of: sodium hydroxide: 8M; liquid to solid ratio: 10:1; temperature: 75 °C; time: 60 minute. Under the mentioned condition, 96.24% of zinc was removed.

Ammonia-ammonium carbonate leaching tests:

a. Effect of concentration of ammonia and ammonium carbonate:

Three different Ammonia concentrations (3, 5, and 7 mol/l) and four different Ammonium carbonate concentrations (0, 0.3, 0.5 and 1 mol/l) were used in the experiments. A plot of metal content leached out into solution against ammonium carbonate concentration was presented in Fig.2 on the condition that 20 g of residue of pretreatment stage was leached for 1 h at 45 °C when L/S=6:1.

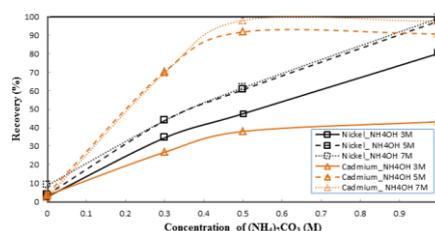


Fig. 2: Effect of ammonium carbonate concentration on metal leaching extraction (temperature=45 °C, L/S=6:1, time=1 h).

The results indicated that dissolution of Ni is dependent on the increase in the ammonium carbonate concentrations. Also it was resulted that dissolution of Ni is not so dependent on ammonium carbonate concentrations.

The Ni extraction efficiency after 1 h at different ammonium carbonate concentrations was found to be 3.79, 43.86, 60.55 and 97.78% respectively. Also the Cd extraction efficiency after 1 h at different ammonium carbonate concentrations was found to be 4.12, 70.48, 91.84 and 90.74% respectively. For economy and due to the constant Cd leaching recovery at high ammonium carbonate concentrations, the ammonium carbonate concentration 1 M is considered the optimal.

In Fig.3, the effect of the ratio of ammonia to ammonium carbonate is presented. As it is shown, at lower ratios nickel and cadmium extraction is higher.

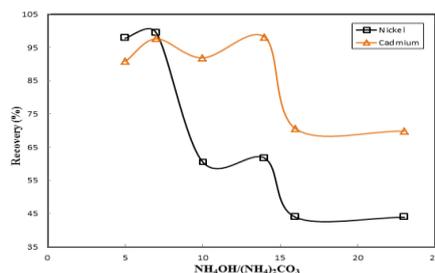


Fig. 3: Effect of $\text{NH}_4\text{OH} / (\text{NH}_4)_2\text{CO}_3$ ratio on metal leaching extraction (temperature=45 °C, L/S=6:1, rpm=600, time=1 h).

b. *Effect of liquid to solid ratio:*

Fig.4. gives the recovery as a function of liquid to solid ratio (volume of liquid/ weight of solid) at 45 °C, $\text{NH}_4\text{OH} / (\text{NH}_4)_2\text{CO}_3$ ratio at 5 and time for 1 h. These results showed that the liquid–solid ratio (L/S) had obvious effect on both Ni and Cd leaching recovery. Nickel recovery increased from 28.44% at L/S 2 to 99.29% at L/S 8. Also, cadmium recovery increased from 29.34% at L/S 2 to 97.08% at L/S 8 Therefore, L/S=8:1 with the maximum Ni and Cd recovery is considered the optimized phase to liquid ratio.

It is indicated that at L/S ratio of 4 Ni and Cd are separated but at other ratios Ni and Cd recovery is near together.

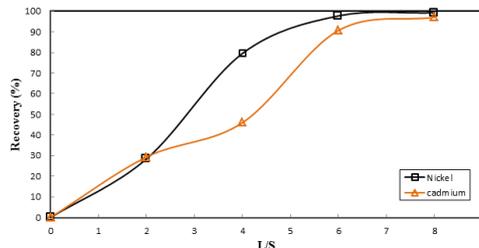


Fig. 4: Effect of L/S on Ni and Cd leaching recovery (temperature=45 °C, $\text{NH}_4\text{OH} / (\text{NH}_4)_2\text{CO}_3$ ratio= 5, time=1 h).

c. *Effect of leaching temperature:*

The effect of temperature on metal dissolution is illustrated in Fig.5. The results show that temperature doesn't play a key role leaching behavior of Ni and Cd in ammonia-ammonium carbonate medium. As it is shown after the temperature of 45°C, the recovery of both nickel and cadmium is downfallen. The Ni leaching recovery decreased from 3.79% at 45 °C to 0.008% at 90 °C and Cd leaching recovery decreased from 4.12% at 45 °C to 0.002% at 90 °C.

