Hydrometallurgy 177 (2018) 34–40

Contents lists available at ScienceDirect

Hydrometallurgy

journal homepage: www.elsevier.com/locate/hydromet

Integrated diffusion dialysis precipitation – Cementation for selective recovery of leaching chemicals and metal values from electroplating sludge

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ARTICLE INFO

Keywords:
Acid dialysis
Recovery of sulfuric acid
Metal recovery
Electroplating sludge

ABSTRACT

The hydrometallurgical valorisation of electroplating metal hydroxides sludge was investigated here. It proposes a complete solution. The hazardous solid waste is both an environmental and an economic global issue. It is considered as a challenging scientific topic. This work aims to recover selectively the leaching acid excess and metal values.

Fast and complete sludge dissolution is a necessary first step that needs a large excess of concentrated leaching chemical (acid, base, complex…). This constitutes a serious drawback of hydrometallurgical processes because this excess is generally lost and constitutes a supplementary pollutant.

The combination chemical leaching – diffusion dialysis (DD) – cementation – precipitation was the main hypothesis of this research.

The acid excess was recovered and reused by DD. This constitutes this research novelty. Finally the DD de acidified retentate, loaded with dissolved metals, was successfully processed by various separation operations: selective precipitation and cementation. DD is a well established membrane process for pickling acid recovery, however this sludge specific and efficient application is quite novel.

Promising results have been obtained; more than 95% of the acid excess was recovered. Copper was successfully cemented (100%). Selective metals precipitation by phosphate was the final recovery step. The studied problem dealt with an important Algerian tap manufacturing plant.

It shows very excellent results (100% of zinc, 99% of nickel, 98, 11% for Lead, 84, 6% Iron and 78, 46% of chromium).

1. Introduction

The metals electroplating remains a dynamic and growing activity and a corner stone for the modern industry. It is directly related to the massive production industries (cars, military, refrigerators, electronic and electric devices, automotives…) where the liquid and solid waste effluents volumes are also enormous. The conventional and most used wastewater treatment of this industry is based on a final precipitation of cationic heavy metals M^{n+} by OH^-, after a CN^- and Cr(VI) detoxification (Crini and Badot, 2007),

$$M^{n+} + nOH \rightarrow M(OH)_n\downarrow$$

This precipitate is called the electroplating sludge, a complex mixture of various metals hydroxides that are considered a hazardous solid waste and a serious environmental threat resulting from this ubiquitous industry.

The various types of pollutants and the enormous quantity of metals generated from this particular industry pose a big problem concerning waste management. In Europe sludge is either recycled or stored in controlled landfills even if they are decreasingly used. Stabilization/Solidification process were used few years ago (Silva et al., 2007; Malviya and Chaudhary, 2006; Li et al., 2001; Poon et al., 2001).

In recent years, several researchers have been interested in the treatment of electroplating sludge; (Pham T. Huyen et al., 2016), applied the combination of acid leaching and electrodeposition to recover copper from a surface treatment sludge. (Xu et al., 2015), were able to recover 93.6% and 88.9% of copper and nickel successively by extraction with N902 in kerosene followed by selective pickling with sulfuric acid. (Sua et al., 2016), have studied the effects of electroplating in hydrochloric acid medium, the rate of success is as high as 80.6% under optimal conditions. Some researchers have even applied methods that rely on the effect of bacteria on metals (Prabhu and Baskar, 2015).

Recently, they are working to find radical solutions to the problem using a technology called “Clean technology “to recover metals from sludge baths. There is three procedures can be used to treat the already

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https://doi.org/10.1016/j.hydromet.2018.02.011
Received 19 October 2017; Received in revised form 22 December 2017; Accepted 17 February 2018
Available online 24 February 2018

existed sludge: hydrometallurgical procedure, pyrometallurgical procedure and finally the combination of both procedures (Amaral et al., 2014), and a lot of other procedures are used to recover metals directly from sludge baths, mentioning for example: selective precipitation (Wang and Zhou, 2002; Wang et al., 2013), membranes techniques (Matis et al., 2005; Fu and Wang, 2011; Zou et al., 2009; Bouhidel and Rumeau, 2000), Ion exchange resin (Franco et al., 2013).

Algeria was one of the most industrialized countries in Africa; this is partly illustrated by the large Arcelor Mittal – Sider steel making plant (2 M t/y) in El Hadjar (Annaba) and by numerous derived metal plating based industries (various galvanizations, taps, bicycles, automotive, refrigerators …manufacturing). Mostly realized during the seventies and eighties these plants were equipped with conventional wastewaters treatment stations based on CN- and Cr(VI) detoxification, ion exchange metals fixation – regeneration and finally the metal hydroxides precipitation with the sludge generation; this situation has not changed until today increasing the sludge stock. The total absence of specialized landfills and realistic heavy metals pollution prevention politics makes the environmental situation more and more hazardous. This Algerian plating sludge problem, topic of this work, was poorly studied in the specialized literature.

This research work deal with the electroplating and metal finishing sludge valorization by a hydrometallurgical process. This aimed leaching totally heavy metals for further recovery and isolating detoxified sludge for safe land filling. In order to do that the combination sludge leaching – diffusion dialysis (leaching agent excess recovery) - heavy metals selective recovery were performed.

The lixiviation first step requires a large leaching chemicals (acids (Miskufova et al., 2006, Tu et al., 2010), bases(Feng et al., 2007), ammonia (Salhi and Bouhidel, 2005), oxidant (Selby and Twidwell, 2006), …) excess for fast and complete sludge dissolution. This excess constitutes the main drawback (loss, pollution…) of the hydrometallurgical process. Its recovery and reuse has never been studied before. Besides the metals analysis and their selective separation objective, this research work aims also to recover the leaching excess acid by diffusion dialysis (DD), a quite novel application.

Acids excess recovery by anion exchange membranes diffusion dialysis is an elegant, efficient and well-established process. It is based on the H+ protons membrane leakage. Its main strength is the separation acid– metallic salt. It has been deeply investigated and largely applied to acidic pickling bathes. The recovery of nitric (Chahrazad Amrane et al., 2017), sulfuric (Xu et al., 2009; Li et al., 2012), phosphoric (Kim et al., 2012), hydrochloric (Luo et al., 2010; Palaty´ and Bendova, 2009), mineral acids from stainless steel, brass and iron pickling bathes have been successfully investigated.

In addition to this scientific objective, this work aimed to contribute solving the Algerian EP sludge problem by showing its feasibility, raising awareness and convincing EP and environmental decision makers.

2. Materials and methods

2.1. Materials

The experimental device consists of a two-compartment Plexiglas cell separated by an anion exchange membrane NEOSEPTA AMX manufactured by the Japanese company TUKUYAMA SODA Co, Ldt. Its basic properties were as follows: Thickness 0.12–0.18 mm, Ion-exchange capacity 1.4–1.7 meq / g, Water content 25–30%. The active surface area is 1 cm^2. The temperature is 30°.

A pH - meter is used: PHM240 pH/ION METER model. MetreLab to monitor the pH evolution of the recovered solution as a function of time and a UV–Visible spectrophotometer: “SHIMADZU UV-1700” model to track the amount of metal passing through the membrane and the recovery efficiencies of each metal (Figs. 1, 2, 7 and 8).

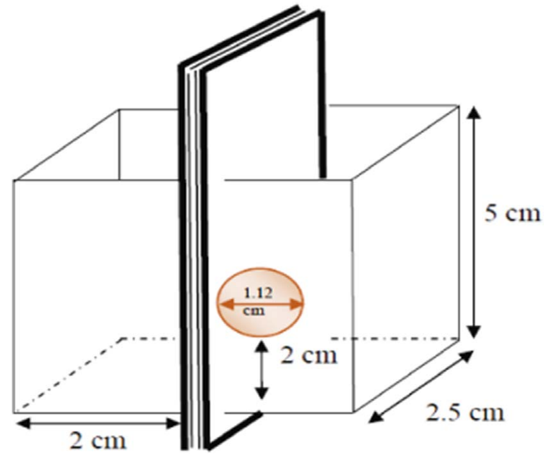

Fig. 1. Experimental device of dialysis.

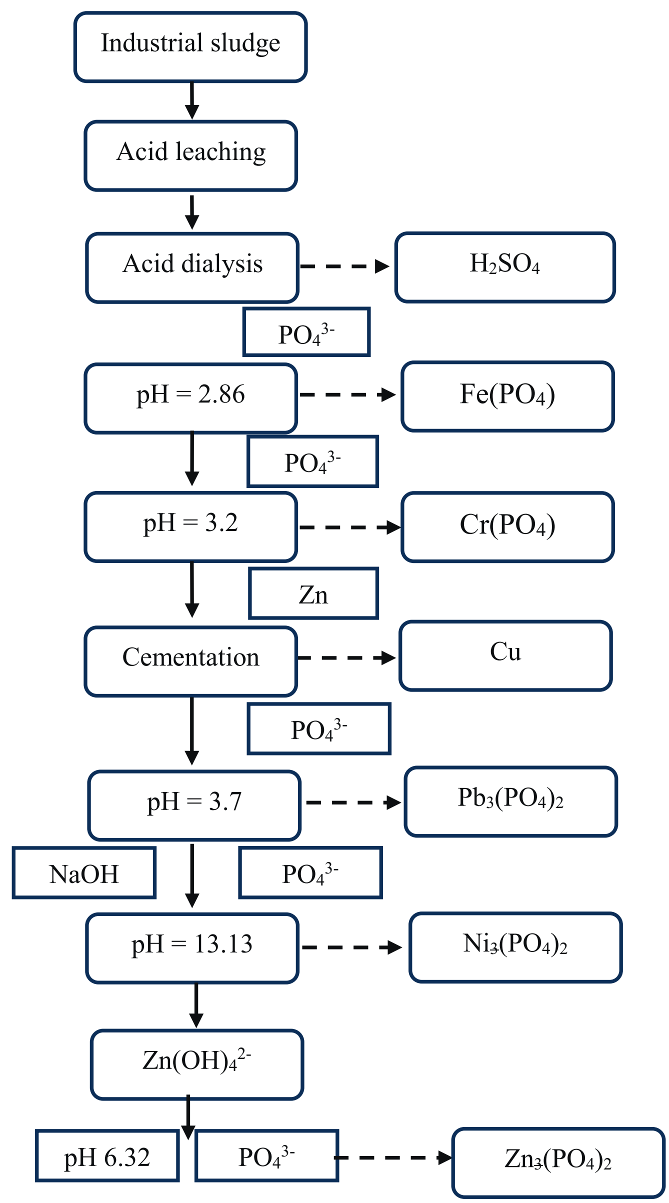

Fig. 2. Procedure of the separation of metals from industrial sludge.