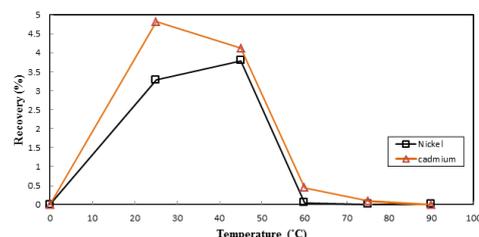


Fig. 5: Effect of temperature on Ni and Cd leaching recovery ($\text{NH}_4\text{OH} / (\text{NH}_4)_2\text{CO}_3$ ratio =5, L/S=6:1, time=1 h).

d. *Effect of leaching time:*

A series of experiments were carried out to clarify the effect of time on the dissolution of Ni and Cd when $\text{NH}_4\text{OH} / (\text{NH}_4)_2\text{CO}_3$ ratio at 5, temperature 45 °C, and L/S 6:1. The effect was studied at eight levels; 15, 30, 45, 60, 90, 120, 150 and 180 minute. The results plotted in figure 6 indicated that Ni and Cd dissolution increases with time up to 60 minutes and after that Ni dissolution remains nearly constant and Cd dissolution doesn't follow an orderly pattern. Nickel and cadmium leaching recovery is 97.78% and 90.74% respectively at the time of 60 minute. The time of 90 minute is considered as optimum time due to low dissolution of Cd and high recovery of Ni.

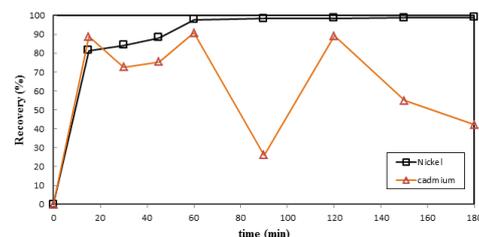


Fig. 6: Effect of time on Ni and Cd leaching recovery (temperature=45 °C, $\text{NH}_4\text{OH} / (\text{NH}_4)_2\text{CO}_3$ ratio =5, L/S=6:1).

Conclusion:

In this study, recovery of nickel and cadmium from zinc plant residue, namely Cold Filter Cake (CFC), was studied. CFC contains significant amount of Zn, Ni and Cd. As pretreatment process 96.24% of zinc is removed from CFC using sodium hydroxide. Then the behavior of Ni and Cd leaching in ammonia-ammonium carbonate medium is investigated. The results show that in ammonia-ammonium carbonate medium is not an important affecting parameter. The optimum condition for ammonia leaching was determined as follows: temperature 45 °C; NH₄OH / (NH₄)₂CO₃ ratio 5; liquid to solid ratio 8:1; stirring speed 400 rpm and time 90 minute.

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REFERENCES

- [1] Altundog˘an, H.S.M., R. Erdem, A. Orhan, F. O˘ zer, 1998. Tu˘men, Heavy metal pollution potential of zinc leach residues discarded in C, inkur plant, Turkish J. Eng. Environ. Sci., 22: 167-177.
- [2] Safarzadeh, M.S., D. Moradkhani, M. Ilkhchi, N. Hamedani Gholshan, 2007. Determination of the optimum conditions for the leaching of Cd–Ni residues from electrolytic zinc plant using statistical design of experiments. Separation and Purification Technology, 58: 367-376.
- [3] Moradkhani, D., B. Sedaghat, M. Khodakarami, I. Ataei, 2014. Recovery of valuable metals from zinc plant residues through separation between cobalt and manganese with N-N reagent, Physicochem. Probl. Miner. Process, 50(2): 735-746.
- [4] Safarzadeh, M.S., 2010. The scale up of cadmium production, scale up in metallurgy, In: Lackner, Maximillian (Ed.), 1st edition. Process. Eng. Technische Universitat Wien. ISBN: 9783902655103.
- [5] Safarzadeh, M.S., D. Moradkhani, 2010a. The effect of heat treatment on selective separation of nickel from Cd–Ni zinc plant residues. Sep. Purif. Technol, 73: 339-341.
- [6] Safarzadeh, M.S., D. Moradkhani, 2010b. The electrowinning of cadmium in the presence of zinc. Hydrometallurgy, 73: 339-341.
- [7] Safarzadeh, M.S., D. Moradkhani, M. Ojaghi-Ilkhchi, 2007b. Determination of the optimum conditions for the cementation of cadmium with zinc powder in sulfate medium. Chem. Eng. Process, 46: 1332-1340.
- [8] Chizhikov, D.M., 1966. Cadmium (translated by D.E. Hayler, 1st English ed.). Pergamon Press.
- [9] D'yachenko, A.N., R.I. Kraidenko, 2010. Processing oxide-sulfide copper ores using ammonium chloride. Russ. J. Non-Ferrous Met., 51: 377-381.
- [10] Hamamcı, C., B. Ziyadanoğulları, 1991. Effect of roasting with ammonium sulfate and sulfuric acid on the extraction of copper and cobalt from copper converter slag. Sep. Sci. Technol, 26: 1147-1154.
- [11] Moradkhani, D., M. Rasouli, D. Behnian, H. Arjmandfar, P. Ashtari, 2012. Selective zinc alkaline leaching optimization and cadmium sponge recovery by electrowinning from cold filter cake (CFC) residue. Hydrometallurgy, 115-116: 84-92.